Environmental Impacts of COAL MINING AND UTILIZATION

(A complete revision of Environmental Implications of
Expanded Coal Utilization)

A study by The Beijer Institute
The United Nations Environment Programme
The USSR Academy of Sciences
The National Energy Administration, Sweden

Editorial Writers:

M. J. CHADWICK

N. H. HIGHTON

and

N. LINDMAN

Published for the Beijer Institute,
UNEP, the USSR Academy of Sciences
and the National Energy Administration, Sweden

PERGAMON PRESS

OXFORD · NEW YORK · BEIJING · FRANKFURT
SÃO PAULO · SYDNEY · TOKYO · TORONTO

U.K.	Pergamon Press, Headington Hill Hall, Oxford OX3 0BW, England
U.S.A.	Pergamon Press, Maxwell House, Fairview Park, Elmsford, New York 10523, U.S.A.
PEOPLE'S REPUBLIC OF CHINA	Pergamon Press, Qianmen Hotel, Beijing, People's Republic of China
FEDERAL REPUBLIC OF GERMANY	Pergamon Press, Hammerweg 6, D-6242 Kronberg, Federal Republic of Germany
BRAZIL	Pergamon Editora, Rua Eça de Queiros, 346, CEP 04011, São Paulo, Brazil
AUSTRALIA	Pergamon Press Australia, P.O. Box 544, Potts Point, N.S.W. 2011, Australia
JAPAN	Pergamon Press, 8th Floor, Matsuoka Central Building, 1-7-1 Nishishinjuku, Shinjuku-ku, Tokyo 160, Japan
CANADA	Pergamon Press Canada, Suite 104, 150 Consumers Road, Willowdale, Ontario M2J 1P9, Canada

First edition 1987

Library of Congress Cataloging in Publication Data

Environmental impacts of coal mining and utilization.
"A complete revision of: Environmental implications
of expanded coal utilization."
"Published for the Beijer Institute, UNEP, and the
USSR Academy of Sciences."
Includes bibliographies.
1. Coal—Environmental aspects. I. Chadwick, M. J.
(Michael J.) II. Lindman, Nils. III. Highton, N. H.
IV. Beijer Institute. V. United Nations Environment
Programme. VI. Akademiia Nauk SSSR. VII. Environmental
implications of expanded coal utilization.
TD195.C58E5927 1986 333.8'221 86-9464

British Library Cataloguing in Publication Data

Environmental impacts of coal mining and
utilization: a study by the Beijer
Institute, the United Nations Environmental
Programme, the USSR Academy of Sciences.
—Rev.
1. Coal—Environmental aspects
I. Chadwick, M. J. II. Lindman, Nils
III. Highton, N. H. IV. Beijerinstitutet
V. United Nations. *Environmental Programme*
VI. Akademiia nauk SSSR VII. Environmental
implications of expanded coal utilization
662.6'2 TD195.C58

ISBN 0-08-031427-9

Printed in Great Britain by A. Wheaton & Co. Ltd. Exeter

Foreword

In 1979, when the Beijer Institute, the United Nations Environment Programme (UNEP) and the USSR Academy of Sciences embarked upon a joint study of the environmental implications of increasing coal use, the world economy was preoccupied with the pervasive impacts of the rising price of oil. At that time, many countries worldwide were re-examining their energy supply options and a number of them came to believe that coal would become the natural substitute for increasingly expensive oil. Much attention was being focused on modern technologies for the preparation of gaseous and especially of liquid fuels from coal in an attempt to emulate the versatility of oil and overcome the last fundamental barrier to its substitution. All these concerns were reflected in the book that emerged from this joint study—*Environmental Implications of Expanded Coal Utilization*, published by Pergamon Press in 1982.

During that period, predictions and forecasts about the future price of oil and, hence, its destiny in the global energy economy were everywhere abundant. But the idea of drastically falling prices as oil suppliers competed for a share in a contracting world market seemed hardly credible when it was mooted at the time. The prevailing mood was much more one of concern as to how the world coal industry could be mobilized to meet the production levels that would most likely be required of it. Now, 7 years on, with oil selling for less than half its 1980 price, forecasters have a much greater respect for uncertainty and realize that arriving at any energy future involving oil will almost certainly produce a very bumpy ride.

Questions about the long-term global availability of oil, political volatility and war in the Middle East, the falling demand for oil as energy conservation measures and economic recession began to reduce world consumption, have all contributed to this overall uncertainty. But the important consequence for coal is that, in the short term, they have led to a steady price decline in oil and seriously reduced the urgency and motivation of pushing ahead with wholesale substitution by coal. This does not mean that coal has no important role to play in the future. It is simply that its true role is currently turning out to be different from what was anticipated for it. Only the most robust developments in coal expansion have survived. These have included the rapid deployment of hitherto largely neglected coal resources in several developing countries, especially in South America and Africa. Additionally, several industrialized countries have pushed ahead rapidly with extensive technological advances in coal combustion methods. By contrast, coal gasification and, in particular, liquefaction operations have been scaled down or abandoned. In fact, only a very few really promising advanced projects have survived in the new economic climate of cheaper oil.

All these considerations have led us to request the authors of *Environmental Implications of Expanded Coal Utilization* to undertake the task of revising the original text in a most fundamental and extensive manner. The whole energy demand and supply scene needed a completely fresh appraisal. New aspects of the resource requirements for coal mine development had to be dealt with. The very interesting new technological advances in coal combustion also required detailed treatment. In addition, the enormous level of activity in the field of acid depositions and their effects on the environment demanded a new assessment of its significance for our understanding of environmental acidification regionally.

We have been joined in these endeavours by the National Energy Administration of Sweden which for some years has been deeply involved in studying the potential role of coal in Sweden.

The result of this fundamental revision is not a new edition of the old text but rather a completely new volume. The actual work of reappraisal and revision had its origins in an International Workshop held by us in Moscow in January 1984, hosted by our Soviet colleagues. Many scientists participated in the new joint study by contributing to the review procedure at the Workshop and/or by detailed technical correspondence. We should like to thank all those who have given their time and effort to this work.

Our especial thanks go to Professor M. J. Chadwick, Dr. N. H. Highton and Dr. N. Lindman, who not only edited all the newly collected material but also provided a substantial proportion of it themselves.

Finally, one thing is clear from the new joint study. Although it seems that the precise role to be played by coal in the global energy economy will always depend on the prevailing fluctuations in the price of oil, there is no doubt that over the ensuing years, with new ways of exploiting it, coal will without any doubt steadily increase in importance as a major world fuel.

For the Beijer Institute
Royal Swedish Academy of Sciences:

GORDON T. GOODMAN
Executive Director, Beijer Institute

For the UN Environment
Programme:

M. TOLBA
Executive Director

For the USSR Academy
of Sciences:

SVENELD EVTEEV
Deputy Chairman
Earth Sciences Section

For the National Energy
Administration of Sweden:

H. RODE
Director

March 1986

Preface

The environmental dimension to energy use is now firmly accepted in energy planning and development. When changes in the spectrum of energy use are proposed, comparison of the environmental impacts is often a significant element in the considerations involved. Public opinion is now more clearly focused and influential in determining the outcome of changes in the balance of fuel use in the economy of many countries, particularly in relation to the environmental implications of such changes.

From 1979 to 1981, the second round of oil price increases led to suggestions that coal would be selected as a major energy alternative. It was widely predicted that, as well as an increase in coal combustion using existing technologies and rapidly developing ones, coal would also feature as a source of liquid and gaseous fuels. Environmental concern focused on the likely impacts both of increased coal mining and transportation and on the outcome of gasification and liquefaction technologies. One of the results of these concerns was the organization of an International Workshop on the *Environmental Implications and Strategies for Expanded Coal Utilization* held in Moscow in 1980. The discussions at the Workshop, the material subsequently submitted and a number of new publications formed the basis of a volume (Chadwick and Lindman, 1982) addressing the major environmental concerns of increased coal utilization. However, subsequent downward trends in oil prices gave rise to less optimism on the future of the international coal market: export prices were depressed, demand did not develop as previously foreseen and excessive production resulted in increased stocks around the world. But surprisingly these factors did not result in quite the downturn in coal utilization that might have been expected: oil-pricing uncertainties and possible future supply difficulties due to the Iraqi–Iranian war, and political volatility combined to ensure a steady substitution of coal for oil and large-scale mining developments (in Colombia, for example) coming on stream to meet this growing demand.

For the reasons outlined above it was decided to update the information on the environmental implications of increased coal production and utilization contained in the previous work (Chadwick and Lindman, 1982). The focus of this new study was rather different from the original activity, however; more emphasis was given to developments in coal combustion technology and there was a consequent reduction in the treatment given to coal gasification and liquefaction. In addition, it was evident that more consideration was being given, worldwide, to the resource requirements and demands made by the development of new coal mines. Further, the increase in the number of investigations of the effects of emissions resulting from coal combustion had to be reflected in the account of acid deposition effects on man-made and natural ecosystems. The result is a new volume rather than a second edition of the original one.

Chapter 1 attempts a fresh assessment of the role of coal in the international energy scene. This involves a consideration of coal reserves, the relationship between energy use and economic growth, factors controlling energy demand and supply, and how current coal trade patterns might develop.

Chapters 2 and 3 address the impacts associated with coal mining developments: land disturbance and reinstatement as well as the resource demands involved. In particular, water needs are considered, as there is some reason to believe that this may limit production in some regions of the world. Chapter 4 deals with the environmental impacts of coal transportation. Chapter 5 considers the developments taking place in coal combustion technology, particularly fluidized bed combustion, coal liquid mixture combustion and the implementation of combined cycles. The effect of these developments on effluent production and emission control is considered. Attention is paid to coal conversion processes in Chapter 6.

Chapter 7 outlines the concepts involved in assessing environmental and health impacts of coal utilization and conversion and this forms an introduction to the treatment of the environmental significance of exposure to trace elements emitted from coal utilization processes (Chapter 8) and carbon compounds (Chapter 9). Chapter 10 looks at the overall occupational and environmental health problems from coal utilization and conversion in an integrated way.

The pollutants traditionally associated with coal combustion are sulphur dioxide and smoke. However, since particulate abatement has been so successfully implemented, interest has focused, in a major way, on sulphur dioxide and other acidic substance emissions. The result has been rapid progress in the study of acid deposition effects. Chapter 11 reviews this in relation to natural and man-managed terrestrial and freshwater ecosystems.

In spite of recent fuel market trends it seems safe to assume that moderate economic growth will occur and that the price of oil will eventually continue on its upward path. Coal demand may not attain some of the previous more optimistic predictions, but will increase, at least in a modest way, for the 20 years beyond 1990. This increased utilization need not take place to the detriment of the environment or at the expense of public and occupational health. This volume sets the scene for the examination of the major factors that need continued attention to make sure that this is so.

February 1986 M. J. CHADWICK N. H. HIGHTON NILS LINDMAN

CHADWICK, M. J. and LINDMAN, N. (1982). *Environmental Implications of Expanded Coal Utilization*. Pergamon, Oxford.

Acknowledgments

Four organizations have sponsored this study. The Beijer Institute, Stockholm, commissioned written reviews on the main areas where recent advances have occurred in relation to coal utilization and environmental impacts. The USSR Academy of Sciences co-ordinated contributions from the Soviet Union. They also arranged (through the State Committee for Science and Technology and the USSR Commission for UNEP) for a Workshop to be held in Moscow in January 1984. The United Nations Environment Programme, together with the Beijer Institute and the USSR Commission for UNEP, gave financial support to enable this Workshop to take place and the Energy Research Commission, Stockholm gave financial assistance with attendance at the Workshop. The National Energy Administration of Sweden generously provided funds to help with editorial work and the writing of new sections of the study after the Workshop. This support has enabled the material for this book to be assembled in three ways: from the commissioned review papers; discussion, comments and written material at the Workshop; reference material and new written material prepared after the Workshop in the light of published material and advances up to December 1985.

The review papers and other written material were prepared by J. N. B. Bell, I. S. Blagov, M. J. Chadwick, A. Grübler, L. D. Hamilton, N. H. Highton, H. Jüntgen, J. Klein, V. A. Knizhnikov, A. A. Krichko, N. Lindman, V. M. Maslennikov, A. Y. Reteyum, V. A. Ruban, M. Y. Shpirt, M. A. Styrikovich, P. J. Svennignsson and R. A. Wadden. A discussion document based on this material was commented upon, in detail, by twenty-two participants at the Workshop from the USSR, the USA, Austria, the People's Republic of China, Colombia, the Federal Republic of Germany, Sweden and the United Kingdom. A paper published by IIASA and Resources and Energy (Elsevier Science Publishers), prepared by J. Alcamo, was also discussed.

Many of the participants, and others, submitted written material at the Workshop and subsequently by post. All these sources have formed the material for this book, together with sections from the previous volume, *Environmental Implications of Expanded Coal Utilization* by M. J. Chadwick and Nils Lindman (Pergamon Press, 1982). The contribution of authors of review papers for this earlier volume is acknowledged here. This new work is, however, so substantially rewritten, and its emphasis changed, that it must be regarded as a new work rather than a second edition.

We gratefully acknowledge the support of the Beijer Institute and its Director (Professor G. T. Goodman) and Deputy Director (Dr. Lars

Kristoferson). We are also grateful for the help rendered by the USSR Academy of Sciences (particularly Academician A. V. Fokin, V. Volgin and Mrs. N. Galubova), the USSR Commission for UNEP, UNEP, Nairobi (Dr. Y. el Mahgary) and the National Energy Administration of Sweden (H. Rode).

The cover illustration is reproduced by kind permission of the NCB Yorkshire Regional Photographic Department (NCB copyright reserved). A substantial part of Chapter 3 appeared as an article by J. Alcamo, *Fire and Water: Water Needs of Future Coal Development in the Soviet Union and the United States*, published by IIASA and Elsevier Science Publishers. We are most grateful for permission to reproduce it. We also make grateful acknowledgment for the editorial assistance received from Isobel Devan, Julian Dison, Solveig Nilsson and Susan Sparrow.

February 1986

M. J. CHADWICK
N. H. HIGHTON
NILS LINDMAN

Abbreviations and Acronyms

(see also Units, Conversions and Equivalents)

AF	Ash free
Ap	Ash content (proportion)
ASTM	American Society for Testing and Materials
BOD	Biochemical oxygen demand
BP	Boiling point
CAPTEX	Cross Appalachian Tracer Experiment
CARBOCOL	Colombia State Mining Corporation
CC	Combined cycle
CFB	Circulating fluidized bed
CHO	Chinese hamster ovary fibroblasts
CIF	Cost insurance and freight
CLM	Coal, liquid mixture
CMEA[1]	Council for Mutual Economic Aid
CMM	Coal, methanol mixture
CMW	Coal, methanol, water mixture
CoGas	Coal gas process
COM	Coal, oil mixture
COW	Coal, oil, water mixture
CPE	Centrally planned economies
CSSR	Czechoslovakian Soviet Socialist Republic
C(t)	Concentration at time *t*
CT	Concentration × time
CWM	Coal, water mixture
daf	Dry, ash free
$	Dollar (US)
DOE	Department of Energy (US)
dwt	Dead weight tonnage
E	Exposure
EC_{50}	Effective concentration at which 50% die
EDS	EXXON donor solvent process
EEC[2]	European Economic Community (Commission of the European Communities)
EF	Enrichment factor
EMEP	Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollution in Europe

EPA	Environmental Protection Agency (US)
ER	Enrichment ratio
F	Flux
FDA	Food and Drug Administration (US)
FGD	Flue gas desulphurization
FOB	Free on board
FRG	Federal Republic of Germany
F-T	Fischer-Tropsch process
GDR	German Democratic Republic
GNP	Gross national product
H-coal	Hydrogen Research Company hydrogenation process
HPLC	High pressure liquid chromatography
I	Intake
ID	Internal diameter
IEA[3]	International Energy Agency
IGC	International Geological Congress
IIASA	International Institute for Applied Systems Analysis
IUFRO	International Union of Forest Research Organizations
K	Proportionality constant
LC_{50}	Lethal concentration for 50% of a test population
LD_{50}	Lethal dose for 50% of a test population
LNTM	Linear no-threshold model
MAC	Maximum allowable concentration
MCG	Medium calorific gas
MEGs	Multi-media environmental goals
MHD	Magnetohydrodynamic
NCB	National Coal Board (UK)
OECD[4]	Organization for Economic Co-operation and Development
OPEC	Organization of Petroleum Exporting Countries
ORNL	Oak Ridge National Laboratory
OTA	Office of Technology Assessment
PBL	Planetary boundary layer
PdC	Pas de Calais
PFA	Pulverized fuel ash
PFBC	Pressurized fluidized bed combustion
pH	Negative logarithm of hydrogen ion concentration
POM	Polycyclic organic matter
PWR	Pressurized water reactor
Q	Amount of pollutant
Qp	Proportionality of high-rate volatiles to proximate volatiles
RAM	Rabbit alveolar macrophages
RSA	Republic of South Africa

RSFSR	Russian Soviet Federated Socialist Republic
SAM	Source analysis model
SANT-H	A coal tar oil
SAR	Synthesegasanlage Ruhr
SASOL	South African Coal, Oil and Gas Corporation
SCOPE	Scientific Committee on Problems of the Environment
SCR	Japanese ammonia injection system for removal of NO_x
SEM	Scanning electron microscopy
SFC	Synthetic Fuels Corporation
SNG	Natural gas substitute
SQI	Solvent quality index
SRC	Solvent refined coal
T	Temperature
tce	tonnes of coal equivalent
TDS	Total dissolved solids
TL	Tolerance limit
TLC	Thin layer chromatography
TLV	Threshold limit value
TOC	Trace organic compounds
toe	tonnes of oil equivalent
TSS	Total suspended solids
TVA	Tennessee Valley Authority
UNESCO	United Nations Educational, Scientific and Cultural Organization
UNGA	United Nations General Assembly
UNSCEAR	United Nations Scientific Committee of the Effects of Atomic Radiation
USA	United States of America
USBM	United States Bureau of Mines
USGS	United States Geological Survey
USSR	Union of Soviet Socialist Republics
WELMM	Water, energy, land, manpower and materials
Wp	Water content (proportion)
X	Concentration of an element

[1] *Council for Mutual Economic Aid*: Bulgaria, Czechoslovakia, German Democratic Republic, Hungary, Poland, Romania and the Union of Soviet Socialist Republics.

[2] *European Economic Community (Commission of the European Communities)*: Belgium, Denmark, Federal Republic of Germany, France, Greece, Ireland, Italy, Luxembourg, the Netherlands, Portugal, Spain and the United Kingdom.

[3] *International Energy Agency*: Australia, Austria, Belgium, Canada, Denmark, Federal Republic of Germany, Greece, Ireland, Italy, Japan,

Luxembourg, the Netherlands, New Zealand, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States of America.

[4] *Organization for Economic Co-operation and Development*: Australia, Austria, Belgium, Canada, Denmark, Federal Republic of Germany, Finland, France, Greece, Iceland, Ireland, Italy, Japan, Luxembourg, the Netherlands, New Zealand, Norway, Portugal, Spain, Sweden, Switzerland, Turkey, the United Kingdom and the United States of America.

Units, Conversions and Equivalents

(Mainly the Système International d'Unités (SI) units have been used throughout the book except where specific use in the original work quoted makes this inappropriate: equivalents and conversion factors are therefore given)

Prefixes for SI Units

Factor	Prefix	Symbol
10^{18}	exa	E
10^{15}	peta	P
10^{12}	tera	T
10^{9}	giga	G
10^{6}	mega	M
10^{3}	kilo	k
10	deca	da
10^{-1}	deci	d
10^{-2}	centi	c
10^{-3}	milli	m
10^{-6}	micro	μ
10^{-9}	nano	n
10^{-12}	pico	p
10^{-15}	femto	f
10^{-18}	atto	a

SI Units

Physical quantity	Unit	Abbreviation	Definition (or equivalent)
length	metre	m	–
mass	kilogram	kg	–
time	second	s	–
thermodynamic temperature	kelvin	K	–
current	ampere	A	–

SI Units

Physical quantity	Unit	Abbreviation	Definition (or equivalent)
amount of substance	mole	mol	–
energy	joule	J	$kg\ m^2\ s^{-2}$
force	newton	N	$kg\ m\ s^{-2}$
power	watt	W	$kg\ m^2\ s^{-3}$
pressure	pascal	Pa	$kg\ m^{-1}\ s^{-2}$
electrical potential difference	volt	V	$J\ A^{-1}\ s^{-1}$
electrical conductance	siemen	S	$A\ V^{-1}$
activity	becquerel	Bq	$(l\ s)^{-1}$
absorbed dose	gray	Gy	$J\ kg^{-1}$
dose equivalent	sievert	SV	$J\ kg^{-1}\ Q$

(Q is the "quality factor" which is unity for β rays, X-rays and γ rays)

Other Units and Equivalents

length	foot	f	0.3048 m
	inch	i	0.0254 m
	mile	(m)	1.609×10^3 m
area	hectare	ha	$10^4\ m^2$
	acre	(acr)	0.4047 ha
	square inch	(si)	$6.45 \times 10^{-4}\ m^2$
volume	normal cubic metre	Nm^3	m^3
	litre	1	$1\ m^3 \times 10^{-3}$
	standard cubic metre	scm	
	standard cubic foot	scf	$2.832\ m^3 \times 10^{-2}$
	gallon	gall	4.54596 1
mass	gram	g	$1\ kg \times 10^{-3}$
	pound	lb	0.4536 kg
	tonne	t	$kg \times 10^3$
	ton (US)	(t)	0.9072 t
time	minute	min	60 s
	hour	h	3600 s
	day	d	24 h
	year	yr	8760 h

Other Units and Equivalents

temperature	degrees Celsius	°C	K + 273.15
	degrees Fahrenheit	°F	1.8C + 32
amount of substance	equivalent	eq	mol valency^{-1}
concentration	parts per million	ppm	mg kg^{-1}
	parts per million (volume basis)	ppmv	μl l^{-1}
	parts per billion	ppb	μg kg^{-1}
pressure	atmosphere	atm	1.013×10^5 N m^{-2}
	bar	bar	10^5 N m^{-2}
	pounds per square inch absolute	psia	6.9×10^3 N m^{-2}
energy	calorie	cal	4.184 J
	British thermal unit	BTU (Btu)	1055 J
	therm	—	1.005 J $\times 10^8$
	thermie	th	0.03968 therm
	tonnes coal equivalent		2.88×10^{10} J
	million tonnes coal equivalent	mtce	2.88×10^{16} J
	tonnes oil equivalent	toe	4.19×10^{10} J
	million tonnes oil equivalent	mtoe	4.19×10^{16} J
	barrel	bbl	5.74×10^9 J
power	megawatt (electric)	MW(e)	10^6 J s^{-1}
	megawatt (thermal)	MW(th)	
	gigawatt (electric)	GW(e)	10^9 J s^{-1}
	kilowatt hour	kWh	3.6×10^6 J
	gigawatt (electric) a year	GW(e)yr	10^9 J s^{-1} 3.15×10^7
radioactivity	curie	Ci	3.7×10^{10} Bq
	rad	rad	0.01 Gy
	rem	rem	0.01 Sv
sound	decibel	dB	comparative unit of intensity of sound

Conversions

	kg	t	(ton US)	lb
kilogram (g)	1	0.001	0.000984	2.20462
tonne (t)	1000	1	1.12	2204.6
ton (US)	907.19	0.9072	1	2000
pound (lb)	0.4536	4.536×10^{-4}	0.0005	1

	10^6 Btu	10^9 cal	toe	tce	GJ
tonne coal equivalent (tce)	27.337	6.88	0.6370	1	28.84
tonne oil equivalent (toe)	40.185	10.128	1	1.57	42.395

Elements and Chemical Compounds

Symbol	Element
A	Argon
Al	Aluminium
Ag	Silver
As	Arsenic
As_4	Arsenic (crystalline or gaseous)
As_2	Arsenic forming at high temperatures: $As_4 \leftrightarrows 2As_2$
Au	Gold
B	Boron
Ba	Barium
Be	Beryllium
Br	Bromine
C	Carbon
Ca	Calcium
Cd	Cadmium
Ce	Cerium
Co	Cobalt
Cr	Chromium
Cs	Cesium
Cu	Copper
Eu	Europium
F	Fluorine
Fe	Iron
Ga	Gallium
Gd	Gadolinium
Ge	Germanium
H, H_2	Hydrogen
Hf	Hafnium
Hg	Mercury
I	Iodine
In	Indium
K	Potassium
Kr	Krypton
La	Lanthanum
Mg	Magnesium
Mn	Manganese

Mo	Molybdenum
N, N_2	Nitrogen
Na	Sodium
Ni	Nickel
O, O_2	Oxygen
P	Phosphorus
Pb	Lead
Po	Polonium
Ra	Radium
Rb	Rubidium
Ru	Ruthenium
S	Sulphur
Sb	Antimony
Sc	Scandium
Se	Selenium
Si	Silicon
Sm	Samarium
Sn	Tin
Sr	Strontium
Ta	Tantalum
Tb	Terbium
Th	Thorium
Ti	Titanium
Tl	Thallium
U	Uranium
V	Vanadium
Xe	Xenon
Zn	Zinc
Zr	Zirconium

Symbol	Chemical compound
Al_2O_3	Aluminium oxide
AsH_3	Arsenic trihydride
BaA	Benz(*a*)anthracene (1,2-benzanthracene)
BkF	Benzo(*k*)fluoranthene (8,9-benzfluoranthene)
$B(OH)_3$	Orthoboric acid
BaP	Benzo(*a*)pyrene (3,4-benzpyrene)
BTX-aromatics	Benzene, toluene, xylene aromatics
CH_3SH	Methyl mercaptan
CH_3OH	Methanol
CH_4	Methane
C_2H_2	Ethene (acetylene)
C_2H_4	Ethylene

Symbol	Chemical compound
C_2H_5SH	Ethyl mercaptan
C_2H_6	Ethane
CN^-	Cyanide
CO	Carbon monoxide
CO_2	Carbon dioxide
COS	Carbonyl oxysulphide
CS	Carbon monosulphide (unstable gas)
CS_2	Carbon disulphide
$CaCO_3$	Calcium carbonate (limestone)
CaC_2	Calcium carbide
$CaMoO_4$	Calcium molybdate
CaO	Calcium oxide
$Ca(OH)_2$	Calcium hydroxide
CaS	Calcium sulphide
$CaSO_4$	Calcium sulphate (gypsum)
DBahA	Dibenz(*a*,*h*)anthracene (1,2–5,6-dibenzanthracene)
DBahP	Dibenzo(*a*,*h*)pyrene (3,4–8,9-dibenzpyrene)
DMBA	7,12-Dimethylbenz(*a*)anthracene (9,10-dimethyl-1,2-benzanthracene)
DNA	Deoxyribose nucleic acid
$Fe(CO)_5$	Iron carbonyl
FeO	Ferrous oxide
Fe_2O_3	Ferric oxide
FeS	Ferrous sulphide
FeS_2	Iron pyrites
$Fe_2(SO_4)_3$	Ferric sulphate
HCl	Hydrochloric acid
HCN	Hydrogen cyanide (gas)
HCO_3^-	Bicarbonate ion
HF	Hydrogen fluoride
HNO_3	Nitric acid
H_2O	Water
HS	Monohydrogen sulphide
H_2S	Hydrogen sulphide (gas)
H_2SO_4	Sulphuric acid
H_2Se	Hydrogen selenide
KOH	Potassium hydroxide
K_2CO_3	Potassium carbonate
K_2S	Potassium monosulphide
K_2SO_4	Potassium sulphate
$MgMoO_4$	Magnesium molybdate

MgO	Magnesium oxide
$MgCO_3$	Magnesium carbonate
MoO_3	Molybdenum trioxide
MCA	3-Methylcholanthrene (3-methyl-1,2-dihydrobenz(*j*)aceanthrylene)
MTBE	Methyl-t-butyl ether
$Ni(CO)_4$	Nickel carbonyl
NH	Amine group
NH_2	Amide group
NH_3	Ammonia (gas)
$NH_4{}^+$	Ammonium ion
$(NH_4)_2SO_4$	Ammonium sulphate
NO_2	Nitrogen dioxide
$NO_3{}^-$	Nitrate ion
NO_X	Nitrogen oxides (usually mixed gases)
N_2O	Nitrous oxide
Na_2S	Sodium sulphide
Na_2SO_4	Sodium sulphate
OH^-	Hydroxyl ion
O_3	Ozone (gas)
$PbCl_2$	Lead chloride
PbO	Lead monoxide
PbS	Lead sulphide
$PO_4{}^-$	Phosphate ion
PAH	Polycyclic aromatic hydrocarbons
PAN	Peroxyacetylnitrate
PNA	Polynuclear aromatics
PVA	Polyvinyl acetate
SO	Sulphur monoxide
SO_2	Sulphur dioxide (gas)
SO_3	Sulphur trioxide
$SO_4{}^-$	Sulphate ion
S_2O	Sulphurous oxide
SiO_2	Silicon dioxide (silica)
UO_2	Uranite

1

Coal in the International Energy Scene

1. Introduction 1
2. Resources and Reserves 3
3. Energy Use and Economic Growth 8
4. Coal Demand 11
    4.1. Energy Prices and Interfuel Substitution 11
    4.2. Coal and Electricity Generation 14
    4.3. Other Industrial Markets 16
    4.4. Coal Conversion 16
5. Coal Supply 19
    5.1. Production and Costs 19
    5.2. Developing Regions 22
    5.3. Transportation 23
6. Coal Trade 23
    6.1. Current Trade Requirements 23
    6.2. Outlook 25
7. Institutional Developments 27
8. References 27

1. Introduction

The utilization of coal as an energy source, both by means of the steam engine and in the form of coke, was a major characteristic of the first Industrial Revolution. It was on coal that nations built their industrial power in the nineteenth and early twentieth centuries. As early as 1860, Great Britain was producing over 80 million tons of pit coal, and it is probably not an exaggeration to say that, with the control of the oceans and overseas dominions, coal made her the dominant industrial power for several decades. Until the First World War it was the main coal-producing nations that held the world's key positions. In 1913 Great Britain, Germany, France and the USA between them produced almost 90% of the coal extracted in the whole world, and this production accounted for virtually all the world's energy supply. Throughout previous decades a 3% annual growth rate bore witness to the vigour of the industrial boom. Oil, however, was beginning to appear on the American market, and its use as a fuel was inconspicuously preparing the way for what was later to be one of the great mutations of the energy market.

Subsequently, the world entered a long period of disorder and trouble. There were two World Wars, an unprecedented energy crisis, and a massive invasion of oil into the energy market of the United States, which became the leading producer and exporter. The result of this shift to oil was a reduction in the share of coal in the world's commercial energy consumption from 61 % in 1950 to 51 % in 1960 and 35 % in 1970. The major transition took place in the mid-1960s. By 1967 oil had overtaken coal's global market share. This pattern was not uniform throughout the world; in the USSR rapid industrial expansion continued to be founded on coal.

After the Second World War oil production began to develop mainly outside the industrialized world—in Latin America, the Middle East, and later in Africa. The USA became, like Western Europe, an energy importer. A new balance in the world energy market came into existence based on the transfer of the developing countries' excess production to the deficient markets of the industrialized nations and by 1970 coal accounted for about 35 % of the world's total commercial energy consumption (UN, 1978).

There was an exceptionally rapid and regular growth in the wealth of both the CMEA (Council for Mutual Economic Assistance) and the OECD (Organization for Economic Co-operation and Development) countries. World energy requirements increased with this rapid economic progress at about 4 % per year in the 1950s and 1960s, but total coal production increased only 2 % per year over this period. Solid fuels now account for no more than 30 % of the total world energy supply and only 21 % in Western Europe (AGIP, 1980). The leading role on the developing world energy scene was played by liquid hydrocarbons, with new discoveries being made and decreasing prices mirroring the abundance of supplies.

If this history of global primary energy substitution between 1860 and 1975 were used to project market shares into the future, coal would be credited with only 10 % by the turn of the century (McDonald, 1981), but something of a crisis arose in the early to mid-1970s. Between 1973 and 1979 the market price of oil rose, in real terms, several-fold, producing a severe challenge to the energy balance which had prevailed for 20 years. Thus intense interest came to be expressed in forms of energy which could act as a substitute for petroleum. The large and diversified resource base of coal, as well as its relative cheapness in relation to other fuels, led to a belief in the return of coal following its post-war decline (IEA, 1978; Wilson, 1980; World Bank, 1979). A period of gradual reversion following its partial replacement by oil was expected.

These predictions of a rapid rise in coal demand have more recently been affected by industrial recession in Western countries and by increased conservation of energy. Industrial restructuring has suggested that a previously supposed link between economic growth and energy use was unfounded (Pindyck, 1979). Future trends in coal production, consumption and trade cannot be predicted accurately and extreme difficulties exist in distinguishing between temporary effects on the international energy scene and those which will result in long-term changes (Long, 1982; Prior, 1982). Coal has now been established as a relatively attractive fuel in many industrial applications, but its overall prospects will depend on economic growth in the major industrialized regions of the world and its acceptability from a social and management point of view. The timing, rate and extent of the introduction of new coal conversion technologies have become particularly uncertain because of trends in relative energy prices and industrial recession.

However, one important change which is occurring, and which will continue in future, is a growing volume of international trade in thermal coal (Long, 1982). But institutions which developed in association with more confined and regionally based coal production and utilization will become increasingly inadequate to deal with the more complex trading and financial arrangements. These inadequacies will also apply to the provision of necessary, long-term co-ordination to coal-related infrastructure projects which take many years to complete. The risks inherent in developing the necessary infrastructure are aggravated by an uncertain economic climate, a factor which itself could inhibit a reintroduction of coal (Fischer, 1984).

2. Resources and Reserves

A major difficulty in assessing coal resources and reserves on a worldwide basis is the lack of an internationally accepted and comprehensive system of resource classification. Concepts and classification systems which apply to mineral resources in general are discussed by Schanz (1975), Modelevsky, Safronov and Egel (1979) and Fettweis (1979a). Difficulties in the formulation of satisfactory classification systems are discussed by van Resenburg (1982); estimates are required in order to determine whether coal production and utilization within a specified time is likely to become constrained. This depends on the geological and technical factors which affect coal supply and those factors which determine demand for coal; for instance, the demand for products of industries which consume coal, the cost of developing transportation and other infrastructure required for coal utilization and the supply costs of other fuels. Controlling factors also include the social acceptability of coal use and legal constraints. Nordhaus (1978) considered the interaction between geological constraints and economic needs and used a model of resource exhaustion to estimate efficient rates of extraction for coal, petroleum and natural gas. The approach is reviewed and contrasted with other world energy models by Ulph (1980).

In a joint agreement adopted in 1973 between the United States Geological Survey and the United States Bureau of Mines (US Department of the Interior, 1976a, 1976b), coal resources are classified on the basis of two independent parameters: known geological status and economic recoverability. This is illustrated in the McKelvey diagram shown in Fig. 1.1. The United Nations's classification is similar, but uses a different system of terminology (UN, 1979; Fettweis, 1979b). Resources are defined in the agreement as "concentrations of coal in such a form that economic extraction is currently or may become feasible". The appropriate time horizon for assessing economically feasible extraction is not clear, and different and often vague conventions are applied. In Canada the period is about 25 years (Department of Energy, Mines and Resources, 1975); some Soviet publications mention 100 years (Kusnetzov *et al.*, 1971); the World Energy Conferences of 1974 and 1980 (WEC, 1974, 1980) refer to the "foreseeable future"; and the World Energy Conferences of 1977/78 (WEC, 1978b) refer to "some time in the future".

Reserves in the United States Bureau of Mines/United States Geological Survey (USBM/USGS) classification are defined as "the portion of the identified coal resource that can be economically mined at the time of determination". The reserve is derived by applying a recovery factor to that component of the "Identified Coal Resource designated as the Reserve Base" (US Department of the Interior, 1976a). There is still confusion, however, about the meaning of "economically" mineable. Some versions of the USBM/USGS classification refer to the portion of the total resource that can be exploited "at a profit" (van Resenburg, 1982). Profitability of new mining operations in the United States and other Western countries is determined by discounting costs and revenues up to 30 years in the future and in addition is affected by the inherent commercial risks of mining ventures. Current and expected energy prices, including factors such as deregulation of energy prices which has occurred in the United States, transport costs, tax policies and environmental regulations all therefore determine the portion of resources economically mineable at the time of determination.

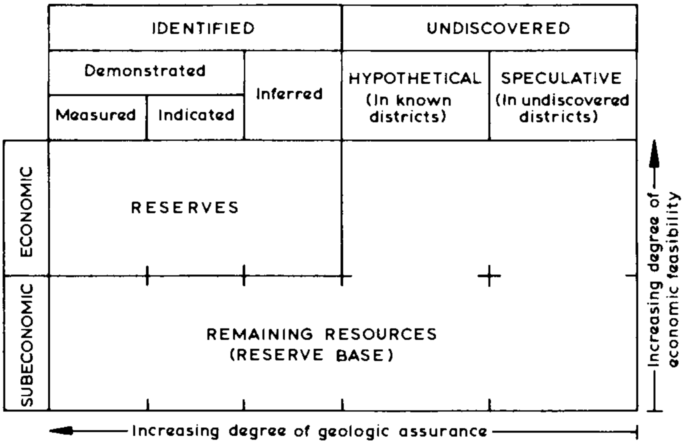


FIG. 1.1. McKelvey diagram: classification of coal resources by the US Geological Survey and the US Bureau of Mines.

In spite of these specific complications, it is understood that, in general, *resources* are quantities that may become economic at some time in the future and *reserves* refer to quantities that are known to be technically and economically recoverable under current conditions. Depending on movements in prices and costs, these specifications change and hence both resource and reserve estimates are subject to continuous re-evaluation (Eden *et al.*, 1981; IEA, 1982a).

The first worldwide survey of coal resources dates back to the 12th International Geological Congress (IGC) held in 1913 in Toronto. Since 1929 the World Energy Conference (WEC), formerly known as the World Power Conference (WPC), has issued worldwide surveys of commercial energy resources normally at intervals of 6 years. Since 1958 this has been done in arrangement with the United Nations. Between the main surveys of 1974 and 1980 (WEC, 1974, 1980) the WEC issued three additional reports dealing with coal resources (WEC, 1977, 1978a, 1978b).

In Tables 1.1 and 1.2 the period from 1960 to 1983 is considered. The figures for 1913, 1936, 1960 and 1962 are taken from an assessment of the WEC surveys by Fettweis (1979a). Data from 1968 onwards are obtained directly from the surveys of energy resources published by the World Energy (World Power) Conference (WPC, 1968; WEC, 1974, 1977, 1978a, 1978b, 1980, 1983). World totals inevitably aggregate data which are not strictly consistent, since they are obtained by following different guidelines in individual countries. For instance, in the 1980 survey the depth limit for hard coal reserves in the USA was 305 m compared with 1500 m in the USSR or FRG (WEC, 1980).

Changes in the way the WEC surveys are prepared, for instance changes in those organizations and individuals responsible for preparing the surveys and for answering detailed questionnaires in reporting countries, including changes in the conditions or limits underlying the resource estimates, also make comparison difficult. Internal inconsistencies and computational errors have also occurred, especially in the 1977 and 1978 intermediary reports (WEC, 1977, 1978b). Estimated ratios of reserves to production are shown in Table 1.3 on the basis of the WEC data. The current reserve-to-production ratio of about 240 years compares with a ratio of about 30 years for oil. This shows the vast potential of coal as an alternative fuel to oil over a very long period (IEA, 1982a). Whilst the WEC surveys are the best publicly available estimates of resources and reserves, it is known that multinational oil companies have additional information from drilling records and it is likely that coal resource estimates for countries in Africa and Latin America shown in Table 1.4, based on the 1980 survey, are underestimates of the true figures. Coal resources have a more even geographical distribution than oil deposits, although most of the reported reserves are in the Northern Hemisphere. Deposits in North America and Western Europe have been extensively exploited, but in regions such as western Siberia and northern China resource and reserve estimates are likely to be increased in future. The

TABLE 1.1. *Evolution of coal resource estimates for selected countries in the World Energy Conference surveys (10^9 t)*[a]

Depth limit	1913 IGC 1200–1800 m	1936 WPC 1200 m	1960 WPC 1200 m	1962 WPC 1200 m	1968 WPC 1200 m	1974 WEC none	1976 WEC none	1978 WEC 2000 m	1980 WEC none[e]	1983 WEC none[f]
China	997	1012	1012	1012	1012	1000	1003	[b]	1467[g]	1567[g]
USA	3234–3839	2913	1673	1506	1506	2925	3598	3600	3600	1570
USSR	234	1200	1213	5979	5528	5714	5710	6790	5926	4406
Australia	166	150	64	110	112	199	350	372	780	785
Botswana	n.a.	n.a.	n.a.	n.a.	[b]	5	[b]	[b]	107	107[h]
Canada	1217–1235	815	86	84	85	109	104	28	474	70
FRG	249	254	286	293	132	287	287	292	285	285
United Kingdom	189	176	170	169	165	163	164	190	190	195
India	n.a.	n.a.	n.a.	n.a.	108	83	83	86	114	115
Poland	n.a.	n.a.	n.a.	n.a.	61	61	[b]	[b]	184	184[h]
South Africa	56	205	70	63	72	44	82	93	93	129
World total	6402–7397	7017	4894	9576	8806	10,754	11,696[c]	13,357[d]	13,476	9703[i]

[a] Rounded figures.

[b] Figures not reported.

[c] Total corrected for improperly accounted resource figures for Mexico and Australia. The original figure in WEC (1977) was 11,505 10^9 t.

[d] Totals published in tce. Figures shown are recalculated from WEC (1978b); missing data are taken from WEC (1980).

[e] Greatest depth reported: 2000 m.

[f] Greatest depth reported: 1500 m (Portugal 1800 m).

[g] WEC 1980 figures including only recoverable reserves in total resource figures. 1983 figure includes *in situ* reserves.

[h] WEC 1980 figure.

[i] Estimated.

n.a. not available.

TABLE 1.2. *Evolution of in situ coal reserves (recoverable coal reserves) for selected countries in World Energy Conference surveys* (10^9 *t*)[a]

Depth limit	1936 WPC 1200 m	1960 WPC 1200 m	1962 WPC 1200 m	1968 WPC 1200 m	1974 WEC none	1976 WEC none	1978 WEC 1500 m	1980 WEC none[b]	1983 WEC none[i]
China	[c]	[c]	[c]	[c]	300 (80)	200[d] (100)	[c]	99 (99)	200 (99)[j]
USA	[c]	77	81	81	364 (182)	396 (198)	398 (200)	398 (223)	429 (257)
USSR	309	247	201	249	273 (137)	277 (137)	276 (1)[c]	276 (233)	288 (240)
Australia	32	50	42	51	74 (24)	102 (48)	109 (62)	83 (59)	91 (66)
Canada	31	43	54	55	9 (6)	15 (9)	[c] (5)	16 (6)	12 (6)
FRG	80	129	133	132	100 (40)	100 (40)	99 (64)	99 (59)	99 (66)
Great Britain	129	128	127	12	99 (4)	101 (4)	[c] (45)	45 (34)	n.a. (5)
India	n.a.	n.a.	n.a.	13	23 (12)	23 (12)	23 (13)	23 (14)	28 (n.a.)
Poland	n.a.	n.a.	n.a.	39	39 (23)	[c]	[c]	76 (39)	76 (39)
South Africa	8	37	21	37	27 (13)	32 (13)	59 (18)	59 (25)	122 (52)
World total	1085	800	769	722	1105[h]–1420 (591)	1177[h]–1374[e] (713)	1283[h]–1427[f] (838)	1320[h]–1464[g] (882)	1414[h]–1520[k] (946)

[a] Figures rounded.

[b] Greatest depth reported: 1700 m.

[c] No figures reported.

[d] No *in situ* tonnage published, estimated from recoverable reserves assuming 50% recovery.

[e] Total not published, estimated from data from individual reporting countries as published in the 1976 survey (possibly slightly underestimated). Poland, Botswana accounted for with 1974 values; China with 200,10^9 tons (see note a).

[f] Total not published, estimated from data from individual countries as published in the 1978 survey. Missing data are taken from the 1980 survey. *In situ* tonnages for China and Great Britain estimated assuming 50% recovery.

[g] Total corrected assuming 50% recovery for China and Great Britain (i.e. *in situ* tonnage twice recoverable reserves).

[h] From 1980 survey. *In situ* assumed equal to recoverable reserves for China and Great Britain.

[i] Greatest depth recorded: 1500 m (Portugal 1800 m).

[j] 1980 WEC figure.

[k] Includes WEC estimates for missing data.

n.a. not available.

TABLE 1.3. *Ratio of world coal resources (R) to production (P) (adapted from IEA, 1982a)*

	World R/P ratio years
1974	189
1976	207
1977	230
1980	239

TABLE 1.4. *Coal resources (10^9 t) for selected countries and continents (from WEC, 1980)*

Region/country	Coal resources
Africa excluding South Africa	125
South Africa	129
Latin America	56
Brazil	16
Colombia	16[a]
Mexico	4
Latin America excluding Brazil, Colombia, Mexico	26
World total	9703

[a] 1981 estimate.

distribution of world resources and reserves on the basis of quality is shown in Table 1.5. The hard coal–brown coal distinction is based on an energy content of 23.8 GJ t^{-1} (ash-free basis).

3. Energy Use and Economic Growth

The relationship between energy use and Gross Domestic Product (GDP) remained relatively stable in Western industrialized countries before 1974. This led to some belief in a fixed proportionality between economic growth and energy consumption. Growth in world energy consumption in comparison with Gross World Product (GWP) between 1925 and 1975 is shown in Table 1.6. Regional differences have been important over the past 50–60 years, with centrally planned economies substantially increasing their share of GWP and energy use, whilst population increases have predominantly

TABLE 1.5. *World reserves of hard coal and brown coal (10^9 t) for selected areas (adapted from Eden et al., 1981)*

	Reserves	
	Hard coal	Brown coal
Africa	34	n.a.
Latin America	5	6
Australia	18	9
Canada	9	1
USA	113	64
China	99	n.a.
USSR	83	27
Western Europe	95	34
Other	37	3
World total	493	144

n.a. not available.

TABLE 1.6. *World energy trends 1925–75[a] (Eden et al., 1981)*

	Average annual percentage growth rates	
	1925–50	1950–75
Energy growth rate	2.4	4.8
Economic (GWP)[b] growth rate	2.6	4.6
Population growth rate	1.1	1.9

[a] Refers to commercial energy.

[b] Gross World Product.

occurred in developing regions (Table 1.7). The increases in energy prices which commenced in 1969 and continued to occur during the 1970s showed a belief in a fixed relationship between energy use and economic activity to be unfounded. A rapid decline in the ratio of energy consumption to industrial output was observed in the seven largest OECD countries in response to oil price rises (see Table 1.8). This trend was reinforced by the oil price increases of 1973 and 1974.

Some of these changes were, however, due to alterations in underlying industrial structure. There are large differences in the energy intensity of different industries (Table 1.9), and reductions in energy consumption per unit of overall industrial output can result from alterations in its composition.

TABLE 1.7. *Regional energy trends (Eden et al., 1981)*

	OECD		Centrally planned economies[a]		Developing regions	
	1925	1975	1925	1975	1925	1975
Percentage of world energy	86	59	9	29	5	12
Percentage of GWP[b]	74	64	13	22	13	14
Percentage of world population	23	18	39	33	38	49

[a] Including China.
[b] Gross World Product.

TABLE 1.8. *Index (1960 = 100) of industrial energy use per unit output (adapted from IEA, 1982a)*

	1960	1975	1980
United States	100	79	71
Japan	100	91	71
Germany	100	78	68
France	100	81	78
United Kingdom	100	79	67
Italy	100	75	63
Canada	100	87	80

For instance, the decline in iron and steel production, which is relatively energy-intensive, partly accounts for these changes. But even having taken these factors into account, it is clear that there has been a strong response to price changes. Adjustments are thought to take place slowly with, for instance in the case of household energy consumption, very gradual turnover of appliance stocks and changes in the nature of housing in relation to standards of insulation.

In Western market-orientated economies these developments have aroused interest in the underlying relationships between energy prices, energy consumption and economic growth. Econometric studies have shown that price is an important controlling factor for both industrial and residential energy consumption. From an analysis of international energy demand, Pindyck (1979) gives a long-term price elasticity for the residential sector of -1.1 in a group of nine countries. This indicates that after a period of adjustment a 1.0% price change results in a 1.1% change in energy use. Estimated price responsiveness for industrial energy users is given by an estimated price elasticity demand of -0.8. Other studies which have investigated the role of prices in the allocation of energy resources are reviewed in UK Department of Energy (1977). The stability of relationships

TABLE 1.9. *Sectoral industrial energy intensities (10^6 J\$$2^{-1}$)[a] for UK, Federal Republic of Germany and Italy (Eden et al., 1981)*

	Energy consumption/value added					
	United Kingdom		Federal Republic of Germany		Italy	
	1954	1974	1954	1973	1955	1973
Food	50	48	19	19	31	23
Iron and steel	371	231	176	107	126	152
Engineering	20	20	13	6	15	15
Chemicals	121	112	91	53	78	132
Textiles	47	29	19	13	19	16
Bricks	241	126	154	72	165	183
Paper	48	31	28	16	17	24
Mining	85	49	179	88	41	53
Total industry	70	49	56	29	45	50

[a] US \$ at 1970 prices.

estimated by these studies from historical data is difficult to predict because of technical change occurring in industry, alterations in the composition of production and social attitudes towards housing conditions and transport.

Most of the increased share in Gross World Product and energy use accounted for by centrally planned economies has resulted from economic growth in the USSR. Two-thirds of the energy produced in these regions is accounted for by the USSR, and economic trends since 1960 indicate some movement towards improved energy efficiency (Table 1.10). Projections indicate that energy efficiency is likely to show a moderate improvement in future.

TABLE 1.10. *Average annual changes in energy consumption and economic growth in the USSR (IEA, 1982a)*

	1961–5	1966–70	1971–5	1978–80	1981–5
Energy consumption	+6.6%	+4.8%	+5.1%	+3.4%	2.8–3.4%
National income	+6.5%	+7.7%	+5.7%	+4.2%	+3.4%
Ratio	1.02	0.62	0.90	0.81	0.82–1.0

4. Coal Demand

4.1. Energy Prices and Interfuel Substitution

Historically, the contribution from coal to total energy use has varied widely (Section 1). During the nineteenth and early twentieth centuries it was the dominant source of energy. In world regions where coal was available and where industrialization occurred, a substitution from wood to coal took place followed by displacement of coal by oil and gas. In the United States coal overtook wood as an energy source in the 1880s and petroleum overtook coal in about 1960. On a global basis the share of coal in commercial energy consumption fell from 61 % in 1950 to 35 % in 1970; the price of oil had been falling in real terms and adequate supplies were available on world markets to meet growing demand.

In 1973 the price of fuel oil rose above the price of coal, expressed on the basis of energy content. After 1973 relative prices stabilized and coal demand in OECD countries rose relatively gradually (0.9 % yr^{-1}). The share of coal in total primary energy continued to decline. Consumption of thermal coal for electricity generation increased in all regions, but this was partly offset by a fall in demand for metallurgical coal in steel making. Steel production contracted in North America and Western Europe, with a shift in production to the Far East. A short-term increase in overall coal demand did take place until after 1979 when a further jump in oil prices occurred. Increasingly, coal was substituted for oil in cement manufacturing. About 45 % of total production costs in cement industries are due to energy use, resulting in rapid responses to changes in relative energy prices. By 1980, 66 % of coal utilized in the OECD area was for electricity generation and the second largest single outlet was cement. Substitution by coal in electricity generation would have been even more pronounced in the absence of newly commissioned nuclear power plants ordered in times of more rapidly increasing electricity demand.

The real price of fuel oil reached a peak in 1981 and has since declined. Coal prices have been relatively stable, but have shown some tendency to follow short-term fluctuations in oil prices. Table 1.11 shows price trends for a group of Western industrialized countries between 1960 and 1980; coal prices rose at 4–8 % yr^{-1}, with the exception of the Federal Republic of Germany where more costly exploitation of deeper seams resulted in more rapid increases. The price of heavy fuel oil rose at a rate of 11–17 % yr^{-1}. Although oil and coal prices are linked in the short term, efficient operation of market forces would ensure that in the long term the price of coal will fluctuate around its supply cost (World Bank, 1979).

The surge in demand for coal, which resulted from the peaking of fuel oil prices, led to supply bottlenecks which were further aggravated by the fall in Polish exports. This led to a temporary rise in coal prices. However, in 1983 coal was still significantly more attractive commercially than heavy fuel oil, in terms of energy content, despite wide regional variations which existed because of differences in production and transportation costs. Steam coal prices, reported in 1983 for selected regions of delivery, are shown in Table 1.12. These are affected by relatively short-term factors and general stagnation in the coal market characterized by temporarily high stocks in Western European countries. Had coal markets been in equilibrium with no excess

TABLE 1.11. *Oil and coal prices 1960 to 1980 in selected countries (IEA, 1982a)*

		Annual average increase (%)[a]	
		1960–73	1973–80
Canada	Coal	−1.7	3.8
	Heavy oil	−1.9	14.3
France	Coal	1.3	7.6
	Heavy oil	−2.3	16.5
FRG	Coal	1.5	11.2
	Heavy oil	−0.5	10.8
Italy	Coal	1.0	7.5
	Heavy oil	−3.4	11.8
Japan	Coal	−2.8	5.3
	Heavy oil	−4.3	15.7
UK	Coal	0.0	3.7
	Heavy oil	−0.5	12.1
USA	Coal	3.7	5.1
	Heavy oil	1.5	16.5

[a] Refers to industrial prices, including both thermal and metallurgical coal, adjusted for inflation.

TABLE 1.12. *International steam coal prices (adapted from ICR, 1983)*

Delivery	Source	Energy content (Btu lb^{-1})	Sulphur content (%)	Price (US \$ t^{-1})
CIF prices[b]				
West Europe	South Africa	11,500	1	42–43
West Europe	Australia	12,000	1	50–51
Nordic	USA	12,000	1	45–47
Nordic	South Africa	10,700–11,000	1	41–43
Italy	USA	12,000	n. a.	43–45
Taiwan	Australia	12,000	1	43–45
FOB prices[c]				
	Germany	12,000	1	60
	Poland	11,000	1	35–37
	South Africa	10,700–11,000[a]	1	32–38
	Australia	11,800	1	39
	UK	12,000	1	26.5–45
	USA	12,000–12,500	1–1.5	38–43

[a] Depends on volatility.
[b] Cost, insurance and freight.
[c] Free on board.

demand or supply (stock building or destocking), prices might have been in the region of \$65 t^{-1} for US coal and \$70 t^{-1} for Australian coal (ICR, 1983).

4.2. Coal and Electricity Generation

Between 70 % and 90 % of the total quantity of coal consumed in planned economies is utilized for electricity generation. Globally this proportion ranges from 70 % in the United States to 50 % in Western Europe and 15 % in Japan. Industrial electricity demand has increased more rapidly in the past 30 years than electricity demand in the commercial and residential sectors. Electricity has displaced the direct use of coal in industry, although not as rapidly as gas and oil. Industrial electricity demand is linked to specialized applications where interfuel substitution is limited or impossible, for instance motive power and electrolysis. In the residential and commercial sectors electricity demand is linked closely with ownership of appliance stocks and a tendency towards saturation in ownership is being approached in certain countries. In applications such as space and water heating, electricity continues to be displaced by other fuels. In the United States, Sweden and the Federal Republic of Germany the development of district heating schemes is enabling coal to contribute to domestic heating requirements.

In spite of these factors it is likely that electricity consumption in OECD countries will continue to increase more rapidly than total primary energy (IEA, 1982a). Oil-based electricity generation is uneconomic in comparison with coal, but relative costs with nuclear power are approximately in balance in a number of countries. International Energy Agency (IEA) estimates for new installations are shown in Table 1.13; these refer to economic conditions in the United States, Western Europe and Japan. The choice between coal and nuclear power will depend on government policies towards environmental implications and trade-offs which exist between the relative capital and operating costs.

Regional imbalances in coal production and energy consumption in the USSR could be alleviated by long distance transmission of electricity generated at coal-fired power stations, but coal may be environmentally less acceptable than natural gas in combined heat and power stations because of proximity to densely populated housing and industry. In other major coal-producing countries such as Poland, Australia and South Africa, the largest single outlet for coal is electricity generation. New generating capacity is planned in Poland which will utilize lignite in minehead power stations and a large number of new coal-fired power stations are expected to be commissioned in Australia which will increase further the share of coal taken by the electricity generation market (James, 1984). In OECD countries the main uncertainties affecting the use of coal for electricity generation are development of nuclear power programmes and the growth of final electricity demand. Until the beginning of the 1990s, however, coal use can be predicted

TABLE 1.13. *Cost estimates (US $) for electricity generation (10^{-3} $ kWh^{-1}) by type of fuel (adapted from IEA, 1982a)*

	Oil 2 × 600 MW		Nuclear	Coal with FGD[b] 2 × 600 MW		
	Low sulphur	High sulphur with FGD[b]	PWR[a] 2 × 1100 MW	US	Europe	Japan
Capital cost	10.8	12.9	24.8	17.1	17.1	17.8
Operating cost	2.5	4.2	4.2	5.1	5.1	5.1
Fuel cost	54.6	47.6	10.0	16.0	26.0	26.0
Total cost	67.9	64.7	39.0	38.2	48.2	48.9
Basis:						
Capital investment ($ kW^{-1})	577	692	1331	920	920	956
(Initial investment $ kW^{-1})	(500)	(600)	(1000)	(760)	(760)	(790)
(Interest during construction $ kW^{-1})	(77)	(92)	(331)	(160)	(160)	(166)
Construction lead time (yr)	3	3	6	4	4	4
Fuel cost ($ toe^{-1})	$ 33 bbl^{-1}	$ 27 bbl^{-1}		$ 40 t^{-1}	$ 65 t^{-1}	$ 65 t^{-1}
	(238)	(194)	(40)	(60)	(100)	(100)
Conversion efficiency	37 %	35 %	34 %	33 %	33 %	33 %
Heat rate (10^3 cal kWh^{-1})	2300	2450	2500	2600	2600	2600

[a] Pressurized water reactor.

[b] Flue gas desulphurization.

with relative accuracy, since little can be done over this period to alter the fuel mix of generating capacity (Prior, 1982). Small amounts of operational overcapacity are required to accommodate unpredicted peaks in electricity demand, but this gives only a limited flexibility in fuel input.

4.3. Other Industrial Markets

Demand for metallurgical coal in iron and steel making can be regarded as a derived demand for products supplied by other industries; in particular, transport equipment, engineering, general manufacturing and construction. These other industries are generally less energy-intensive than iron and steel making itself; estimates for the United Kingdom (Eden *et al.*, 1981) indicate that in 1972 energy use per unit of value added was 354×10^6 J $\$^{-1}$ in iron and steel and between 20 and 35×10^6 J $\$^{-1}$ in engineering and manufacturing. In OECD iron and steel industries demand has been in the region of 250×10^6 t yr^{-1} since the 1960s (IEA, 1982a). Increases in the production of crude steel have been accompanied by modernization and improved energy efficiency. The use of fuel oil will continue to decline as a supplementary heat source and the overall world market for metallurgical coal is likely to remain stable. Changes in the pattern of trade will occur as production continues to shift away from traditional centres.

Energy requirements in the production of aluminium are dominated by electricity required in electrolysis. Heat requirements in other energy-intensive industries such as chemicals, paper manufacture, building materials and petroleum refining give scope for the additional utilization of coal in place of oil and gas. This will take place in large-scale industrial establishments which are favourably located with respect to sources of coal supply. Coal use is particularly suitable in cement production and significant cost reductions could be achieved for bulk steam raising in the manufacturing of textiles and engineering.

4.4. Coal Conversion

The considerable interest in coal conversion which resulted from the oil price increases of the 1970s has, more recently, been affected by a slackening of oil prices and economic recession in Western industrialized countries. The very large investment required for full-scale projects is not currently regarded as commercially viable. In the longer term, development would take place over a sequence of phases which is characteristic of new industries. In an initial phase it is necessary that processes be established both as technically and economically viable. Industrial capacity can then be expanded in order to establish synthetic fuel plants on a large scale. A final stage of development is achieved when production can be brought to levels to meet both national demand and export requirements. Each of the first two stages might take between 7 and 10 years. The extent to which synthetic fuels will ultimately be introduced depends on their attractiveness in comparison with the direct use of coal and the use of heavy crudes from tar sands and oil shale.

Conversion plants would be large because of economies of scale; for an H-coal process capital costs are estimated to be \$3.48 × 10^9 (1981 prices) assuming a "pioneer" plant with a coal input capacity of 22.5 × 10^3 t d^{-1} and based on economic conditions in the United States (de Vaux, 1982). A learning process generally occurs which can reduce the cost of subsequent plants. Capital costs might be halved, but estimates are necessarily speculative. Table 1.14 shows the estimated cost of coal conversion products for liquefaction processes which are under development or in operation. The main markets for coal-derived fuel oil would be ceramics, bricks, tiles, glass and other products where the direct use of coal for process heat presents technical difficulties. The use of fuel oil in large boilers, including electricity generation, would be displaced by direct use of coal as opposed to coal conversion products. Medium and light distillates from coal liquefaction would be used predominantly as a transport fuel. Naphtha could be used as a petrochemical feedstock.

Estimated costs of gasification for both high calorific gas and synthesis gas are shown in Table 1.15. A limited use for high calorific gas will be to supplement supplies of natural gas in some regions. Utilization of medium and low calorific gas will depend on the supply costs of other fuels, particularly direct use of coal, but certain manufacturing conditions are better suited to a clean coal derived gas. The use of gasification products as a chemical feedstock is likely to displace natural gas.

Possible rates of development for synthetic fuels are shown in Table 1.16. These are based on estimates formulated in 1980 and reported in IEA (1982b). They show low and high scenarios for the introduction of new coal technologies beyond the year 2000 and indicate rapid development after 1990. A number of factors will constrain these projected rates of development. An

TABLE 1.14. *Estimated mean cost (\$ GJ^{-1}) of coal conversion products (adapted from IEA, 1982b)*

Coal price (\$ GJ^{-1})	Hydrogenation mainly giving fuel oil (H-coal)[a]	Hydrogenation mainly giving light/medium distillates (NCB[b] process)	Synthesis mainly giving light/medium distillates (SASOL)[c]
1	4.0	5.2	8.3
2	5.6	6.7	10.9
3	7.1	8.1	13.5

[a] Hydrocarbon Research Company, Trenton, New Jersey, USA.
[b] UK National Coal Board.
[c] South African Coal, Oil and Gas Corporation.

TABLE 1.15. *Estimated cost ($ GJ^{-1}) of synthetic natural gas and medium calorific gas (adapted from IEA, 1982b)*

Coal price ($ GJ^{-1})	Product (and plant size) SNG[a] (3 GW)	MCG[b] (3 GW)	MCG (0.5 GW)
1	4.1–4.7	2.6–3.5	3.0–4.3
2	5.8–6.5	3.9–5.1	4.4–5.9
3	7.5–8.3	5.2–6.6	5.7–7.4

[a] Substitute natural gas.
[b] Medium calorific gas.

TABLE 1.16. *The contribution (10^6 tce) of new coal technologies under two scenarios*[a] *(IEA, 1980 in IEA, 1982b)*

	Scenario I				Scenario II			
	1990	2000	2010	2020	1990	2000	2010	2020
Coal liquefaction	5	39	223	467	11	109	420	646
High calorie gasification	13	24	53	78	13	31	48	73
Low/medium calorie gasification	–	–	10	18	–	1	10	18
Combined cycle	–	17	32	36	3	11	44	41
Fluidized bed combustion	2	14	14	3	2	16	8	4
Magnetohydrodynamics	–	–	–	8	–	–	3	16
Coal gas fuel cell	–	–	2	–	–	–	–	–
In situ coal gasification	–	–	1	2	–	–	2	3

[a] Scenarios I and II refer to "minimum system cost" and "accelerated security" respectively.

important factor is demand for synthetic fuels at prices which reflect costs of production. The timing of their introduction has therefore become increasingly uncertain. In the absence of adequate planning, any rapid rates of development could be constrained by supplies of specialized materials such as chromium, and adequacy of transport facilities, particularly railways, ports and new pipelines for synthetic products. In Western countries commercial constraints could arise from the availability of finance for relatively large-scale and risky undertakings. Development of coal conversion plants in the USSR would alleviate the difficulties of transporting Siberian coal. Installations to gasify and liquefy lignite have been constructed in the Moscow and Kansk-Achinsk regions.

5. Coal Supply

5.1. Production and Costs

The nine largest producers of coal (USA, USSR, People's Republic of China, Poland, FRG, UK, Australia, South Africa and India) account for about 85 % of total world output. In the past three decades diverse trends were established in the regional pattern of production. Between 1950 and 1959 coal production tripled in East European countries and remained stagnant in Western industrialized countries. Production in developing countries increased at a modest rate over this period (2.8 % yr^{-1}).

The restructuring of energy production and consumption to reduce reliance on oil has presented a range of coal production and transportation difficulties. In western areas of the USSR production is affected by unfavourable geological conditions and exhaustion of low seams. The major coal regions of the USSR are the Donetsk Basin in the Ukraine and South European Russia, the Moscow Basin, the Pechora Basin located inside the Arctic Circle, the Urals, the Karaganda and Ekibastuz Basins in Central Asia, the Kuznetsk Basin in western Siberia, the Kansk-Achinsk lignite deposits and the South Yakutsk region located in the East. Reserves in the Donetsk Basin are mostly located in thin and deep seams and production costs are high. Exploitation of shallow deposits is therefore required and technical developments are being introduced to enable more efficient mining of thin seams. Power stations located in the Moscow area are supplied predominantly from coal obtained in the Moscow Basin, but ash content is relatively high and calorific content low. Coalfields in the Pechora Basin are an important source of supply of coking coal to local steel industries, but mining conditions are difficult. Supplies from coal mines in the Urals for local industry have been declining.

Production costs are low in the Kuznetsk Basin where high quality deposits are located in thick seams. New strip mines have been planned. It is less costly to utilize this coal in the European region of the USSR than to use locally supplied coal in spite of the additional costs of transportation. Conditions are more favourable in the Ekibastuz Basin, where mining from thick seams can be undertaken at even lower cost. However, the coal has high ash content and specialized boilers are required for its utilization. Output expanded rapidly in the 1970s and further developments have been considered. Possible projects include electricity generation in large plants with high voltage, long distance transmission to the Moscow region. This would relieve pressures on railway capacity which have occurred. Mining costs are also low in the Kansk-Achinsk Basin, but there is a lack of local need for lignite. Coal conversion plants or transportation by slurry pipeline are technical possibilities, but would involve high capital cost. Various other coalfields in the USSR supply local needs, but deposits in other regions are not likely to be required on a large scale. Production in the different coal-mining regions of the USSR is shown in Table 1.17.

TABLE 1.17. *Production of coal (10^6 t) in the USSR*[a] *(de Vaux, 1982)*

	1970	1975	1979	1980	1981
USSR total[b]	624.1	701.3	719 (752)	716 (790–810)	
Minugleprom:	–	694.6	–	–	
Donetsk	218.0	221.5	208	203	(213)
Kuznetsk	113.3	134.0	(154.9)	(162.4)	(149)
Karaganda	38.4	46.3	(47)	(48.6)	(49)
Pechora	21.5	24.2	28.9	28	(29)
Ekibastuz	22.6	45.8	59.2	66.8	(72)
Kansk-Achinsk	–	27.9	33	34.5	(46)
Moscow	36.2	34.1	27	25	(23)
Degree of plan fulfilment %	–	102.5	95.6	88.2–90.5[c]	–

[a] Column figures in parentheses denote plan targets.
[b] Total includes coal produced outside Minugleprom, the Soviet coal Ministry.
[c] Based on original Five Year Plan targets.

The main producer of coal in Eastern Europe is Poland. Productivity is high by international standards, as a result of heavy investment which has been undertaken in modern underground mining techniques. In order to avoid bottlenecks in railway distribution which occurred in the 1970s, additional capital investment will be required. Prior to the early 1980s Poland was the world's largest exporter of steam coal. Coal will continue to be a major source of foreign exchange.

The major coal-mining areas of Australia are New South Wales, Queensland and Victoria. In New South Wales deep mining is predominant and the coal is suitable for coking. In the future surface mining is likely to become increasingly important. Currently over half the national output is obtained through surface mining, mostly in Queensland and Victoria. Historically, industrial labour relations in the Australian coal-mining industry have been difficult.

Good mining conditions and low wages allow South African coal to be sold at low prices on world markets. Steam coal is exported through Richard's Bay. Major domestic markets are electricity generation and conversion to liquids and chemical products. Demand for electricity in South Africa has been rising at a rapid rate. Coal is obtained mostly by underground mining, but surface mining is becoming increasingly important.

Coal deposits in the Appalachian Basin of the United States supply about half of the national output, mostly for metallurgical industries and electricity generation. Surface mining has become increasingly important, but exhaustion will lead to some reversion to underground mining in the 1990s. In many areas production is below potential capacity, with associated high labour unemployment. Moderate increases in production may be achieved through growth of exports, but the United States is a marginal source of supply on international markets. About 25% of national coal demand in the United States is supplied from the eastern interior. The coal is high in sulphur, with low ash content, and is used primarily for electricity generation. Production from this area could expand rapidly because of the proximity to potential users. This would, however, involve the disturbance of high quality agricultural land. The western interior of the United States supplies more limited markets.

The major world coal-importing region is Western Europe. Although the European Economic Community (EEC) contains some of the world's largest coal producers, high production costs will favour continually increased imports from other areas. A large number of coalfields are located in the United Kingdom, but the industry has been in decline for several decades and has suffered from financial problems as well as labour unrest. It is utilized mainly for electricity generation. Future prospects will partially depend on the nuclear power programme. The coal industry in the Federal Republic of Germany has also been in decline. Production takes place mostly from deep mines in the Ruhr region, and in some areas deposits are approaching effective exhaustion. Various government subsidies support the coal industry in the Federal Republic of Germany. Coal production in France has fallen dramatically over the past 30 years and the country is now one of the world's largest importers.

Amongst Western industrialized countries, production costs are lowest in South Africa, Australia and, to some extent, North America. Because of the exploitation of thinner seams at greater depths, production costs are highest in European countries. Polish coal is produced from underground mines at costs comparable with the United States. In all areas costs are generally lower for surface mining, which is a relatively capital intensive technique, and higher for underground mining which is relatively labour intensive. Table 1.18 shows

TABLE 1.18. *Coal production costs ($ t^{-1}) in selected countries*[a] *(adapted from IEA, 1982a)*

	Surface mining	Underground mining
United States	8–15	20–30
Canada	6–15	20–45
Australia	12–15	–
France	–	80–95
Germany	–	–
United Kingdom	–	45–75
South Africa	8–10	10–12
Poland	–	18–25

[a] 1978 prices (US $): refers to production in existing mines, as opposed to new mines.

estimated production costs for a selection of countries. Differences in calorific value and sulphur content do not permit a precise international comparison from these data.

5.2. *Developing Regions*

India produces about 48×10^6 t yr^{-1} and is planning a rapid expansion programme in order to supply domestic requirements. Korea produced 8×10^6 t in 1980 and Colombia is likely to expand production to over 15×10^6 t yr^{-1} from a rate of 2.5×10^6 t yr^{-1} in 1980. A large deposit located on the Caribbean coast of the country is being jointly exploited using surface mining techniques by a multinational oil company in collaboration with the state mining corporation (CARBOCOL). Feasibility studies have been undertaken for new mines in Zimbabwe and Botswana, but the landlocked location of these countries restricts export opportunities. Other coal-producing areas with potential for further expansion include Mexico, Brazil, Swaziland and Taiwan. World coal production by region and country is summarized in Table 1.19.

TABLE 1.19. *World coal production (10^6 tce) by region and country (IEA, 1982a)*

				Average annual rate of increase (%)	
	1950	1973	1979	1950–73	1973–9
OECD[a]	1065	929	1029	−0.6	1.7
North America:	522	530	635	0.1	3.1
United States	506	513	607	0.1	2.8
Canada	16	17	28	0.3	8.7
Oceania:	20	62	83	5.0	5.0
Australia	19	60	81	5.1	5.1
Japan	40	22	17	−2.6	−4.2
Europe:	483	315	294	−1.8	−1.1
Germany	150	133	126	−0.5	−0.9
United Kingdom	219	118	110	−2.7	−1.2
Non-OECD/non-CPE[b]	80	155	208	2.9	5.0
South Africa	26	53	76	3.1	6.2
India	33	57	75	2.4	4.7
CPE[b]	435	1224	1500	4.6	3.5
China	43	369	544	9.8	6.7
USSR	220	475	511	3.4	1.2
Poland	79	168	212	3.3	4.0
Czechoslovakia	35	61	66	2.4	1.3
World total	1580	2308	2737	1.7	2.9

[a] Organization for Economic Co-operation and Development.
[b] Centrally planned economies.

5.3. Transportation

Variations in transport costs result in differences in supply costs at points of final consumption. The cost of inland railway transportation for distances over 400 km is comparable with costs for transportation by inland waterway. Inland transportation costs vary widely in different countries. Maritime transportation benefits from substantial economies of scale; costs for transportation in vessels of 100,000 to 150,000 dwt between Australia and Europe can be half that for vessels of 25,000 dwt. Large vessels, however, require large-scale port facilities and long lead times are required to introduce necessary equipment. Voyage rates varied between \$4 and \$14 t^{-1} in 1983, but these prices were depressed by short-term slack capacity. Costs can be up to \$30 t^{-1} for transportation in relatively small vessels.

Approximately 90 % of South Africa's coal exports pass through Richard's Bay, which has the capacity for vessels up to 150,000 dwt. Planned increases in loading rates have been postponed to keep export capacity in line with export demand. The volume of exports through Richard's Bay reached 29×10^6 t in 1981 compared with export capacities of 1×10^6 t yr^{-1} and 2.5×10^6 t yr^{-1} in Maputo and Durban respectively.

After 1981, congestion at port facilities on the east coast of the United States was relieved by stagnation in the international market and several projects were cancelled at the planning stage. Projects under construction on the Gulf of Mexico will increase future capacity by 69×10^6 t yr^{-1}. Expansion of capacity on the east and west coasts of the United States will give an additional 62×10^6 and 14×10^6 t yr^{-1} respectively. In British Colombia port capacity was 31×10^6 t yr^{-1} in 1982 and an approximate doubling is planned. Expansion plans are also continuing in Australia with facilities to handle vessels of up to 160,000 dwt. Export capacity in New South Wales and Queensland is approaching 130×10^6 t yr^{-1}. In the major importing regions of Europe and Japan port expansion plans are consistent with projected capacity requirement. In certain areas of the Pacific, such as Taiwan and the Philippines, additional facilities may be required.

6. Coal Trade

6.1. Current Trade Requirements

In the early 1970s international trade took place mainly in relatively scarce and costly metallurgical coals. The use and production of steam coal were more regionally based with relatively insignificant international trading. Approximately 10 % of overall world production was exchanged on international markets. Fundamental change has now occurred through an increased volume of trading accompanied by relative stagnation in markets for coking coal and with increases in demand for steam coal for electricity generation. A consequence of this was serious congestion of port loading capacity in the United States and Australia. Operators in the United States were estimated to have lost exports amounting to 10 × 10^6 t due to lack of infrastructure (Long, 1982).

Major exporting countries are the United States, Poland, Australia, the USSR, Canada and South Africa, whilst the major importers are Japan, France and Italy. Imports into the United Kingdom and the Federal Republic of Germany are constrained by official quotas designed to protect domestic coal industries. The pattern of trade between world regions in 1980 is summarized in Table 1.20. In 1981 United States exports reached a record of approximately 100 × 10^6 t, but growing stockpiles of steam coal in Taiwan, The Netherlands, Denmark, Finland, Spain and Japan led to reduced orders and a decline in exports to approximately 95 × 10^6 t in 1982. The market for coking coal was less depressed than that for steam coal.

Major markets for US coal exist in Latin America, in particular Brazil, Argentina and Mexico. Supplies to these countries are mainly of coking coal. One of the largest importers of US coal is Japan. Europe accounts for about half of United States' exports, with most of the remainder taken by Brazil and Japan. European countries obtain coal from the other principal areas of Australia and South Africa, and intermittent imports are obtained from Poland and the USSR. Stagnation in Japan's production of crude steel has affected export opportunities from Australia, the United States and Canada. Increasingly, Japan is importing coal from China, the USSR and South Africa at prices generally below those from North American sources.

TABLE 1.20. *World coal trade (10^6 tce) in 1980 (IEA, 1982a)*

	Importers					
	OECD[a] Europe	Japan	Canada	USA	Others	Total
Exporters						
Australia	9	29	–	1	4	43
Canada	2	11	–	–	1	14
Europe (OECD)	31	–	–	–	–	31
Japan	–	–	–	–	2	2
Poland	15	1	–	1	15	32
South Africa	20	4	–	1	4	29
USA	36	21	16	–	8	81
USSR	4	2	–	–	17	23
Others	0	3	–	–	–	3
Total	117	71	16	3	51	258

[a] Organization for Economic Co-operation and Development.

6.2. Outlook

Because of the movement of steel production away from traditional centres, the volume of coking coal which is traded on world markets may show some increase in spite of stagnation in the size of the overall market. In addition to this, low cost opencast mining in developed countries, particularly the United States, Canada and Australia, as well as existing or planned low cost operations in South Africa, Colombia and Indonesia, will lead to a rapid increase in international steam coal trade over the next two decades. The rate of increase in the volume of trade will be more rapid than overall production and consumption. This continuing internationalization of coal trade will be encouraged by pressure on high cost producers, particularly the United Kingdom, the Federal Republic of Germany, Belgium and France, to substitute imported for domestically produced coal. The extent to which this occurs will, however, depend on the domestic energy policies pursued in these countries.

Increases in steam coal trade will be due mainly to direct combustion in large-scale energy-intensive installations which can benefit from the advantages of bulk transport. Conversion to coal is most favourable for new or replacement boilers, but slow economic growth in Europe has reduced the rate of turnover of installations. Increases in imports are likely to Denmark, The Netherlands, Spain and Sweden. In addition, Japan, South Korea and Taiwan are likely to become major importers by the end of the 1980s, but in Pacific countries the use of steam coal is relatively new and increases in consumption will be preceded by development of power stations and ports. In the longer term, substitution to coal on a larger scale is possible in these areas. Australia will be the main source of supply, although the USSR and China may supply significant quantities to importers in the Pacific. Colombia and South Africa will be important for European consumers. Production potential for selected developing regions is summarized in Table 1.21; actual production will be constrained by industrial requirements in developed countries.

Future development of the overall regional pattern of coal production, consumption and trade is subject to the major uncertainty of general

TABLE 1.21. *Coal production potential (10^6 tce) in developing countries or regions (IEA, 1982a)*

	1980	1985	1990	2000
India	107	102–118	143–157	220–259
Korea	17	16–28	28–35	33–42
Mexico	8	9–28	11–19	24–33
Brazil	6	9–28	16–20	19–24
Colombia	6	9–13	17–24	39–47
Taiwan	3	8–11	9–14	11–16
Others	5	8–13	13–28	126–231

economic growth in Western countries. Prospects depend on economic growth in the major industrialized countries as opposed to constraints on supply or excessively high prices relative to other forms of energy. If economic growth proceeds rapidly in the 1990s, a renewal in the need for rapid port developments could occur. Beyond the end of the century commencement of coal conversion on a large scale would lead to additional import requirements. Global developments which can be expected are shown in Table 1.22.

TABLE 1.22. *Outlook for coal production, consumption and trade ($10^6 t$) in world regions*

	1980	2000
Demand:		
Western Europe	400	600
Centrally planned economies	1339	2457
North America	616	900
Other regions	282	764
Total	2737	4721
Supply:		
Western Europe	294	310
Centrally planned economies	1369	2507
North America	695	1046
Other regions	379	858
Total	2737	4721
Net exports:		
Western Europe	– 106	– 290
Centrally planned economies	30	50
North America	79	146
Other regions	57	94

Total production and consumption are likely to increase by approximately 70 % by the year 2000 in comparison with 1980. This would represent a more rapid growth than other forms of primary energy and a continuing increase in the share of coal in overall energy use.

Low cost supplies from developing countries and possibly from centrally-planned economies will result in increased imports from these areas into Western Europe. Relatively high cost North American sources would moderately increase exports for European consumption. An important component of trade, not shown in Table 1.22, will be between Japan, South Africa, Australia and developing countries.

7. Institutional Developments

Growing internationalization of coal will make it a more dependable source of energy with less reliance on a limited number of suppliers. Although coal deposits are regionally dispersed, an increasing concentration of control over production and trade may occur. A significant growth of trade will give rise to financing requirements equivalent to about 4% of overall investment requirements in OECD countries. This could continue over a period of two decades or more, but sudden surges in financing requirements would compete with other industrial investment needs through increases in interest rates. In developing countries, financing problems are likely to be the most severe, so here emphasis will continue to be on private investment in export projects. Concentration of control will be encouraged by the more complex and co-ordinated planning required in international trade, including expertise in marketing, finance, transportation and, in particular, by the need to reduce the risks inherent in long-term coal infrastructure projects. Difficulties in raising finance partly account for increased concentration of production which has already occurred in the United States of America and Australia. In the USA price increases in the 1970s raised the profitability of the coal industry, but since then a worsening financial situation has been accompanied by the operation of larger mines by fewer companies. Multinational oil companies have been increasingly active in developing countries and new problems, in relation to the economic, environmental and social impacts in these areas, will arise and will require effective management.

A limited degree of competition may emerge between exports from centrally planned economies and organizations controlling coal exports in Western countries; Western Europe and Japan will continue to import coal from all global regions. Priorities for the coal industry in the USSR will be influenced by the imbalance between regional supplies and consumption and general development priorities for the eastern and western areas. Coal development will also be dependent on priorities given to new oil and gas deposits and requirements for foreign exchange obtainable from coal exports.

8. References

AGIP (1980). *Eni: Energy and Hydrocarbons 1978*. AGIP, Rome.

AVERITT, P. (1975). *Coal Resources of the United States, January 1, 1974*. USGS Bulletin 1412, US Geological Survey, Washington, D.C.

DEPARTMENT OF ENERGY, MINES AND RESOURCES (1975). *Departmental Terminology and Definitions of Reserves and Resources (Metals, Industrial Minerals and Coal)*. Interim document, Department of Energy, Mines and Resources, Ottawa.

EDEN, R., POSNER, M., BENDING, R., CROUCH, E. and STANISLAW, J. (1981). *Energy Economics: Growth, Resources and Policies*. Cambridge University Press, Cambridge.

FETTWEIS, G. B. (1979a). *World Coal Resources. Methods of Assessment and Results*. Elsevier, Amsterdam.

FETTWEIS, G. B. (1979b). A proposal for distinguishing between occurrences and resources of mineral commodities with special reference to coal. *Future Coal Supply for the World Energy*

Balance (ed. by M. Grenon). Third IIASA Conference on Energy Resources. Pergamon, Oxford.

FISCHER, W. (1984). *Coal Trade Statistics Volume II.* Robertson Research, Llandudno.

ICR (1983). *International Coal Report No. 65.* Financial Times Business Information Ltd., London.

IEA (1978). *Steam Coal Prospects to 2000.* OECD, Paris.

IEA (1982a). *World Energy Outlook.* OECD, Paris.

IEA (1982b). *The Use of Coal in Industry.* Report by the Coal Industry Advisory Board, OECD, Paris.

JAMES, P. (1984). *The Future of Coal.* 2nd edition. Macmillan, London.

KUSNETZOV, K. K., JASSTREBOV, A. I., DABYDOVA, E. A., DZIOV, V. A., KLEPIKOV, L. N., MKRTYCJAN, P. I., SLOBODCIKOV, P. I., SMIRNOV, V. M. and TRETJAKOV, K. M. (1971). *Coal Deposits for Opencast Mining.* Nedra, Moscow.

LONG, R. (1982). *Constraints on International Trade in Coal.* IEA, EAS Report G3/82, London.

MCDONALD, A. (1981). *Energy in a Finite World: Executive Summary.* IIASA, Laxenburg.

MARCHETTI, I. E. and NARICENOVIC, N. (1979). *The Dynamics of Energy Systems and the Logistic Substitution Model.* RR-79-13, IIASA, Laxenburg.

MODELEVSKY, M. S., SAFRONOV, D. S. and EGEL, L. E. (1979). Coal reserve classification used in different countries. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Third IIASA Conference on Energy Resources. Pergamon, Oxford.

NORDHAUS, W. D. (1978). The allocation of energy resources. *Brookings Papers on Activity*, **3**, 529–70.

PINDYCK, R. S. (1979). *The Structure of World Energy Demand.* MIT Press, Cambridge, Massachusetts.

PRIOR, M. (1982). *Steam Coal and Energy Needs in Western Europe Beyond 1985.* Economic Intelligence Unit, Report 134, London.

SCHANZ, J. J., Jr. (1975). *Resource Terminology: An Examination of Concepts and Terms and Recommendations for Improvement.* EPRI Report No. RP384-1, Electric Power Research Institute, Palo Alto, California.

UK DEPARTMENT OF ENERGY (1977). *Report of the Working Group on Energy Elasticities.* Energy Paper No. 17. Department of Energy, London.

ULPH, A. M. (1980). World energy models—a survey and critique. *Energy Economics*, **2**, 46–59.

UN (1978). *World Energy Supplies: 1972–1976.* UN, New York.

UN (1979). *The International Classification of Mineral Resources.* Economic Report No. 1. Centre for Natural Resources, Energy and Transport, UN, New York.

US DEPARTMENT OF THE INTERIOR (1976a). *Principles of the Mineral Resource Classification System of the US Bureau of Mines and the US Geological Survey.* USGS Bulletin 1450–A, Washington, D.C.

US DEPARTMENT OF THE INTERIOR (1976b). *Coal Resource Classification System of the US Bureau of Mines and US Geological Survey.* USGS Bulletin 1450–B, Washington, D.C.

VAN RESENBURG, W. C. J. (1982). The relationship between resources and reserves. Estimates for US coal. *Resources Policy,* March 1982, 53–8.

de VAUX, G. R. (1982). Coal liquids—costs and timing. *Energy*, **7**, 12–4.

WEC (1974). *Survey of Energy Resources, 1974.* WEC Central Office, London.

WEC (1977). *Survey of Energy Resources, 1976.* WEC Central Office, London.

WEC (1978a). *World Energy Resources 1985–2020.* Executive summaries of reports on resources, conservation and demand to the Conservation Commission of the WEC 1977, Istanbul. IPC, Guildford.

WEC (1978b). *Survey of Energy Resources, 1978.* WEC Central Office, London.

WEC (1980). *Survey of Energy Resources, 1980.* Federal Institute for Geosciences and Natural Resources of the FRG, 11th World Energy Conference, Munich.

WEC (1983). *Survey of Energy Resources, 12th WEC Congress Edition.* World Energy Conference, Central Office, London.

WILSON, C. (1980). *Coal—Bridge to the Future: Report of the World Coal Study, WOCOL.* Ballinger, Cambridge, Massachusetts.

WORLD BANK (1979). *Coal Development Potential and Prospects in the Developing Countries.* Washington, D.C.

WPC (1968). *Survey of Energy Resources, 1968.* WEC Central Office, London.

2

Land Disturbance and Reclamation after Mining

1. Introduction 29
2. Classification of Mining Land Disturbance 30
3. Land Reclamation 31
   3.1. Reclamation Objectives and Methods 31
   3.2. Framework of Land Reclamation Requirements 32
   3.3. Developments in Mining Technology and Land Reclamation 36
      3.3.1. Strip-mining techniques 37
      3.3.2. Improvements in site engineering technology 37
      3.3.3. Improvements in surface soil treatment 38
      3.3.4. Land-use efficiency improvements 39
      3.3.5. Improvement of underground mining waste reclamation 39
   3.4. Progress in Land Reclamation 40
      3.4.1. General considerations 40
      3.4.2. Land disturbed by surface mining 41
      3.4.3. Land disturbed by underground mining 44
4. The Economics of Reclamation 45
5. References 45

1. Introduction

Irrespective of the actual methods of coal mining being used, mining processes unavoidably bring about land-surface disturbances, and increased mining for expanded coal supplies will inevitably lead to extended land disturbances. Such disturbed land tends to create a negative effect on the surrounding environment and reduces its economic value.

The character of the disturbance depends upon the disposition of the mined strata, and upon the methods used to mine them (Fig. 2.1). Continued improvement of coal-mining methods is associated with the exploitation of deeper coal seams (below 1000 m in deep mining operations and a few hundred metres in surface or strip-mine operations). Generally speaking, progress in mining has led to greater use of open coal extraction methods which produce larger-scale disturbances on the earth's surface. At the same time, research and development has provided greatly improved engineering and biological methods of landscape reclamation.

Although dramatic changes in landscape may sometimes occur as a result of open mining, underground coal extraction on occasion causes such gross land disturbance that reclamation runs into much greater difficulties and costs than in open mining. If geographical, social and other conditions are similar, such land characteristics as the physical and chemical properties of the rocks and soil in the rooting zone, site permeability and moisture content predetermine the method and technology of the reclamation required. Therefore, study and classification of such properties of disturbed substrates are considered as very important.

2. Classification of Mining Land Disturbance

The following classification, of land disturbance due to coal mining, is adapted from a more general and comprehensive classification of Motorina and Ovchinnikov (1975) and Bauer and Weinitschke (1973). The classifications of Motorina and Ovchinnikov (1975) include one based on relief features and another on overburden characteristics. Although designed for the USSR, they can be adapted for other countries.

Land disturbance due to coal mining

1. Land disturbed by surface mining (strip-mining and open-pit mining).
   1.1. Open-pit mining disturbance.
      1.1.1. Terraced excavations usually over 30 m deep; deposit usually dipped at >30°; overburden stored externally (Fig. 2.1A).
      1.1.2. Terraced excavations over 10 m deep; deposit usually 8–30°; most over-burden stored externally (Fig. 2.1B).
      1.1.3. Terraced excavations usually 10–30 m deep with deposit dipped at 8°; some internal storage of overburden (Fig. 2.1C).
   1.2. Strip-mining disturbance.
      1.2.1. Area strip-mining of horizontal deposits relatively near the surface (Fig. 2.1D).
      1.2.2. Contour strip-mining of horizontal or steeply dipped deposits in mountainous regions (Fig. 2.1E).
2. Land disturbed by underground (deep) mining.
   2.1. Land subsidence.
      2.1.1. Canyon subsidence caused by working medium or thick seams that dip steeply (>45°).
      2.1.2. Terraced subsidence resulting from the working of seams that show 27–45° dipping and where the land surface slopes.
      2.1.3. Bowl or cirque-type subsidence caused by the working of medium or thick seams dipping up to 27°.
      2.1.4. Trough-type subsidence resulting from mining low and medium thickness, horizontal or slightly inclined seams; appear like natural depressions.
   2.2. Surface waste deposits associated with pitheads.

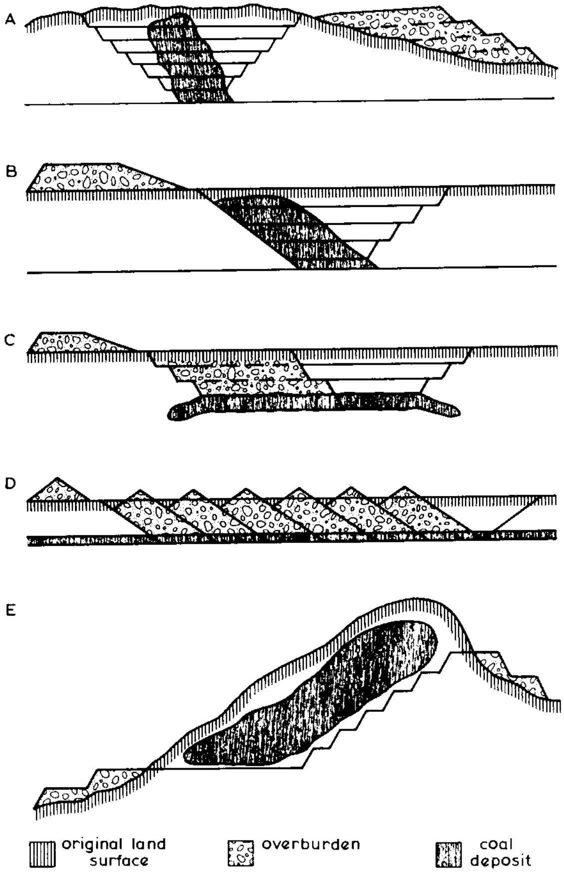


FIG. 2.1. Land disturbance by mining: A-open pit deposit deeply dipping; B-open pit with medium dip to deposit; C-open pit with horizontal deposit; D-conventional area strip-mining; E-contour strip-mining of deeply dipped deposit.

2.2.1. Plateau-shaped tips resulting from single- or multi-level tipping from road or rail transport.
2.2.2. Crest-shaped tips resulting from cableway dumping.
2.2.3. Conical tips resulting from skip or tip-wagon dumping.

Further detailed consideration of surface mining disturbance is given by Grim and Hill (1974).

3. Land Reclamation

3.1. Reclamation Objectives and Methods

The changes in the environment effected by the coal industry are so profound that often no natural recovery of disturbed landscapes can occur.

Land reclamation has emerged as a method of countering the negative after-effects of extracting coal and other minerals. Land reclamation is also a comprehensive interdisciplinary subject which covers the problems of landscape redevelopment and the restoration of its productivity, ecological integrity and economic and aesthetic value.

Disturbed lands constitute a form of environmental damage produced by the mining industry. If reclamation can be included in the production process, the mining industry could become, given a reasonable time-span, something near a "closed-cycle" industry producing no wastes, However, some overriding climatic factors may result in permanent losses, or the need for continued inputs to the system. This would be true, for example, in very high-rainfall or semi-arid regions respectively.

At the present time land reclamation has to be implemented sequentially (Motorina and Ovchinnikov, 1975). The first step involves planning, followed by research and development. The second step is site preparation for a specific use, consisting of earth-moving and shaping of the surface, water regime control and sometimes the application of productive layer, like topsoil or organic material on the surface. The third step entails biological reclamation which will allow the development of a restored site or landscape. The goal of the third step is to restore fertility and biological productivity to the disturbed land. The six main uses of reclaimed land are as follows, and depend on natural and socio-economic factors of the locality:

(1) agriculture (arable land, orchards, meadows or grazing land);
(2) forestry (commercial or non-commercial forests);
(3) recreation (recreation zones, parks, or public open space);
(4) water use (fishing, swimming, boating and yachting);
(5) construction (sites for light industry, houses or service purposes);
(6) wildlife habitats (wilderness areas).

These uses may be closely interrelated and often form a comprehensive development in an area of extracted deposits.

3.2. Framework of Land Reclamation Requirements

In recent years many countries have developed and adopted laws, national programmes and specific policies for environmental protection.

The basis of most laws applicable to the mining industry and its effect on land seeks to control land management, protect resources and regulate land reclamation and landscape restoration. Although these laws are often specific to activities of the mining industry, they constitute an integral part of national efforts to optimize living conditions for man. The costs involved in environmental improvement are borne, in the first place, by those responsible for the mining activity. There is no doubt that legal provisions of this kind contribute to rises in production costs.

The Constitution of the USSR provides that land and its resources are owned by the Soviet society. Since the national economy is planned, the country has all prerequisites for genuinely integrated and rational use of natural resources. Article 18 of the Constitution emphasizes the importance of natural resource protection in the interests of present and future generations.

All land reclaiming activity in the country is controlled by a series of legal Acts. The fundamentals of land reclamation are decreed in *Fundamentals of Land Legislation*, the land codes of the Union Republics and other legislation. For example, Article II of the *Fundamentals cf Land Legislation* makes it mandatory to reclaim the lands allocated for surface and underground mining of minerals; to bring disturbed cultivable lands and forests into a usable state; to do everything possible to restore agricultural land to a usable state in parallel with the production process, and if this is impossible to do, carry out the necessary reclamation no later than a year after production has been completed; to remove and store fertile soil layers in any work which involves soil disturbance so that it can be reused during reclamation.

An increasing concentration of disturbed land in mining areas of the USSR has made the problem of land reclamation the most important long-term programme of the nation in the field of natural resource management. The Special Degree of the Council of Ministers of the Soviet Union of 2 June 1976—*On the Reclamation of Lands, Preservation and Rational Use of Fertile Soil Layers in Mining Materials and Peat, Exploration Surveys, Construction and Other Works*—provides that the reclamation of the areas where minerals have been already extracted shall be funded by the state from special allocations, and industries with active mines shall reclaim disturbed lands at their own expense, all the costs involved being covered from production costs. The research institutes of the Ministry of Agriculture of the USSR are assigned the task of developing reclamation programmes. State control of the programme's implementation is exercised through local authorities and land management offices.

This logical sequence of laws and codes of practice in the field of land reclamation envisages a high rate of recovery of disturbed lands and during the ninth 5-year plan by 1975, the areas of reclaimed lands increased two and a half times compared with 1971. The proportion of the lands returned to agriculture increased considerably. For the Ukraine and Steppe region of the RSFSR it reached 70–75 %. This high rate of reclamation is a result of setting up special land reclamation divisions within coal production associations. Within the Ministry of Coal Industry there is a division for the protection of nature with its own research institute. The institute conducts comprehensive studies on problems related to reclamation of land disturbed by coal mining.

In the German Democratic Republic (GDR), land reclamation was first officially endorsed in 1951 in the law on the *Restoration of Economic Value of Territory Occupied by the Mining Industry, Including Waste Storage*. At the present time various codes of practice in the GDR strictly define the rights and obligations of production units whose activity causes land disturbance. Legislation determines the procedures to be followed if damage to former users occurs, including the procedure for reimbursement of losses by farmers' co-operatives when the disturbed area is greater than 20% (Motorina and Zabelina, 1968).

In Czechoslovakia (CSSR), laws concerning the use of natural resources (1957), protection of land resources (1976), forest law (1977) and building code (1976), the obligations of industrial establishments in relation to soil, landscaping, water regimes and other subjects are specified. Legislation requires long-term plans for the reclamation of lands in the large industrial areas of the country. For example, the master plan for the restoration of the North Czech brown coal area contained detailed maps and calculations of the total land-use up to 1980. Projects provide for parks and green zones around industrial establishments. The costs involved in reclamation activity in the CSSR are borne by the mining industry.

In Poland, planned reclamation started in 1961 when the Economic Committee of the Council of Ministers made it mandatory, by a special decree for the Ministry of Mining Industry and Power, to reclaim disturbed lands. Further, a considerable effect on land reclamation activity was produced by the 1966 Act of the Council of Ministers and the 1971 Law on the *Conservation of Agricultural and Forestry land and its Recultivation.* This legislation made land reclamation obligatory during mining operations, specified necessary procedures, and dealt with legal and financial considerations. Reclamation work is carried out by the coal producers as a cost to them.

Land reclamation is legislated for in Bulgaria, Hungary, Romania and Yugoslavia. Although there are differences in organization and management, a number of common principles can be discerned:

(1) land reclamation activity is included in national plans and constitutes a part of natural resource management;
(2) objectives and methods of reclamation must be determined at the design stage of the mine; mining plans must incorporate land reclamation as an integral part of production processes;
(3) before reclamation plans are drawn up a comprehensive study is carried out.

In the United States of America the *National Environmental Policy Act* of 1969 covers a wide area of national policy in relation to environmental problems. The Act requires the preparation of a document to be used by the federal authorities in their decision process for all major federal actions that significantly affect the environment. The federal authorities must take into account:

(1) the environmental impact of the proposed action;

(2) any adverse environmental effects which cannot be avoided should the proposal be implemented;
(3) alternatives to the proposed action;
(4) the relationship between local short-term uses of man's environment and the maintenance and enhancement of long-term productivity;
(5) any irreversible and irretrievable commitments of resources which would be involved in the proposed action should it be implemented.

Such a document is required wherever federal action is involved in the mining process, either direct or indirect (for example, the federal licensing of a related activity).

Until 1977, individual states in the USA exercised primary jurisdiction over the restoration of mined lands. State legislation usually authorized a state agency to issue permits to mines on receipt of details on the mine site, mining and reclamation plans, and a bond or other security to cover the restoration.

Surface and mineral rights are owned separately and diversified ownership among private individuals, the federal government and the state governments exists. Until 1976 it had been the policy of the federal government to allow the individual states to act for it where federally owned coal was being mined, but this was changed by the 1977 federal *Surface Mining Control and Reclamation Act*. This created overall regulations for coal mine reclamation and established minimum reclamation performance standards. The standards include restoration to the approximate original contour, segregation and replacement of topsoil, establishment of vegetation comparable to pre-mining conditions, and protection from adverse hydrological effects. Operators must take responsibility for successful revegetation for 5 years after seeding (for 10 years where mean annual precipitation is less than 66 cm).

Thus, since 1977, reclamation of surface-mined land for coal is initially the responsibility of the Office of Surface Mining within the US Department of the Interior, but individual states are allowed to take this over where they develop state programmes that comply with the procedures in the Surface Mining Act and enforce permanent standards at least as stringent as the federal ones. If state programmes are not developed, they have no regulatory authority over the reclamation of coal-mined land. The Surface Mining Act placed a levy per ton on all coal produced in the USA. This fund can be used to finance the restoration of areas that were not reclaimed in the past and other action needed to alleviate off-site impacts of the unreclaimed areas.

From 1981 a new consideration has been given to the Surface Mining Control and Reclamation Act. Without amending the Act, it is intended that the rules should be streamlined to reduce federal involvement in the Act's enforcement and increase the discretion of state authorities in interpreting the law according to regional conditions. It will probably result in a reduction in site inspection by the federal agencies and remove certain specific requirements such as those relating to the grading of coal haulage roads and the method of treating mining water runoff. Regulations relating to the planting density of forest trees on reclaimed forest land may also be relaxed in the light of local conditions.

In the Federal Republic of Germany considerable preplanning of reclamation has occurred as coal mining has impinged on areas of intensive agriculture. As early as 1920, a law established an association to deal with regional planning in the Ruhr. The *Regional Planning Act* of 1962 made this body responsible for a development plan which includes refuse disposal planning and land utilization policy. Open space and amenity areas must be secured, often by the renewal of derelict areas. The location of tips is determined, suggestions for combining waste materials are made and landscaping plans and planting schemes approved. This provides integrated planning control for land restoration.

In the Rhenish brown coal district of the Federal Republic of Germany a soft brown coal has been mined by surface methods since the 1950s. An integrated plan defines the limits of mining, the location of industry, agriculture and forestry and the reclamation schedule. This operational plan is approved by the *Land* parliament.

In the United Kingdom local government has had powers to acquire derelict land for restoration since 1944 (*Town and Country Planning Act*) and in 1951 the *Mineral Workings Act* provided for a fund for financing the restoration of land made derelict by surface mining by stripping. Various Acts made provision to meet the high cost of land restoration by making grants available. These have ranged from 50% to the total cost in special development, development, intermediate and derelict land clearance areas. Since 1974, as a result of an amendment to the *Town and Country Planning General Development Order 1973*, a county planning authority may require the National Coal Board to submit a scheme aimed at ensuring that waste material from deep mines is tipped in such a way as to facilitate subsequent landscaping and restoration to improve visual amenity. Strip-mining land restoration has been developed extremely successfully by the Opencast Executive of the National Coal Board.

In the People's Republic of China the Standing Committee of the Fifth National People's Congress approved the Environmental Protection Law in 1979 for trial implementation. In 1982 the Ministry of Urban and Rural Construction and Environment Protection was created which implements and supervises the carrying out of the national guidelines, policies, laws and acts relating to environmental protection. Land protection figures prominently in many of the articles of the law (Geping and Lee, 1984).

3.3. Developments in Mining Technology and Land Reclamation

During the last 15–20 years considerable progress has been made in reclamation technology geared to the biological improvement of disturbed land and the creation of optimally organized landscapes. This generally involves:

(1) careful separation and placement of different categories of mining waste;
(2) earth movement and landscaping to meet fully the requirements of potential users;
(3) careful analysis and amendment of waste materials to ensure good plant establishment and growth;
(4) attention to drainage and other hydrological factors.

International experience shows that the site engineering stage of reclamation is the most costly part of the operation. If, however, engineering works are included in the production process and planned in the course of mining operations, they may become economically effective and efficient.

3.3.1. Strip-mining techniques

Experts in Czechoslovakia believe that the opening pioneer cuts in surface mining operations can be located in such a way that, at the final stage of mining, overburden and soil material is strategically placed for the final stages of reclamation work. This approach makes it possible to provide the best soil at the surface of reclaimed land when levelling the area (Stys, 1981). When large horizontal coal deposits are mined in the USSR, GDR or FRG by surface mining, open pits are arranged in such a way that when each new pit is opened spoil is disposed of in the old mined-out pit. This technique provides for maximal filling of old pits and a reduction in the areas needed for external waste tips (Koliadny and Ovchinnikov, 1976). A large number of operational improvements have occurred in recent years in the USA in the surface mining of coal (Grim and Hill, 1974).

3.3.2. Improvements in site engineering technology

The degree of site engineering work required is largely determined by the desired after-use of the site. Generally, land that is to be reclaimed to arable use requires the greatest degree of site engineering.

Requirements for site engineering can most easily be met if waste and waste storage transport systems are planned in advance. Stripping that creates a "hill and dale" topography with up to 15–20 m difference in elevation requires a great volume of earth-moving to be completed (up to 20,000 $m^3 ha^{-1}$) if the surface is to be rendered suitable for ploughing. A reduction of levelling work can be achieved if the width of stripping face is reduced, or if waste storage is formed by fan techniques. This method of pile formation has been introduced at a number of open pits in the USSR

(Estonian shale, Cheremchovo coal) and is also used extensively in the USA (Ovchinnikov, 1967; Kaar and Lainoja, 1971; Grim and Hill, 1974).

The greatest experience in reclamation of crest-shaped waste storages has been accumulated in the USA, because waste is stored internally (Fig. 2.1D) without intermediate transport. The geological configurations of the coal deposits favour such methods of mining.

The technology and processes which are used in reclamation are dependent on the types of stripping and dumping machinery that are used and the way in which they are deployed. Many experts believe that draglines are the most efficient means of levelling waste surfaces. Caterpillar and wheeled dozers have also become widely accepted means of levelling surfaces in connection with both surface and deep mining coal.

Wheeled-bucket loaders of 8 m^3 or more capacity and wheeled scrapers are used on a large scale. The efficiency of scrapers in reclamation works is determined by the distance it is required to move the overburden. If the distance of earth-moving is significant, scrapers can be efficient.

Reclamation work in the USA often leaves surfaces incompletely levelled and it is claimed that this provides some biological advantages in reclamation (Riley, 1973). Surface levelling can be divided into three categories: complete, terrace-forming, and incomplete (peaks having a crest width of 2.5–5 m at the top). Complete levelling results in an even horizontal surface and is rarely undertaken in the USA. The most widespread technique is incomplete and terrace-forming levelling. This is much less costly, while the surface suits quite well the establishment of grazing lands and forests. In the climates with low precipitation (the south-western states) the surfaces of reclaimed waste storages are levelled to form closed drainless depressions which are used as reservoirs for gravity irrigation.

3.3.3. Improvements in surface soil treatment

One of the main preconditions of successful biological reclamation is a substrate for plant growth that is physically, chemically and biologically fertile. If stripping and dumping of the overburden in strip-mining operations is planned beforehand, coarse materials can be buried and overlain by fertile substrates. During stripping, toxic wastes if identified can be selectively stripped and channelled to the bottom of storage dumps.

In the GDR, at a number of open pits of the Niederlausitz brown coal basin, selective topsoil stripping is ensured by using excavators and transport bridges.

In the Rhenish brown coal basin thick loess deposits are found in the northern part of the basin, but in the southern part no appropriate material is present. A solution to this problem is found by transporting the loess by railroad for over 40 km. After delivery, the loess is discharged hydraulically to levelled land (the "loess flush system"). The loess is applied in layers. First a

20–30 cm layer is applied and is partially mixed with the waste. After this layer has dried off it acts as an impermeable stratum and prevents water from subsequent loess applications from seeping into the pile. The water is collected and recycled. The plots are drained after the last application of loess and cultivations can then take place within 6 months. In addition, trees of high economic value have been planted successfully on forest gravel—a mixture of loess and sand, gravel and crushed rock (Kausch, 1979).

3.3.4. Land-use efficiency improvements

"Land ratio" in surface mining of coal is an important consideration. It is defined as the area of land taken up or disturbed to produce 1 million tons of coal. Horizontal deposits usually result in a lower land ratio than inclined deposits, because the latter usually involve external storage of the waste (compare Figs. 2.1B and D). For this reason studies have been made to suggest designs for internal waste storage where steep and inclined seams are mined (Tomakov, 1975). If, however, external waste storage has to be deployed on fertile land, the most effective method of waste disposal is often in single large sectional banks, rather than as high multibank stores. This method results in somewhat higher land ratios, but reclaimed surfaces can be brought into agricultural production at a faster rate as single-bank storage gives a greater ratio of horizontal to sloped surface, meeting agricultural requirements better.

3.3.5. Improvement of underground mining waste reclamation

Most underground coal mines no longer tip colliery spoil to form conical tips. The most progressive form of tipping involves the establishment of a common dumping site for a group of mines utilizing motor transport and producing plateau-shaped or contoured tips. Such a procedure is now common in many coal-producing countries in Europe and North America.

Especially interesting is a method of forming large multibank waste tips (of a "step-pyramid" type) in the Ruhr. Spoil is hauled by dump trucks to the tip site. The waste is formed from the periphery to the centre of the dumping area, thus making reclamation of the perimeter possible right from the start of dumping. A pioneer retaining bank 8–10 m high is formed at the outset, this height being maintained for the first stage of tipping. The outer slope forms part of the final tip, unchanged. This waste platform is then used for the next stage of tipping and a second similar bank constructed. A distance of 4 m between the upper edge of the first bank and the bottom edge of the second is left as an access roadway. This pattern is followed until the tip reaches the desired height. Completed tips have four to six banks (terraces) with 1:2 or 1:3 outer slopes. The outer slopes are composed of fine refuse and these are seeded or planted with trees. By the time waste tipping is completed, the outer slopes are fully reclaimed. Tree growth softens the outline of the tip, but recently more consideration has been given to landscaping, and areas that are to receive deep mine spoil are now often prepared to receive it by first scraping off the fertile topsoil. The area is then used as a dumping site, the volume of tip being calculated to match the surface area to be covered by soil. Some areas that suffer from subsidence can make use of this, so that as soon as the coal is extracted the roof will cave in and the tip becomes flush to the surface of the earth.

Some systems have been developed which utilize mined-out underground voids for colliery spoil stowage. This reduces land subsidence to a minimum and removes the need to occupy land with spoil tips. Experiments have been conducted to find the best method of "back stowage". No universal approach, however, has yet been found. Moreover, if thick coal seams are mined, the back stowage material available may be insufficient in quantity and special quarries may have to be opened up to produce it. This situation exists in the Upper Silesia Coalfield in Poland. Here 90% of the coal produced underground is mined using a system of back stowage. Land subsidence is negligible. The stowage material, however, has to be produced separately from sand open pits of 40 million m^3 capacity. Reclamation of this is often achieved by filling them with colliery spoil.

The reclamation of disturbed land is not only a problem, it is a challenge as new land-use patterns may be developed and the original productivity improved. If "land capability analogues" and "waste material analogues" can be recognized, from region to region and country to country, a system can be made available to optimize land restoration practice. However, it must be admitted that some very real problems still remain. Arid or semi-arid regions present restoration problems that are only just beginning to be squarely faced. In developing countries such problems may be particularly acute. Even more conventional reclamation situations may not attract the expertise necessary for their solution. There is a real need for restoration expertise and experience to be matched with the conditions likely to prevail in potentially new centres of coal-mining activity.

3.4. Progress in Land Reclamation

3.4.1. General considerations

All reclamation will depend on such characteristics as the nature of the land disturbance, the climatic and environmental conditions prevailing, economic considerations and social requirements. For example, in regions where fertile soils occur, near densely populated areas, normal requirements for restoration will be to agriculture. Less fertile regions or industrial areas that are in need of measures against air pollution will usually be restored by the establishment of forest areas. Some reclamation may make use of flooded pits to construct reservoirs, water recreation and amenity areas.

It is held by many reclamation scientists that the selection of objectives can be made more rational and cost-effective if a classification of disturbed land types is produced showing their compatibility with reclamation objectives. Classification by spoil type, relief, rock composition and characteristic colonizing vegetation of broader application have had to be developed. In Poland six categories of disturbed lands have been identified on the basis of relief, soil and water regime characteristics.

In the USA a coding system has been developed for disturbed lands based on aerial photography. In the USSR a classification of disturbed lands has been developed based on landscape and technological features.

Although classifications based on straightforward mineralogical composition and the chemical and physical properties of wastes are insufficient alone to identify broad reclamation potential, they can be nonetheless extremely important. This is particularly true of coal waste materials in relation to iron pyrite (FeS_2) content and form, once the waste material is exposed to water and the dissolved oxygen it carries. Pyritic oxidation is responsible for toxic substrates due to low pH values (as low as 1.2–1.4) and subsequent phytotoxic levels of aluminium, and some other heavy metals. Nitrogen, phosphate and even potassium deficiency (Caruccio, 1975; Chadwick, 1973) can also occur. Acid mine drainage water causes pollution of aquatic ecosystems (Glover, 1975). Methods of dealing with these problems have been intensively studied.

3.4.2. Land disturbed by surface mining

Reclamation of land disturbed by mining for coal is now firmly established in most developed countries, on the scientific analysis of substrates, experimental determinations of amendment requirements and detailed planting and sowing regimes. As a result, land is restored to a wide range of after-uses. Scientifically and technically, it is now possible to deal with a wide range of reclamation situations in a sucessful way (Motorina, 1975). This is not to say, however, that no scientific or technical problems remain, or that the application of reclamation knowledge always results in satisfactory land restoration. One of the largest environmental disturbances resulting from the surface mining of coal in Europe is to be found in the FRG, in the Rhenish brown coal district. Resources amount to 55 × 10^9 tonnes. A 175 km^2 area has been mined in an area with a population of over 1 million and a population density of 413 inhabitants km^{-2}. About 116 km^2 have been reclaimed, and 23,700 inhabitants resettled in 55 resettlement villages (Kausch, 1979). The methods of reclamation have been described in a previous section (3.3.3).

In the GDR brown coal is mined in two main areas: Halle-Leipzig and Niederlausitz. This produces about 260 × 10^6 tons per annum. 88,900 ha have so far been mined and over half of this area restored. Reclamation is complicated in some areas by the existence of a thick stratum of sulphide-containing rock. Where fertile loess exists, this is extracted to a depth of 1 m and stored separately from the rest of the overburden. In the Niederlausitz area infertile sands and gravels overlie clays and sands with a high sulphur content (4–6 %). These clays and sands are often replaced in reclamation as the surface layer, but being acid (pH 2.8–3.5) soil improvement measures must be taken. Either lime or other neutralizing materials must be added. In the "Domsdorf" method (Knabe, 1959, 1973) applications of hygroscopic calcareous brown coal ash, from nearly power plants, are combined with fertilization and deep soil cultivation. Various modifications of the method have been worked out (Illner and Raasch, 1966). Blast furnace slag and the residue of waste-water treatment from coking plants ("Koyne" method) have also been used as ameliorants. On agricultural areas good yields of lucerne (*Medicago sativa*), rye (*Secale cereale*) and sunflower (*Helianthus annuus*) have been obtained. Poorer soils are devoted to forestry, planted initially with buckthorn (*Hippophae* spp.), alder (*Alnus* spp.) and black locust (*Robinia pseudoacacia*) as pioneer species, followed by poplar (*Populus* spp.), pines (*Pinus* spp.), birch (*Betula* spp.) and oak (*Quercus* spp.).

Reclamation in the CSSR, following the surface mining of coal in northern Czechoslovakia and the Sokolovsky region, concentrated for some years on improving the air quality conditions of industrial regions by afforestation. Red oak (*Quercus borealis*), European larch (*Larix decidua*), ash (*Fraxinus excelsior*) and field maple (*Acer campestris*), after European alder (*Alnus glutinosa*), grey alder (*A. incana*) and sea buckthorn (*Hippophae rhamnoides*), have been used as "soil improvers". In the Sokolovsky brown coal basin apples, pears, plums and cherries, as well as soft fruit, are grown on reclaimed land.

Recently, reclamation to agriculture has been favoured and research has centred on the ways in which topsoil applications can be reduced in depth from 50 to 20 cm. This has been achieved by fertilizing and growing a green manure grass–legume mixture for the first 3 or 4 years. Subsequently good yields of wheat (*Triticum aestivum*) and barley (*Hordeum vulgare*) are gained.

Reclamation of brown coal mined areas in other Eastern European countries (Hungary, Romania, Bulgaria and Poland) has also successfully produced land suitable for a range of agricultural crops and forestry.

Strip-mining for coal is practised in the UK, where the ratio of overburden to coal (by volume) is 20:1 for bituminous coal and 30:1 for anthracite. The stripping operations in the UK are now planned not only to reclaim the areas currently being exploited, but also to incorporate spoil material from old deep mines. Care is taken to remove topsoil and layers of overburden and store these separately. Care is also taken in replacement. After soil replacement cultivation is needed to restore soil structure; often high nitrogen dressings are required to restore losses from the soil dump by denitrification. Restored sites in the UK have generally given good yields of arable crops and satisfactory tree growth (Bradshaw and Chadwick, 1980).

Over 30% of coal in the USSR is produced by surface mining. It is envisaged that this will quickly rise to 40–45%. As conditions of climate, geology and soil type differ enormously over the USSR, specific approaches to reclamation have to be adapted from region to region.

In the Ukrainian SSR surface mining disturbs fertile chernozem soils that are used extensively for agriculture. Either the upper layers of the soil (the black A_1 horizon) are replaced on the surface to form the growth substrate or, in some areas, it has been found that the lower loess deposits are more fertile and these are held back for surface spreading. Usually legumes are sown first, and where these are used as green manures high yields of winter wheat (*Triticum aestivum*) and other crops are obtained (Novikova and Savitch, 1980). Considerable emphasis is given to crop rotations.

Two areas of investigation have been pursued vigorously in recent years: the screening of toxic sulphide-containing rocks with impermeable or carbonate rocks before topsoil application, and the necessary depth of topsoil under different conditions.

Restoration of land to forest and screens of trees is undertaken. In the Ukraine forty species of woody plants have been established on coal spoil, including elm (*Ulmus* spp.), black locust (*Robinia pseudoacacia*), lime (*Tilia* spp.), apple (*Malus* spp.), and species of pine (*Pinus*), maple (*Acer*), poplar (*Populus*) and birch (*Betula*).

Consideration has also been given to the reclamation of power plant ash (fly ash) and settler and sludge pond material. Generally these lack major plant nutrients (nitrogen, phosphorus and potassium) and are physically poor. Layers of soil, peat or organic wastes have been used, in conjunction with fertilizers, to improve the chemical and physical condition. Arable and pasture crops have been grown and tree species established.

In the USA, area and contour strip-mining are widely practised for coal gaining. In the areas where deposits are shallow, or there are seams of considerable width, there is less difficulty with reclamation to a satisfactory standard than where moisture is lacking, contour mining is required or high iron pyrite contents are encountered. It is estimated that 80% of the land disturbed can be reclaimed, 25% of it to agriculture. A specific feature of the reclamation activity in the USA is that the surface of the disturbed land may only be partially levelled to enhance water availability and, in some cases, reduce toxicity by the creation of a "ravine-ridge" surface topography (Riley, 1973). Negative effects of complete levelling have been demonstrated. Considerable expertise has been built up on the technological and biological aspects of reclamation and many good summaries of all aspects of this work exist (Hutnik and Davis, 1973; Grim and Hill, 1974; Schaller and Sutton, 1978; Bradshaw and Chadwick, 1980; Sendlein, Yazicigil and Carlson, 1983). In particular, the mechanization of reclamation has been developed and hydraulic sprinklers, hydraumatic seeding and the aerial application of fertilizer and seed have been extensively applied.

3.4.3. Land disturbed by underground mining

Although in many countries the percentage of coal mined by surface mining methods is increasing, current deep mine waste production still produces considerable environmental degradation to add to the many unreclaimed spoil tips produced in previous decades. Generally, these tips are to be found in densely populated regions and the possibility of their arrival with any major coal-mining development is one of the major deterrents to public acceptance of plans for coal mining in an area.

Colliery spoil heaps can be satisfactorily restored to blend into the landscape and to be biologically productive, but in almost all countries one or several factors may prevent this easily being achieved. Iron pyrite, a primary mineral associated with colliery spoil, is responsible for rendering the medium acid. Usually acidity is produced, as this is oxidized on the tip and there is seldom sufficient natural carbonate mineral, or other buffering components, present to neutralize the acidity. Spoil pH values may fall below 3. The complicated relationship between pyritic content and acid generation has been the subject of many studies, and these are well summarized by Caruccio (1975). The result of pyritic oxidation on coal tips is to produce surface layers of low pH and this in turn has a detrimental effect on spoil components that renders the spoil even less suitable for satisfactory plant growth. Low pH reduces the spoil cation exchange capacity so that beneficial ions are more easily lost by leaching; clay minerals become degraded and aluminium in solution reaches phytotoxic levels; the mineral ankerite is degraded to release high levels of available manganese; clay mineral distortion may lead to the release and loss of potassium. In addition, low pH values (accompanied by high iron and aluminium concentrations) cause phosphate fixation and reduce nitrogen availability.

In some regions, as in the Silesian Coalfield in Poland, spoil may show salinity problems due to a high content of water-soluble salts originating from ground water. Salinity can also develop from weathering reactions involving carbonate minerals in the spoil, calcium, magnesium and sodium accumulating, mostly as sulphate and chloride. In humid climates these salts are relatively easily leached from spoil material and salinity problems are fairly short-lived. This is not true for more arid climates.

Spoil materials from deep-mined coal are highly variable from region to region, tip to tip, place to place on a single tip and from time to time (Chadwick, 1974), and this must be taken into account in formulating reclamation procedures.

After earth-moving and landscaping work is completed, correction of spoil reaction is usually carried out by addition of lime—even amendments in excess of 50 tonnes ha^{-1} may be required, but care needs to be taken in order not to induce temporary phosphate unavailability due to high pH values. Most colliery spoils will require additions of nitrogen and phosphorus fertilizer, in amounts that can be determined by analysis, and then suitable plant species and cultivars must be selected. Legumes are often employed in order to contribute fixed nitrogen, although under-planting trees with these may cause a reduction in tree survival and growth. The various planting, seeding and spoil-amendment techniques suitable for colliery spoil in different parts of the world have been the subject of much research, and a summary is provided by Zaitzev, Motorina and Danyko (1977) and Bradshaw and Chadwick (1980). In particular, it is now realized how important careful attention to maintenance procedures can be, and the part site monitoring must play in this.

4. The Economics of Reclamation

As land is taken out of circulation by mining processes and the environment degraded by the wastes of industry, sectors of the economy naturally suffer. The efficiency of investments into the mining industry must be evaluated taking into account the real costs involved. The costs of environmental protection measures have to be internalized.

In all countries the cost of the earth-moving stage of reclamation makes up 70–90 % of the total costs. This is a high cost–low risk activity, whereas the cultivation, sowing and planting part of reclamation is a low cost–high risk activity. Costs vary enormously, making land reclamation costs difficult to estimate realistically even within one country (Misiolek and Noser, 1982). It is not possible to make sensible comparisons from country to country.

It is difficult to separate the economic effects of reclamation from widespread social effects. Some of the assessments published suggest that sociological factors far outweigh economic effects. A correct assessment of the importance of reclamation will only be possible when socio-economic aspects are given their proper emphasis.

Finally, the economics of the total coal production and utilization cycle need to be analysed from the standpoint of energy consumption. Such a technique would evaluate the energy balance of the total coal cycle, including reclamation.

5. References

BAUER, L. and WEINITSCHKE, H. (1973). *Landschaftsphlege und Naturschutz*. Jena.

BRADSHAW, A. D. and CHADWICK, M. J. (1980). *The Restoration of Land*. Blackwell, Oxford.

CARUCCIO, F. T. (1975). Estimating the acid potential of coal mine refuse. *The Ecology of Resource Degradation and Renewal* (ed. by M. J. Chadwick and G. T. Goodman). Blackwell, Oxford.

CHADWICK, M. J. (1973). Methods of assessment of acid colliery spoil as a medium for plant growth. *Ecology and Reclamation of Devastated Land*, vol. I (ed. by R. J. Hutnik and G. Davis). Gordon & Breach, New York.

CHADWICK, M. J. (1974). Variation in the chemical characteristics of colliery spoil and its relation to vegetational establishment. In *Grune Halden in Ruhrgebeit*, pp. 22–38. SVR, Essen.

Geping, Q. and Lee, W. (1984). *Managing the Environment in China*. Tycooly International, Dublin.

Glover, H. G. (1975). Acid and ferruginous mine drainages. *The Ecology of Resource Degradation and Renewal* (ed. by M. J. Chadwick and G. T. Goodman). Blackwell, Oxford.

Grim, E. C. and Hill, R. D. (1974). *Environmental Protection in Surface Mining of Coal*. EPA-670/2-74-013. US Environmental Protection Agency, Cincinnati, Ohio.

Hutnik, R. J. and Davis, G. (1973). *Ecology and Reclamation of Devastated Land*, vols. I and II. Gordon & Breach, New York.

Illner, K. and Raasche, H. (1966). Zur Bestimmung des Kalkbedarfs fur die Melioration von Schwefelhaltigen Tertiarkippen. *Zeitschrift fur Landeskultur*, **7**, 285–90.

Kaar, E. and Lainoja, L. (1971). *Polevkivikarjaaride Rekultivfeerimine*. Tallinn.

Kausch, P. (1979). Brown coal opencast mining and landscaping in Rhineland. *Energy Research*, **3**, 275–88.

Knabe, W. (1959). Vorlaufige Richtlinien fur die Bepflanzung und Melioration forstlicher Kippenstandorte in der Niederlausitz. *Forst. und Jagd.*, **9**, 229–34.

Knabe, W. (1973). Development and application of the "Domsdorf ameliorative treatment" on toxic spoil banks of lignite opencast mines in Germany. *Ecology and Reclamation of Devastated Land*, vol. II (ed. by R. J. Hutnik and G. Davis). Gordon & Breach, New York.

Koliadny, M. F. and Ovchinnikov, V. A. (1976). *Rekultivatsia Narusiennich Ziemli v FRG*. Moscow (Russian).

Misiolek, W. S. and Noser, T. C. (1982). Coal surface mine land reclamation costs. *Land Economics*, **58**, 67–85.

Motorina, L. V. (1975). *Experience of Reclamation of Land Disturbed by Mining in the USSR and Abroad*. Moscow.

Motorina, L. V. and Ovchinnikov, V. A. (1975). *Industry and the Recultivation of Land*. M'isl, Moscow (Russian).

Motorina, L. V. and Zabelina, N. M. (1968). *The Recultivation of Land Damaged by the Mining Industry—A Review of the Literature*. All Union Scientific and Technical Institute for Agriculture, Moscow (Russian).

Novikova, N. A. and Savitch, A. I. (1980). *Vosstavnovlienie Potivennovo Plodorodia na Otvaliach Otkrutuch Razrabotok pri Biologitieskoy Rekultivatsii*. Moscow.

Ovchinnikov, V. A. (1967). The interrelationship between natural and technical mineralogical (mining) factors in the recultivation of land damaged by opencast mining operations. In *The Restoration of Land Following Industrial Operations*. M'isl, Moscow (Russian).

Riley, C. V. (1973). Chemical alterations of strip-mine spoil by furrow grading—revegetation success. *Ecology and Reclamation of Devastated Land*, vol. II (ed. by R. J. Hutnik and G. Davis). Gordon & Breach, New York:

Schaller, F. W. and Sutton, P. (1978). *Reclamation of Drastically Disturbed Lands*. American Society of Agronomy, Madison, Wisconsin.

Sendlein, LK. V. A., Yazicigil, H. and Carlson, C. L. (1983). *Surface Mining Environmental Monitoring and Reclamation Handbook*. Elsevier, New York.

Stys, S. (1981). *Rekultivace tezenu Postizenych Tezbou Nerostnych Surovin*. SNTL, Prague.

Tomakov, P. I. (1975). Nekatorie napravlienia po snizieniyu otritzatelnovo vliyania otkrutruch razabotok na okruziaynsyuy sredu. In *Teoretitieskie i Praktitieskie Problemy Rekultivatsii Narusienych Ziemli*. Moscow.

Zaitzev, G. A., Motorina, L. V. and Danyko, V. K. (1977). *Land Reclamation for Forestry*. M'isl, Moscow (Russian).

3

Resource Demands of Coal Production

1. Introduction 47
2. Resource Requirements 48
3. Water Needs and Coal Development 50
    3.1. Future Coal Technologies 50
    3.2. The Geography of Coal 52
        3.2.1. USSR 52
        3.2.2. USA 54
    3.3. Regional Coal Scenarios 55
    3.4. A Water-conscious Future 57
    3.5. Water Requirement Model 58
        3.5.1. Surface mining 58
        3.5.2. Underground mining 59
        3.5.3. Coal preparation 60
        3.5.4. Slurry pipelines 60
        3.5.5. Flue gas desulphurization 60
        3.5.6. Power plants 61
        3.5.7. Liquefaction 64
        3.5.8. Constants for the equations 65
    3.6. Water Requirements 65
    3.7. Water Balance 67
    3.8. Conclusions 71
4. References 71

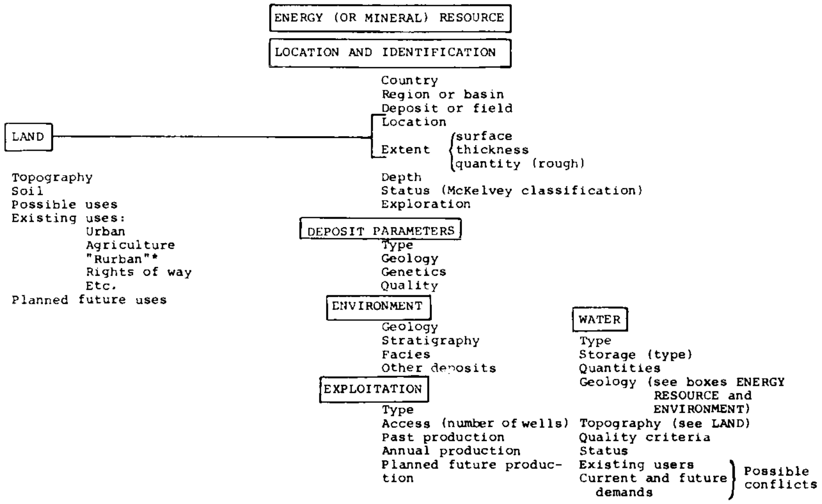
1. Introduction

The impacts of coal mining may be regarded as a chain reaction where the initial effect of coal extraction, in turn, produces other impacts which pass through the reaction chain like a series of impulses. As the impulses pass through the chain, their influence on the environment decreases. For example, the initial effect of underground mining is the deposition of waste on the surface above the mine and subsidence and caving, causing land degradation at the surface. This usually adversely affects water courses, vegetation, animal communities and man-made structures. Drainage patterns will be changed and different vegetation types develop; disposal of mine drainage water will alter or degrade aquatic biota; solid waste will produce new substrates to be colonized; but also run-off and particulate loading of the atmosphere may cause increased heavy metal and other elemental dispersion to surrounding biota as absorption by plants and migration via food webs occur. Ventilation emissions from the mine will give localized atmospheric alteration. The initial impacts in this chain of events, land degradation and disturbance, have been discussed in Chapter 2. In some countries, like the United Kingdom, these are assessed as being the major impacts on the environment of whole coal cycle (Commission on Energy and the Environment, 1981).

However, land and the structures and communities it bears are not the only resource to be utilized in the process of coal mining. Water (W), energy (E), materials (M) and manpower (M), as well as land (L), WELMM in aggregate, are the total natural resources consumed in making available fresh resources of fuel (Grenon, 1979) as in coal mining, coal conversion and further use.

2. Resource Requirements

To assess the range of natural (and other) resources required to achieve access to a desired quantity of an energy resource like coal, it is necessary to evaluate the factors outlined in Fig. 3.1. Not all will be of major significance in any one development. The variation that may occur can be gauged by reference to Tables 3.1 and 3.2 concerning mine and environmental characteristics relating to three current coal mining developments at Selby in the United Kingdom, Garsdorf in the Rhine region of the Federal Republic of Germany and Burnham, New Mexico, USA. These three mine developments differ considerably in the characteristics of the resource to be gained (the coal) and in the environmental and other resources that will be called upon to gain this resource.



*New types of human settlements in rural areas

FIG. 3.1. Resource information in relation to coal mining, conversion and further utilization (Grenon, 1979).

TABLE 3.1. *Main mine characteristics of three coal mining developments (after Grenon, 1979)*

Location	Selby, North Yorkshire, UK	Garsdorf, Rhine area, FRG	Burnham, New Mexico, USA
Coal characteristics	Bituminous	Lignite	Sub-bituminous
Calorific value (kJ kg^{-1})	35,000	7950–8790	9625
Sulphur content (%)	1.4	low	0.72
Ash content (%)	4	8 to 8	22
Moisture content (%)	2.5–8	50 to 60	17
Deposit characteristics			
Estimated resources (Mt)	600+	1000	615
Seam thickness (m)	2.75–3.35	45–60	3.50 (in 4 seams)
Depth (m)	about 760	up to 300	up to 50
Type of mining	Underground	Open pit	Stripping
Main equipment	Long wall	Bucket wheel excavator	Dragline and power shovel
Annual production (10^6 t yr^{-1})	10 plus around 1988–9	42 in 1974 50 forecasted	maximum 17 in 1982+
Status	Started in 1981	Started mid-1960s	Planned (1980s)

TABLE 3.2. *Some environmental characteristics of the Selby, Garsdorf and Burnham Mines (Grenon, 1979)*

Location	Selby, North Yorkshire, UK	Garsdorf, Rhine area, FRG	Burnham, New Mexico, USA
Total area (km^2)	390		162
Area mined (km^2)	290	25–26	89
Type	Very rich farmland Rural	Very rich farmland High population density Large cities nearby	Poor grazeland Scarce population Indian reservation
Main problems	Impact on traditional rural area Subsidence of agricultural land	Deepest open pit water table Resettlement of communities	Uncertainty of reclamation potential

Another approach within the WELMM framework might be to assess the total water (or other resource) requirements for coal use in a region. Water resources are required at every stage of coal development, from mining to its final conversion to useful energy (Alcamo, 1984). Even with a high degree of recycling, 1 tonne of coal equivalent (tce), as coal-fuel delivered, requires 1–2 tonnes of water under conditions encountered in the USA and USSR. Indeed, some studies have suggested that water availability may be the limiting factor in the development of coal resources (Harte, 1979; Harte and El-Gasseir, 1978).

3. Water Needs and Coal Development

3.1. Future Coal Technologies

Alcamo (1984) has recently attempted a comparative evaluation of the quantity of water required to expand coal production to 2.9×10^9 tce in North America (Canada and the USA) and to 3.8×10^9 tce by the socialist countries of Eastern Europe by 2030. Such increases represent five-fold and four-fold expansions respectively. The methodology for such an evaluation is outlined in Fig. 3.2. Having decided on the coal scenario, the model is used to evaluate the water needed for such a development and this is then compared with an estimate of the water available.

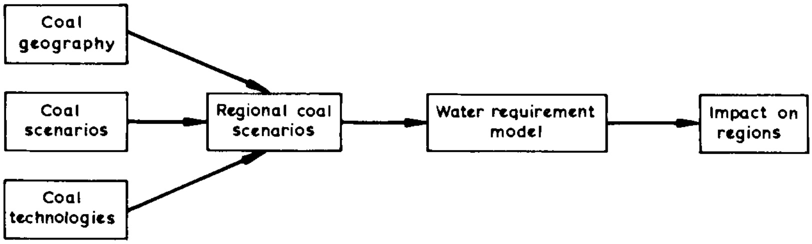


FIG. 3.2. Analytical approach to determining the impact of coal development on water resources.

Obviously the water requirements of future coal development in the Soviet Union and the USA will depend on the kind of technology that will be used in the future coal industry. Unfortunately, it is not so obvious what these technologies will be. It becomes easier to speculate once one realizes that the "lifetime" of a mine is of the order of 30 years or longer, as is the lifetime of a well-designed and maintained power plant. It follows that much of the coal infrastructure that will exist in the year 2030 will have been designed or even constructed before the end of this century. With this in mind, it seems appropriate to select only those technologies that are currently in use or are at an advanced stage of development.

Alcamo (1984) assumed coal use for conversion to liquid fuel, but not to gas, as the scenario he used assumed that natural gas was sufficient to meet near-term gas demand in the USA and the USSR. He also assumed that much of the coal would be used for metallurgical processes.

Figure 3.3 shows that in order to meet a "demand" a path or "chain" through each of the six major coal sectors—(1) mining; (2) local transport;

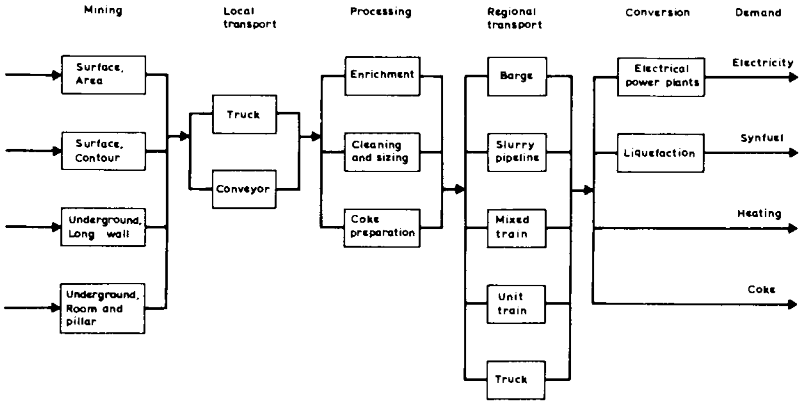


FIG. 3.3. Selection of technologies for the coal industry in the future.

(3) processing; (4) regional transport; (5) conversion; (6) demand—must be followed. As Fig. 3.3 notes, there are several possible technologies for each of these sectors.

1. Two types of mining are distinguished: surface and underground. Underground mining is, in turn, subdivided into two categories, long-wall and room and pillar. The latter is by far the most common type of underground mining in the USA whereas long-wall is the predominant method used in Europe and the USSR. Hydraulic mining is being discussed as an alternative to long-wall mining in the USSR, but its future share of total underground mining is still unclear (Gontov, 1979; Astakhov, 1979). Alcamo (1984) assumed, therefore, that long-wall mining will continue to be the principal form of underground mining in the USSR.

2. "Local transport" refers to the movement of coal between mining and processing centres, which are often in close proximity. Two forms of local transport are included—truck and conveyor.

3. Three alternatives are specified for coal processing: (a) enrichment facilities for low-grade coals destined for power plants; (b) cleaning and sizing facilities for higher-grade coals used in power plants or for residential or commercial heating; and (c) coke preparation for coking and other industrial coals.

4. "Regional transport" represents the distribution of coal from processing centres to either conversion facilities or demand centres. Five different transport modes are selected: barge, slurry pipeline, mixed train, unit train and trucks. Barges are used in the Appalachian and Central coal regions of the USA but not in the Soviet Union. Slurry pipelines are considered a future transportation alternative, even though they are considered speculative by some. It seemed reasonable to include them in this model, however, because

one is already in operation in the Four Corners coal region of the southwestern USA. Slurry pipelines have been included in projections of future coal use in the USA (WOCOL, 1980). There also has been some discussion about their future use in the USSR (Baibakov *et al.*, 1979), though Alcamo (1984) assumed no slurry pipeline transport for the USSR. A "mixed train" was used to refer to a train that carries non-coal cargo in addition to coal. A "unit train" carries only coal. Both types are currently used in the USSR and the USA. Trucks are currently used for short-distance haulage to conversion facilities or to other transportation modes in the Appalachian coalfields of the USA.

5. Figure 3.3 specifies two possibilities for coal conversion—electrical power plants and liquefaction. Power plants are assumed to be of the conventional combustion type. Liquefaction plants are assumed to use the synthoil process, which is used by Probstein and Gold (1978) to project water requirements of the future synfuels industry in the USA. According to Probstein and Gold (1978), synthoil consumes about the same amount of water as other feasible hydrogenation processes.

6. The last sector, demand, specifies four possible forms of energy from coal—electricity, synfuel, heating (which includes electrical production via cogeneration) and coke (which includes all industrial uses of coal, including feedstocks).

3.2. The Geography of Coal

To proceed with the model, the patterns and distribution of coal production and use in the two countries must be assessed. This was termed the "geography of coal" by Alcamo (1984).

3.2.1. USSR

A key feature of the coal geography of the USSR is the shift that is currently taking place in the location of coal production. Currently, over three-quarters of Soviet energy consumers live on one-quarter of its territory in the European part of the USSR. These energy consumers also use over three-quarters of the Soviet Union's total energy, and by the year 2000 it is unlikely that they will use less than 65–70 % of the Soviet Union's energy. Much of this energy is provided by coal from a few European coal regions. By far the largest producer is the Donetsk Region, whose output overshadows all other coalfields in the USSR, as noted in Fig. 3.4. High-quality coal from the Donetsk Region's underground mines satisfies much of the coking-coal needs of European–USSR industry, as well as its heating and power production requirements. However, there are other important sources of high-quality coal, such as the Asian–USSR coalfields in Kuznetsk and Karaganda. In addition, lower-quality brown and hard coal is extracted cheaply from surface

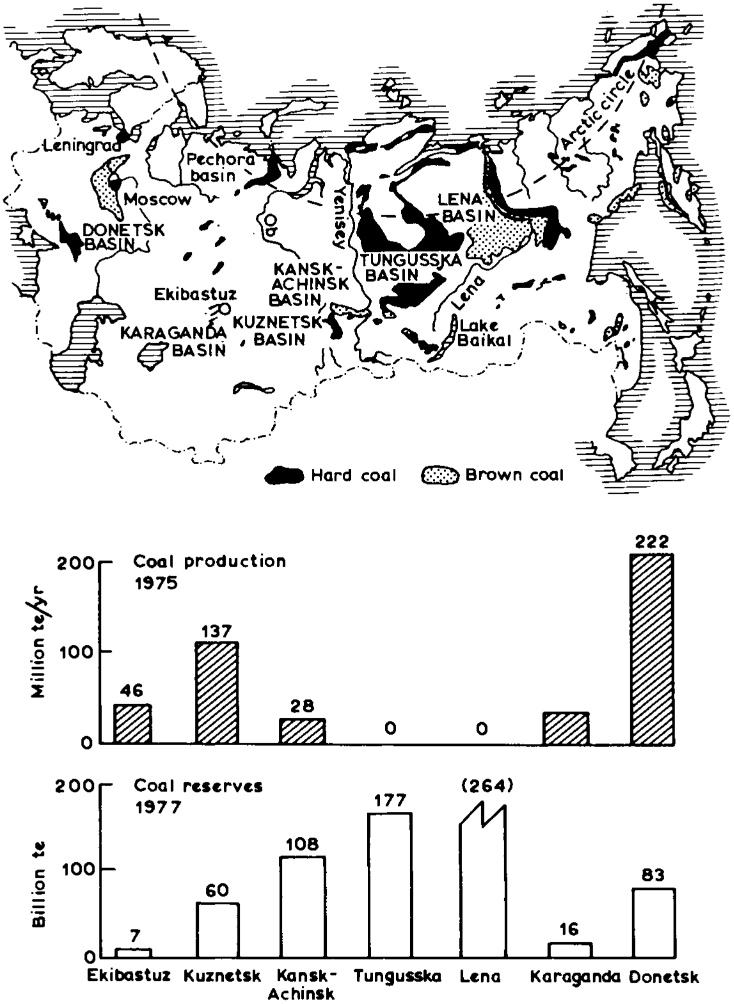


FIG. 3.4. Coal production and reserves in the USSR. (Sources: Coal production data from Shelest (1979). Coal reserves data are low estimates of known/identified reserves cited by Astakhov (1977). See text for explanation of estimates from Tungusska and Lena).

mines in other Asian fields such as Ekibastuz and the vast coalfields of Kansk-Achinsk where the lignite coal is in seams up to 100 m thick. Unfortunately, the lower value of this coal, and its tendency to spontaneous combustion when moved far, makes it uneconomic to transport it to demand centres in the west of the country. It is therefore necessary to convert it to useful products. An example of this is the current plan to construct four huge electricity generating stations in Ekibastuz, each possessing eight power plants with a capacity of

500 MW. 40 % of this electricity will be transmitted 2400 km to the European power grid.

Unfortunately for the coal industry of the USSR, however, the output of the Donetsk Region is static and may soon even decline. The root of the problem is the increasingly difficult mining conditions—one-third of the mines are already worked down to depths of 1.2 km or deeper (Astakhov, 1979) and they become 12 m deeper each year (Astakhov, 1977). Deepening mines result in more difficult working conditions, as well as increasing technical problems. In addition, some entire coalfields in the Donetsk Region have been exhausted and few undeveloped fields remain. All this adds up to increasing costs and lack of possibilities to increase production.

If the Donetsk Region cannot meet the possible four-fold expansion in production, then where will this coal come from? It is clear from the diagram of coal reserves in the Soviet Union (Fig. 3.4) that it will have to come mostly from the rich reserves such as the Siberian fields of Lena and Tungusska, which are not only undeveloped but virtually unexplored. However, the reserve estimates for these fields, shown in Fig. 3.4, represent a possibly optimistic 10% of total resources (Fettweis, 1979). But if this estimate is correct, then an enormous quantity of coal lies in these fields.

In summary, it is clear from Fig. 3.4 that production by the year 2030 will have to shift to the eastern part of the USSR. Furthermore, much of this coal will probably be converted to useful products near the coal mines and then sent to the European part of the USSR. However, the possibility of transporting at least part of the coal instead cannot be discounted, and this would switch some of the water use requirements to other parts of the USSR.

3.2.2. USA

A locational shift of the same magnitude is also occurring in coal regions of the USA. Figure 3.5 shows that much of current production of the USA originates in the Appalachian coalfields. High-quality coal from these fields is used in the industrial east, while Central region high-quality coals serve much of the industrial corridor in the vicinity of Detroit and Chicago (Fig. 3.5). Production of the mostly lower-quality coal in the west is now substantially lower than the sum of Appalachian and Central coal production, but, as Fig. 3.5 notes, the USA coal reserves are largely in the west, where the coal can also be extracted rather easily in surface mines. Other constraints notwithstanding, it is likely that future production will shift from east to west in the USA.

These major geographic shifts set the "tone" for future coal development in the USSR and the USA, and provide a departure point for the construction of the regional coal scenarios.

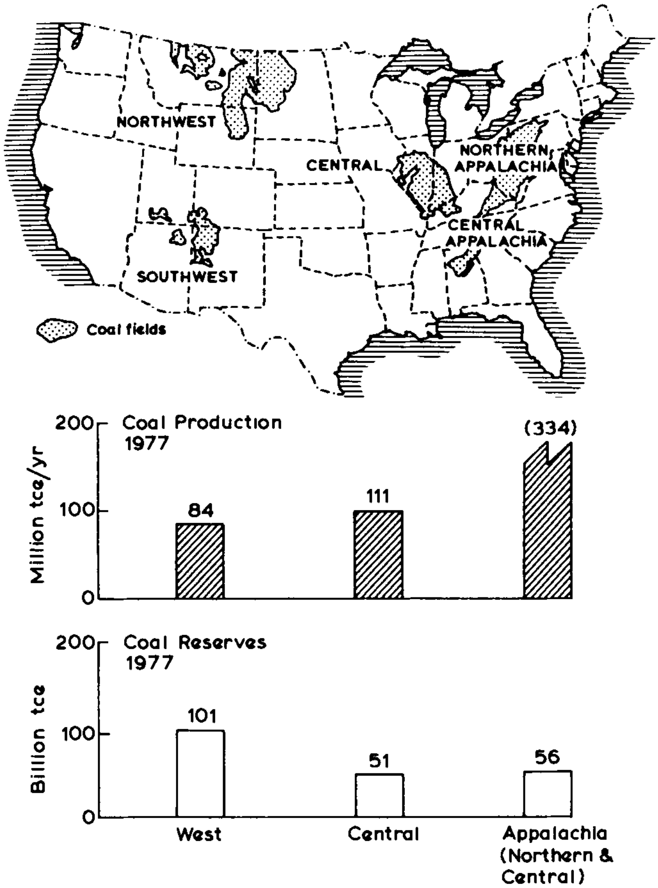


FIG. 3.5. Coal production and reserves in the US (Sources: Coal production data adapted from OTA (1979) using average coal heat values from Hittman (1974). Coal reserves data from Conaes (1977).)

3.3. *Regional Coal Scenarios*

Knowledge of the current and future coal geography may be combined with the future coal technologies selected in order to devise regional scenarios for future coal development in the USSR and the USA. Alcamo (1984) clarifies the term "regional scenario" as follows: each scenario consists of the following information for each coal-producing region:

- total coal production;
- quantity of different coal "products" (liquid fuels, electricity, heating and industrial coal);

- type of coal technologies employed with each region (surface or underground mining);
- coal characteristics.

The main guidelines used in constructing these scenarios were as follows. First, upper limits were set on total coal production in a region when this information was available. In addition, a rough upper limit of about 6 % yr^{-1} was set on the rate of expansion of coal production for any one region. Recent expansion of coal production in Ekibastuz demonstrates that this is an achievable upper limit for at least a short period of time. Between 1975 and 1979 production was reported to have increased by 31 %, which is equivalent to a 5.5 % yr^{-1} expansion for that 5-year period (Shabad, 1980). But this rate of expansion has also been sustained for a much longer period of time in the Kuznetsk Basin, where production increased from 21 to 134 million t yr^{-1} between 1940 and 1975, an annual rate of expansion of 5.4 % yr^{-1} (Astakhov, 1977).

Secondly, the quality of coal in a particular coal region was matched with the "type" of coal products needed. For example, the Kuznetsk Region possesses over 50 % of the USSR's coking coal reserves (Lelyukhina, 1973). This region was therefore allocated most of the country's total coke production. The proximity of the coal region to potential consumers is also an important factor. In the case of Ekibastuz, for example, it would be uneconomic to transport low-quality coal thousands of kilometres to European demand centres for residential heating or industrial use, but it may be economic to convert it first to electricity, as is currently planned. Therefore, a significant amount of the future coal–electricity requirement for the USSR was assigned to Ekibastuz.

The third guideline is that regional scenarios were based wherever possible on existing authoritative forecasts. At least two such forecasts exist for the USA, one from the World Coal Study (WOCOL, 1980) and the other from the Office of Technology Assessment (OTA, 1979). Both estimate the same magnitude of future coal production in the USA as the IIASA low scenario (Häfele, 1981). The OTA figures were used by Alcamo (1984) because they were available on a state-by-state basis. Regional scenarios were unavailable for the USSR.

Finally, current plans to expand coal production, such as those to construct four power plant complexes at Ekibastuz over the next 10 years, were incorporated into the scenarios.

Following these guidelines, the low and high regional scenarios presented in Table 3.3 were constructed. The specific assumptions behind these results were presented in the original study carried out by Alcamo (1983).

An important question raised by these scenarios is: "Do these regions have adequate reserves to reach such levels of production?" If an exponential growth in production from 1975 to 2030 is assumed, the Ekibastuz will

TABLE 3.3. *Low and high regional coal scenarios for the year 2030 (coal production in 10^8 tce yr^{-1})*

Region	Low scenario	High scenario
Soviet Union[a]		
(1) Ekibastuz	0.1	0.1
(2) Kuznetsk	0.5	1.0
(3) Kansk-Achinsk	0.4	1.0
(4) Donetsk	0.2	0.2
(5) Tungusska	0	1.0
United States[a]		
(1) South-west	0.15	0.27
(2) North-west	0.41	0.75
(3) Central	0.23	0.43
(4) Northern Appalachia	0.26	0.48
(5) Central Appalachia	0.32	0.58

[a] See Figs. 3.4 and 3.5 for location of the regions.

consume roughly three-quarters of its reserves by 2030 under both high and low scenario assumptions. Similarly, the Kuznetsk coalfield will deplete 50% of reserves and Appalachia 60% of reserves by the year 2030 under the high scenario. It follows that coal mining will be very expensive in these regions by the year 2030. It is also important to note that reduced production levels in these areas would only shift the supply burden to the reserves of other regions.

Another important finding from these regional scenarios is that existing Soviet coalfields could probably meet the requirements of the low scenario, but it may be necessary to open up entirely new coalfields (or import coal) in order to meet the production targets of the high scenario. In order to meet these additional requirements domestically, the choice seems to be between two largely undeveloped Siberian coalfields, the Lena and Tungusska. Tungusska was selected by Alcamo (1984) because it is located further south from the Arctic Circle than the Lena Basin and may, therefore, have better climatic conditions. It also possesses higher-grade coal (Astakhov, 1977).

3.4. *A Water-conscious Future*

Having estimated the quantity and location of future coal production and processing, and the methods by which this coal will be extracted, transported and processed, Alcamo (1984) computed the industry's water requirements. He assumed a "water-conscious" future in which planners would be aware of regional or local constraints on water use and therefore have the economic and political incentive to maximize water conservation. It follows that all wastewater streams will be treated and recycled, and that other strict conservation measures will be taken in water-short areas. But is this an economically reasonable assumption? One set of investigators (Probstein and Gold, 1978) maintain that the cost of water treatment to provide recycled water in a synthetic fuels complex will probably not exceed 5 % of the final cost of the product. Maximum water conservation may therefore be affordable. This water-conscious future also influences the selection of power plant and liquefaction cooling techniques, which are among the most significant water consumers in the coal industry (Harte and El-Gasseir, 1978).

Assuming maximum water recycling, Alcamo (1984) listed ways in which water would be consumed (Table 3.4). These include water lost by evaporation during dust control in mining, processing and conversion facilities. Water is also evaporated during the cooling processes of liquefaction and power plants, and is the "basic ingredient" of pipeline slurry and various process streams in liquefaction plants. Theoretically it can be reclaimed from pipeline slurry. Alcamo (1984) assumed that it would be uneconomic to do so.

Water is lost in the disposal of sludges originating from ash residues of liquefaction plants and scrubber equipment of pollution control devices. As with the pipeline slurry, it was assumed that water associated with these sludges could not be economically recovered. Water is also lost with the flow of "scrubbed" air in pollution control devices.

To compute the amount of water required for each of these uses, an analytical tool or "model" was developed. Equations in the model were based mostly on the work carried out at the Massachusetts Institute of Technology and Water Purification Associates (Probstein and Gold, 1978; Gold *et al.*, 1977).

TABLE 3.4. *Water requirements of various stages of coal processing*

Mining	Conversion (power plants, liquefaction)
–Dust control	–Process water
–Revegetation	–Cooling water
	–Pollution control
Preparation	–Ash disposal
–Dust control	–Dust control
Transport	
–Slurry water	

3.5. Water Requirement Model

3.5.1. Surface mining

For surface mining the only significant water loss was assumed to occur through evaporation during fugitive dust control of roads at the mining site. As in Probstein and Gold (1978), this loss was assumed to be significant only in regions where the potential evaporation rate clearly exceeded the precipitation rate. In rainy regions fugitive dust was assumed to be controlled by the rain itself. The amount of evaporated water per year was found simply by multiplying the annual potential evaporation rate by the area that is wetted down (i.e. the road area). It was also assumed that the road area is equal to 12% of the total mine area; it follows that the amount of evaporated water at surface mines will be:

$$W_{11} = (b/e)\,(y_{11}a_{11} + y_{12}a_{12}), \tag{1}$$

where W_{11} = water requirement of surface mining ($m^3\ yr^{-1}$),
b = annual potential evaporation rate (cm) to prevent double counting of temporal units (b is input in units of cm in this equation),
e = coalfield yield ($t\ ha^{-1}$),
y_{11} = coal input, area mining ($t\ yr^{-1}$),
a_{11} = efficiency, area mining (fraction),
y_{12} = coal input, contour mining ($t\ yr^{-1}$),
a_{12} = efficiency, contour mining (fraction).

In developing equations for the water requirement model, the following convention was used to denote the variables: water requirement is assigned the variable W, the amount of coal input to a process y, and the energy equivalent of this coal x. A double-digit subscript is assigned to each variable; the first refers to the major coal development sectors noted in Fig. 3.3, the second to a particular process in one of these sectors. For example, in Fig. 3.3 the first sector is mining and the first process in this sector is surface-area mining. Therefore, a variable referring to surface-area mining would have a subscript 11. The water requirement of a surface mine is thus denoted W_{11}.

3.5.2. *Underground mining*

As in surface mines, dust control was also assumed to be the principal water consumer in underground mining. Probstein and Gold (1978) give the quantity of water used in Appalachian underground mines as 100–300 gall min^{-1}, or roughly 33–100 pounds (lb) water per 1000 lb of coal. This range reflects different levels of water availability and management in the mines. For the water requirement model, an intermediate value of 67 lb water per 1000 lb coal was selected; since this is equivalent to 0.067 m^3 water per ton of coal, the simple expression is obtained

$$W_{13} = 0.067\,(y_{13}a_{13} + y_{14}a_{14}), \tag{2}$$

where W_{13} = water requirement of underground mining ($m^3\ yr^{-1}$),
y_{13} = coal input, long-wall mining ($t\ yr^{-1}$),
a_{13} = efficiency, long-wall mining (fraction),

y_{14} = coal input, room and pillar mining (t yr^{-1}),
a_{14} = efficiency, room and pillar mining (fraction).

3.5.3. *Coal preparation*

The various processes described in Fig. 3.3 as "cleaning and sizing" include breaking, conveying, screening, crushing and other standard procedures. Washing is not included. Probstein and Gold (1978) note that in coal preparation most water is used in dust control at transfer points such as surge bins, storage sites, etc. The amount of water used for this purpose in US mines is 10–15 lb per 1000 lb of coal. Using an intermediate value of 12.5 lb water per 1000 lb coal:

$$W_{32} = 0.0125\, y_{32} a_{32}, \tag{3}$$

where W_{32} = water requirement of coal preparation (m^3 yr^{-1}),
y_{32} = coal input, coal preparation (t yr^{-1}),
a_{32} = efficiency, coal preparation (fraction).

3.5.4. *Slurry pipelines*

The water consumed in slurry pipelines was assumed to be simply the water used for slurry make-up:

$$W_{42} = f y_{42} \tag{4}$$

where W_{42} = water requirement of slurry pipelines (m^3 yr^{-1}),
f = water/coal ratio (t water t^{-1} coal),
y_{42} = coal input, slurry pipeline (t yr^{-1}).

3.5.5. *Flue gas desulphurization*

The water requirement of only one pollution control device was included in the water requirement model. This was the flue gas desulphurization unit (FGD) that is used to control sulphur emissions. There are two principal ways in which water is lost in these devices: (1) with the scrubbed flue gas; (2) in the water used to dispose of the spent scrubber sludge. For losses of the first type, Probstein and Gold (1978) present the following equations based on mass balance and stochiometric considerations:

$$\frac{\text{moles flue gas}}{\text{lb coal}} = 4.76\,(1 + a')\left(\frac{c}{12} + \frac{s}{32}\right) + (3.76 + 4.76a)\left(\frac{h}{4} - \frac{x}{32}\right) \tag{5}$$

and

$$\frac{\text{lb water}}{\text{lb coal}} = \left(\frac{\text{moles flue gas}}{\text{lb coal}}\right)\left(\frac{\text{moles water vapour}}{\text{moies dry flue gas}}\right)18 - w - \frac{h}{4}, \tag{6}$$

where a' = excess air fraction (wt. fraction),
c = carbon content of coal (wt. fraction),
s = sulphur content of coal (wt. fraction),
x = oxygen content of coal (wt. fraction),
h = hydrogen content of coal (wt. fraction),
w = moisture content of coal (wt. fraction).

If an average experimental value of 0.13 is assigned to moles water vapour/moles dry flue gas, and a value of 0.15, as Probstein and Gold (1978) suggest, and we combine equations (5) and (6), then:

$$WF_1 = 1.07c + 0.4s + 2.51\,h - 0.33x - w, \tag{7}$$

where WF_1 = water lost in FGD unit with scrubbed flue gas (t water t^{-1} coal).

Other variables are defined above.

The water required for ash disposal in the FGD unit is a function of the amount of sulphur removed from the flue gas, and can be expressed as

$$\frac{\text{lb make-up water}}{\text{lb sulphur}} = \frac{\text{lb ash solid}}{\text{lb sulphur}}\left(\frac{1-m}{m}\right), \tag{8}$$

where m = solid concentration in scrubber sludge.

We can assign a value of 40% to the solid concentration in the scrubber sludge (m) and 5.9 to the ash solid/sulphur ratio (lb ash solid/lb sulphur). Using these values in equation (8) yields

$$WF_2 = 8.85s, \tag{9}$$

where WF_2 = water lost in FGD scrubber sludge (t water t^{-1} coal),
s = sulphur content of coal (wt. fraction).

3.5.6. Power plants

Water is consumed in coal-fired power plants in two major ways: (1) in cooling processes; (2) in pollution control equipment. The computation of cooling water requirements is rather complicated, involving assumptions of the type of cooling process used, efficiency of the process selected, and many other variables. Since these computations were outside the scope of the water requirement model, a "black-box" approach was taken by Alcamo (1984) to compute the amount of water used. The water requirement is given simply by

$$W_{51a} = gX_{51}, \tag{10}$$

where W_{51a} = water requirement for power plant cooling (m^3 yr^{-1}),
g = cooling water required per energy input (m^3 10^{15} J),
X_{51} = energy equivalent of coal input (10^{15} J yr^{-1}).

Assumptions of cooling mode, efficiency, etc., are built into the parameter g. The selection of values for g is discussed in the footnotes of Table 3.5.

TABLE 3.5. *Constants in the water requirement model*

Parameter	Soviet Union[a]					United States[a]				
	(1)	(2)	(3)	(4)	(5)	(1)	(2)	(3)	(4)	(5)
Evaporation rate (cm yr^{-1})[b]	70	55	55	80	NA	125	102	NA	NA	NA
Surface mining yield (t ha^{-1})[c]	79,000	79,000	79,000	79,000	NA	67,000	116,000	NA	NA	NA
Water-coal slurry ratio (t water t^{-1} coal)[d]	NA	NA	NA	NA	NA	1	1	1	1	1
Power plant cooling water (m^3 water 10^{-15} J input)[e]	152,000	152,000	152,000	152,000	152,000	152,000	152,000	152,000	190,000	190,000
Liquefaction process water (m^3 water 10^{-15} J input)[f]	8000	8000	4000	9000	6000	5594	3996	6660	9590	9590
Liquefaction cooling water (m^3 water 10^{-15} J input)[g]	19,000	19,000	19,000	19,000	19,000	18,648	18,648	42,624	41,292	41,292
Coal characteristics (wt. fraction)										
Ash	0.370^{h}	0.109^{h}	0.104^{h}	0.225^{h}	0.100^{l}	0.157^{k}	0.068^{k}	0.089^{k}	0.147^{k}	0.112^{k}
Carbon	0.450^{j}	0.650^{j}	0.450^{j}	0.600^{j}	0.600^{l}	0.570^{j}	0.458^{l}	0.591^{j}	0.693^{j}	0.736^{l}
Sulphur	0.007^{h}	0.004^{j}	0.005^{h}	0.028^{h}	0.030^{l}	0.006^{k}	0.009^{k}	0.029^{k}	0.031^{k}	0.009^{k}
Hydrogen	0.050^{j}	0.040^{j}	0.040^{j}	0.040^{j}	0.040^{l}	0.036^{l}	0.034^{l}	0.041^{l}	0.049^{l}	0.049^{l}
Oxygen	0.040^{j}	0.100^{j}	0.050^{j}	0.060^{j}	0.060^{l}	0.093^{l}	0.113^{l}	0.083^{l}	0.053^{l}	0.053^{l}
Moisture	0.080^{h}	0.094^{j}	0.348^{h}	0.040^{j}	0.160^{l}	0.124^{l}	0.304^{l}	0.161^{l}	0.023^{l}	0.023^{l}
Coal heating value (kcal kg^{-1})	4250^{m}	6150^{m}	3560^{m}	6000^{m}	6000^{l}	5457^{k}	4878^{k}	5890^{k}	6557^{k}	6723^{k}

[a] Numbers refer to coal-producing regions: *USSR*: (1) Ekibastuz, (2) Kuznetsk, (3) Kansk-Achinsk, (4) Donetsk, (5) Tungusska. *USA*: (1) south-west, (2) north-west, (3) central, (4) northern Appalachia, (5) central Appalachia.

[b] Potential evaporation: data for USSR from UNESCO (1978), interpolated from sheet 18. Data for USA based on range of 'open surface evaporation' presented in Probstein and Gold (1978). NA (not applicable) indicates that average annual potential evaporation does not exceed average annual precipitation. It is assumed in these cases that water is not consumed by dust control in surface mines.

[c] Estimate for the USSR represents an average national figure derived as follows from Mel'nikov (1979): Overburden ratio, 1975 USSR average for surface mines 3.8 $m^3 t^{-1}$. Typical seam + overburden thickness $\simeq$ 30 m. Therefore, "average" coalfield yield $\simeq$ (30 m 3.8 $m^{-3} t^{-1}$) × 10,000 $m^2 ha^{-1}$ $\simeq$ 79,000 t ha^{-1}. US estimates from Probstein and Gold (1978).

[d] Assuming no slurry pipelines in the USSR, US data assume slurry mixture 50% coal, 50% water (after Probstein and Gold, 1978).

[e] Gold *et al.* (1977) present economically optimal water requirements for six proposed western US coal-fired power plants. From their calculations we derive 0.4 km^3 10^{-18} J output, which is also the lower range of wet-cooling tower water requirements presented by Harte and El-Gasseir (1978). It was therefore assumed that this represents a reasonable water requirement for water-scarce areas. Since the water requirement model calls for "water required per energy input" to the plant, 0.4 km^3 10^{-18} J output was converted assuming 38% plant efficiency: 0.4 × 0.38 = 0.152 km^3 10^{-18} J input. For plants in water-plentiful areas, an intermediate value for wet-cooling tower water requirements was used (Harte and El-Gasseir 1978): 0.5 km^3 10^{-18} J output, which is equivalent (at 38% plant efficiency) to 0.19 km^3 10^{-18} J input. Power plants in the USSR were assigned "water-scarce" values.

[f] US data were derived from Probstein and Gold (1978, fig. 9.6), converted into input values assuming 80% process efficiency, as do Probstein and Gold (1978). Soviet water requirements for the synthoil process were based on the moisture content of Soviet coal, and the trend of US data, converted to input values assuming 80% process efficiency.

[g] US estimates from Probstein and Gold (1978, fig. 9.6), converted to input values assuming 80% process efficiency. Numbers in Probstein and Gold (1978) for "maximum high wet cooling", but these authors note that in water-scarce areas these water requirements may be halved. For the Southwest and Northwest coal regions half of the water requirements specified in Probstein and Gold (1978) were assumed.

[h] From Astakhov (1977), mid-range values.

[i] Deduced from Astakhov (1977) and assigned typical characteristics of high sub-bituminous or low bituminous coals.

[j] Assigned based on typical values of coal rank.

[k] From Hittman (1974, 1975).

[l] "Representative" coals as given by Probstein and Gold (1978); their regions and those used in this report are matched as follows:

Probstein and Gold	This report
Four Corners	1. Southwest
Powder River	2. Northwest
Central Illinois	3. Central
Appalachia	4. North Appalachia
Appalachia	5. Central Appalachia

[m] From Astakhov (1979).

The amount of water consumed in the FGD unit of a power plant is computed using equations (7) and (9), such that

$$W_{51b} = (WF_1 + WF_2)\, y_{51}, \tag{11}$$

where W_{51b} = water requirement of power plant FGD unit ($\mathrm{m^3\, yr^{-1}}$),
WF_1, WF_2 = FGD water losses, as computed in equations (7) and (9) (t water $\mathrm{t^{-1}}$ coal),
y_{51} = coal input to power plant ($\mathrm{t\, yr^{-1}}$).

3.5.7. Liquefaction

In addition to cooling process and FGD unit water requirements, liquefaction facilities also consume process water and water for dust control. As noted earlier, the model process used to compute these water requirements is the synthoil process. Make-up water in a synthoil plant is needed for the major process streams, including coal preparation, slurry preparation, catalytic reactions, and oil and gas separation. These water requirements are aggregated into the parameter j of the following expression:

$$W_{52a} = jX_{52}, \tag{12}$$

where W_{52a} = process water requirement for liquefaction ($\mathrm{m^3\, yr^{-1}}$),
j = process water required per energy input ($\mathrm{m^3\, 10^{-15}\, J}$),
X_{52} = energy equivalent of coal input ($\mathrm{10^{15}\, J\, yr^{-1}}$).

Estimates of j are presented in Table 3.5.

For dust control, Probstein and Gold (1978) report that about 8–12 lb water 1000 $\mathrm{lb^{-1}}$ coal will be required by synthoil plants currently being designed in the USA. Using an intermediate value of 10 lb water 1000 $\mathrm{lb^{-1}}$ coal (equivalent to 1 $\mathrm{m^3}$ water 100 $\mathrm{t^{-1}}$ coal), the estimated water requirement will be:

$$W_{52a} = 0.01\, y_{52}, \tag{13}$$

where W_{52a} = dust control water requirement for liquefaction ($\mathrm{m^3\, yr^{-1}}$),
y_{52} = coal input to liquefaction ($\mathrm{t\, yr^{-1}}$).

Equations (12) and (13) are then combined in the water requirement model, yielding

$$W_{52a} = jX_{52} + 0.01\, y_{52}. \tag{14}$$

The same kind of approach used to compute power plant cooling water and FGD unit water requirements was also used to compute the water requirements in liquefaction facilities. The cooling water requirement is expressed as

$$W_{52b} = kX_{52}, \tag{15}$$

where W_{52b} = water requirement for liquefaction plant cooling ($\mathrm{m}^3\,\mathrm{yr}^{-1}$),
k = cooling water required per energy input ($\mathrm{m}^3\,10^{-15}\,\mathrm{J}$),
X_{52} = energy equivalent of coal input ($10^{15}\,\mathrm{J\,yr}^{-1}$).

The water requirement of the FGD unit in a liquefaction facility is given by

$$WP_{52c} = (WF_1 - WF_2)y_{52} \tag{16}$$

where W_{52c} = water requirement for liquefaction plant FGD unit ($\mathrm{m}^3\,\mathrm{yr}^{-1}$),
WF_1, WF_2 = FGD water losses (t water t^{-1} coal),
y_{52} = coal input to liquefaction (t^{-1} yr).

3.5.8. *Constants for the equations*

Values of the constants in the preceding model equations are presented in Table 3.5. The source of these values is described in the footnotes to Table 3.5.

3.6. *Water Requirements*

Using the water requirement model with the prescribed inputs from each of the coal regions, we obtain the results presented in Table 3.6 for the low and high regional scenarios. The amount of water consumed for various regions ranges from about 0.1 to 1.0 $\mathrm{km}^3\,\mathrm{yr}^{-1}$. For perspective, we can compare this range to average flows of major water delivery projects in the USA and the USSR. For example, the Volga-Moscow Canal transfers about 2.13 $\mathrm{km}^3\,\mathrm{yr}^{-1}$ from the Volga River to the thirsty industrial and residential areas around Moscow, while the California Project in the USA brings 4.2 $\mathrm{km}^3\,\mathrm{yr}^{-1}$ of water from water-rich northern California to rapidly growing and arid southern

TABLE 3.6. *Water requirements of the high and low regional scenarios for the year 2030, with pollution controls (in $km^3\,yr^{-1}$)*

Region	Low scenario	High scenario
Soviet Union		
(1) Ekibastuz	0.27	0.27
(2) Kuznetsk	0.43	0.77
(3) Kansk-Achinsk	0.69	0.84
(4) Donetsk	0.07	0.07
(5) Tungusska	—	0.58
United States		
(1) South-west	0.26	0.47
(2) North-west	0.65	1.01
(3) Central	0.55	0.99
(4) Northern Appalachia	0.46	0.51
(5) Central Appalachia	0.63	0.62

California (Golubev and Vasiliev, 1978). Since the projected requirement for water in coal-producing regions (0.1–1.0 $km^3 yr^{-1}$) approaches the magnitude of these water projects, one may conclude that a significant water resource engineering effort will be necessary to meet these water requirements.

Alcamo (1984) found it also instructive to look at the "water intensity" of coal development, i.e. the amount of water required to deliver a specific quantity of coal from mining to delivery of final fuels. This is obtained simply by dividing the total water requirement for the USA and the USSR (in $km^3 yr^{-1}$) by the amount of coal-fuel delivered (in billions of tce yr^{-1}). This calculation yields a water intensity for the two countries of about 1–2 km^3 water per billion tce coal. In other words, 1–2 t of water are consumed for every ton-equivalent of coal-fuel mix delivered.

It is important to recall, however, that these figures are based on the assumption of strict water conservation practices. A sensitivity analysis described in appendix C of the original study (Alcamo, 1983) notes that less water-conscious practices could use over 50% more water; i.e. 1.5–3.0 t of water may be necessary for each ton-equivalent of coal-fuel delivered.

Also of interest is the breakdown of total water requirements according to the different sectors of coal development. Figure 3.6 shows, for instance, that water consumed by the flue gas desulphurization equipment necessary to control air pollution is about 30% of the total water requirement, while mining and processing use 6% or less. Figure 3.6 also notes that the slurries that are assumed to transport some of the coal in the north-western USA consume over 15% of the total amount for the coal industry in that region.

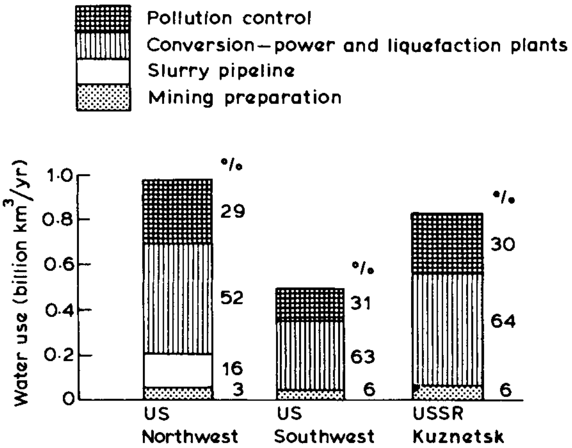


FIG. 3.6. Breakdown of coal-related use of water, based on high scenario calculations.

3.7. *Water Balance*

Now that the quantity of water needed for coal production in the major coal-producing regions is known, the question may be put "Will there be enough water available to satisfy this need?" The answer to this question can be attempted by comparing the water requirement for coal with the amount of surface runoff of the water basin in which the coal-producing region is located. This is an arbitrary yet reasonable judgement, since it assumes that a coal region can draw more economically on surface water than on groundwater. It is an especially realistic assumption for the USA, because groundwater overdrafting is already a major problem throughout much of the country. It also implies that it is more economic to take advantage of topography and gravity and draw water from within a basin than from outside it. This approach has already been used in studies for the US government conducted by Harte and El-Gasseir (1978) and March (1974).

For the USA, the demarcation of major drainage basins by the US Water Resources Council was used. Figure 3.7 notes the basins that are assumed to provide water for the coal-producing regions. Figure 3.8 compares the mean annual surface runoff in these basins (column 2) with computed water requirement for coal in the year 2030 from the high scenario case with pollution control (column 1). It is clear that there should be no *absolute* shortage of water for coal development, but this is, of course, a simplistic conclusion since this amount of water is not available each year. More reasonable measures of

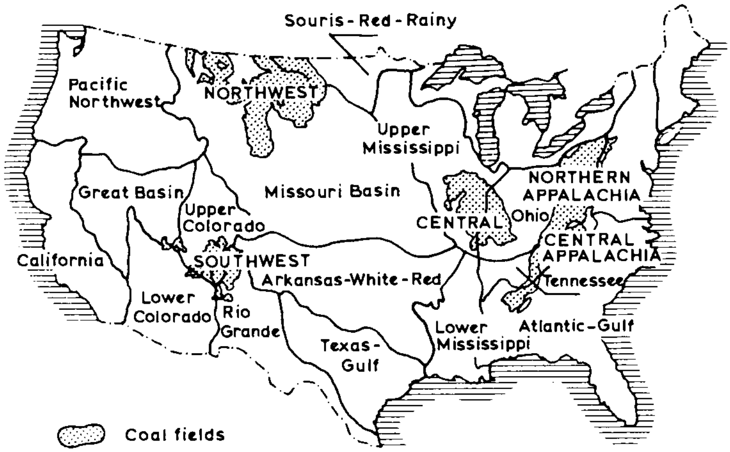


FIG. 3.7. US coal regions and major drainage basins. Note that regions are matched with basins as follows: Northwest – Missouri basin; Southwest – Upper Colorado and Rio Grande basins; Central – Upper Mississippi basin; Northern Appalachia – Ohio basin; Central Appalachia – Tennessee and Atlantic Gulf basins. (Source: details of water basins from US Water Resources Council (1978a)).

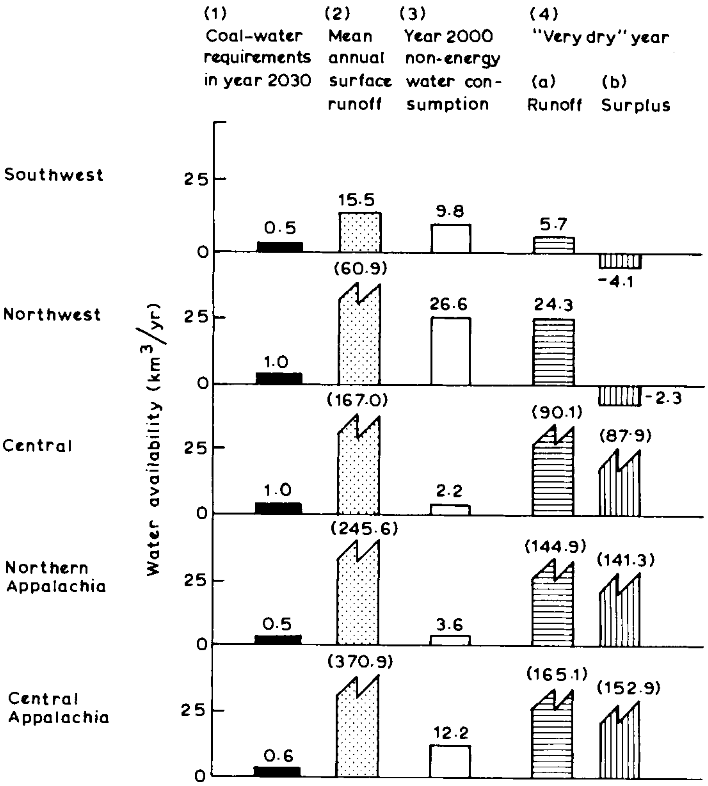


FIG. 3.8. Future availability of water for coal production in the US column (4b) gives the difference between dry-year runoff (column 4a) and non-energy water consumption (column 3). Sources: Water requirement for coal is computed elsewhere in this report and refers to the high scenario. Runoff data and non-energy water consumption have been taken from US Water Resources Council (1978a, 1978b).

long-term water reliability are the basins' drought flows presented in the column 4a. These are the annual flows of 95 % exceedance, i.e. those flows that are exceeded in 95 out of 100 years and occur during "very dry years" (US Water Resources Council, 1978b).

By comparing the coal water requirements (column 1) with these low flows (column 4a), we can see that there is still sufficient water for coal development, though in some cases coal requires a large fraction of this flow. For example, water needed for coal development in the south-west is nearly 10% of the region's low flow.

But for a better grasp of the future availability of water we should also account for water uses that will compete with the coal industry. Column 3 in Fig. 3.8 presents the projected water requirements for non-energy activities estimated by the US Water Resources Council (1978a, 1978b) for the year 2000. Column 4b gives the surplus water remaining after this non-energy water demand has been subtracted from the low flows. In the case of the south-west and north-west USA, a water deficit is observed, which implies that the coal industry will displace other projected water needs, such as irrigation and municipal water supply, during dry years in these regions. For the Central and Appalachian coal regions, Fig. 3.8 suggests that ample water should be available for all uses, even during low flow years.

However, it is important to note that this analysis takes a somewhat conservative approach and may therefore underestimate the possible severity of the future water supply problem in a few significant ways. First, as discussed earlier, a good deal of water-consciousness has been assumed for the future coal industry. As also noted previously, water requirements could actually be 50% greater than this report assumes. Secondly, non-energy water requirements were probably underestimated because projections for the year 2000 instead of for 2030 were used. Since it is likely that water requirements will continue to increase beyond the year 2000, it is also likely that this report underestimates the non-energy water requirements of the year 2030.

Finally, "in-stream" water requirements were neglected. These "in-stream" requirements, which are necessary for both maintenance of fish and wildlife habitats as well as navigation channels, may amount to 50% *or more* of the *mean annual surface runoff* of these water basins (US Water Resources Council, 1978a, 1978b).

For the USSR, the type of information used in the preceding analysis was unavailable in the published English literature. For example, the size of the drainage basins that would provide water for the coal-producing regions was unknown. But we can devise a crude estimate of water availability by assuming that the drainage basins of the USSR are of the same scale as those that provide water for coal in the USA (roughly 50,000–200,000 km^2). Table 3.7 uses this rough estimate, together with known values of mean annual surface runoff (in cm yr^{-1}), to compute average runoff values in coal regions of the USSR. Low flows in these regions are taken to be 50% of the mean flow. Although these figures are rough, they are nonetheless informative. For instance, they indicate that coal development in Ekibastuz may consume much of the region's available water; in Kansk-Achinsk and Tungusska it may deplete up to a quarter of the estimated low flows; and that the Kuznetsk Region may also experience pressure for water. In addition, note that competitive water uses in these regions (for example, for agriculture or municipal water supply) were not accounted for.

A more direct comparison between coal-water requirements in the USA and the USSR is made in Table 3.8, which presents estimated drainage area sizes that will be needed to provide water for each coal region. As expected, the drier regions will require larger areas from which to draw water for their coal industry. Ekibastuz, the driest of the coal regions, with only about 1 cm yr^{-1} of

TABLE 3.7. *Estimates of water availability in USSR coal regions*

Region	Coal-water requirement (km^3 yr^{-1})[a]	Mean annual surface runoff (km^3 yr^{-1})[b]	Low flow (km^3 yr^{-1})[c]
Ekibastuz	0.27	0.5–2.0	0.3–1.0
Kuznetsk	0.77	25–100	12.5–50
Kansk-Achinsk	0.84	10–40	5–20
Donetsk	0.07	5–20	2.5–10
Tungusska	0.58	7.5–30	3.8–15

[a] From this report, high scenario with pollution controls.
[b] Computed as the product of the mean annual surface runoff (cm yr^{-1}), from UNESCO (1978), and drainage area (50,000–200,000 km^2).
[c] Low flow = 50% mean annual surface runoff.

TABLE 3.8. *Drainage area required to satisfy requirements*

Coal region	Coal-water requirement (km^3 yr^{-1})	Drainage area required (km^2)[b]
Soviet Union		
(1) Ekibastuz	0.27	27,000
(2) Kuznetsk	0.77	1540
(3) Kansk-Achinsk	0.69	3450
(4) Donetsk	0.07	700
(5) Tungusska	0.58	3870
United States		
(1) South-west	0.47	1770
(2) North-west	1.01	2690
(3) Central	0.99	780
(4) Northern Appalachia	0.51	190
(5) Central Appalachia	0.62	230

[a] From Table 3.6, high scenario.
[b] For the USSR, computed from (coal-water requirement (km^3 yr^{-1}) mean annual runoff (cm yr^{-1}) × 10^{-5}). For the USA, computed from (coal-water requirement (km^3 yr^{-1}) × drainage area (km^2)/mean annual runoff (km^3 yr^{-1})). Runoff and drainage areas for the USA were obtained from US Water Resources Council (1978a, 1978b). Runoff for USSR from UNESCO (1978). All values have been rounded.

surface runoff (UNESCO, 1978), will need the runoff from about 27,000 km^2 during an average year to meet its coal-related water requirements. The arid north-west and south-west coal regions of the USA will need drainage areas of the same scale (on the order of a few thousand km^2) as do three of the five coal regions of the USSR: Kansk-Achinsk, Kuznetsk and Tungusska. These numbers suggest that the level of effort needed to provide water for coal in the American West will be comparable with the effort needed in the major coal regions of the USSR.

3.8. Conclusions

The "Order of magnitude" calculations presented by Alcamo (1984) are informative despite the rough approximations they invoke. It was found that it will be difficult to meet implicit production targets for certain coal regions in the USSR and the USA. In trying to attain a high scenario coal future, the USA would probably deplete a significant proportion of its Appalachian coal reserves, the USSR its Kuznetsk and Ekibastuz reserves. Furthermore, for this scenario it may be necessary for the USSR to develop an entirely new and remote Siberian coalfield, such as Tungusska.

Alcamo (1984) also found that, assuming a water-conscious future in which the coal industry is motivated to conserve water, then about 1–2 t of water will be required in both countries for each ton-equivalent of coal-fuel delivered. This figure might be 50% larger if the coal industry is less concerned about water conservation.

In addition, we found that the water requirement for coal in the USA coal regions was relatively small compared with future non-energy water uses in these regions. However, if we subtract these non-coal water uses from the water available during low flow years in the south-west and north-west, it seems that no water will be left for coal, no matter how small the water requirement. Coal will probably have to displace other uses in those regions, such as crop irrigation and municipal water supply.

In the USSR intense pressure for water in Ekibastuz will be experienced, while less severe competition may be seen in Kansk-Achinsk, Tungusska and Kuznetsk, in that order.

Overall, it appears that a four- or five-fold expansion of coal production in the USSR and the USA is likely to be constrained to some degree by the lack of readily available water. Both nations possess rich coal reserves, but both must confront the same problem of how to develop these with limited water resources available.

4. References

ALCAMO, J. (1983). *Water and Fire: Water Needs of Future Coal Development in the Soviet Union and the United States.* RR-83-11, International Institute for Applied Systems Analysis, Laxenburg.

ALCAMO, J. (1984). Water and fire: water needs of future coal development in the Soviet Union and United States. *Resources and Energy*, **6**, 77–105.

ASTAKHOV, A. (1977). The geological-mining appraisal of major coal basins in the USSR. Materials for the Coal Resources Working Group, Energy Project, International Institute for Applied Systems Analysis, Laxenburg.

ASTAKHOV, A. (1979). Development possibilities of USSR coal mining in the first quarter of the

21st century. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Pergamon, Oxford.

BAIBAKOV, S. N., BELYCH, B. N., MOROZOV, L. L., OLOPHYNSKY, E. J., RUKIN, E. J. and STEPIN, B. S. (1979). Economics of mineral and brown coal pipeline hydrotransport. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Pergamon, Oxford.

COMMISSION ON ENERGY AND THE ENVIRONMENT (1981). *Coal and the Environment*. HMSO, London.

CONAES (1977). Report of the Coal Subpanel to the Committee on Nuclear and Alternative Energy Systems. US National Academy of Sciences, Washington, D.C.

FETTWEIS, G. B. (1979). *World Coal Resources: Methods of Assessment and Results*. Elsevier, New York.

GOLD, H., GOLDSTEIN, D. J., PROBSTEIN, R. F., SHEN, J. S. and YUNG, G. (1977). *Water Requirements for Steam Electric Power Generation and Synthetic Fuels in the Western US*. US Environmental Protection Agency Report EPA-600/7-77-037, Washington, D.C.

GOLUBEV, G. and VASILIEV, O. (1978). Interregional water transfers as an interdisciplinary problem. *Water Supply and Management*, **2**, 67–78.

GONTOV, A. E. (1979). Hydraulic mining in the USSR. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Pergamon, Oxford.

GRENON, M. (1979). The WELMM approach to coal mining. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Pergamon, Oxford.

HÄFELE, W. (1981). *Energy in Finite World: A Global Systems Analysis*. Ballinger, Cambridge, Massachusetts.

HARTE, J. (1979). The risks of energy strategies: an ecological perspective. *Energy Risk Management* (ed. by G. T. Goodman and W. D. Rowe). Academic, London.

HARTE, J. and El-GASSEIR, M. (1978). Energy and water. *Energy II: Use, Conservation and Supply* (ed. by P. Abelson and A. Hammond). American Association for the Advancement of Science, Washington, D.C.

HITTMAN (1974). *Environmental Impacts, Efficiency and Cost of Energy Supply and End use, Volume I*. Hittman Associates Inc., USA.

HITTMAN (1975). *Environmental Impacts, Efficiency and Cost of Energy Supply and End Use, Volume II*. Hittman Associates Inc., USA.

LELYUKHINA, N. D. (1973). The cost effectiveness of location of the iron and steel industry. Quoted in Dienes, L. and Shabad, T. (1979). *The Soviet Energy System*. Wiley, New York.

MARCH, F. (1974). Water Resources Crosscut Team. *Water for Energy Development* (ed. by G. M. Kavadi and R. J. Krizek). U.S. Water Resources Council, USA.

MEL'NIKOV, N. LV. (1979). Open-cut mining in the USSR: techniques and economics. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Pergamon, Oxford.

OTA (1979). *Direct Use of Coal*. Office of Technology Assessment Report USGPO-052-003-00664-2, Washington, D.C.

PROBSTEIN, R. F. and GOLD, H. (1978). *Water in Synthetic Fuel Production: the Technology and Alternatives*. MIT Press, Cambridge, Massachusetts.

SHABAD, T. (1980). News Notes. *Soviet Geography*, **21**, 246–52.

SHELEST, V. A. (1979). Large-scale coal use in the USSR fuel/energy complex. *Future Coal Supply for the World Energy Balance* (ed. by M. Grenon). Pergamon, Oxford.

UNESCO (1978). *World Water Balance*. Compiled by the Hydrometeorological Service of the USSR. UNESCO, Paris.

US WATER RESOURCES COUNCIL (1978a). *The Nation's Water Resources 1975–2000, Volume I*. US Water Resources Council, USA.

US WATER RESOURCES COUNCIL (1978b). *The Nation's Water Resources 1975–2000, Volume II*. US Water Resources Council, USA.

WOCOL (1980). *Future Coal Prospects: Country and Regional Assessments* (ed. by R. P. Green and J. M. Gallagher). Ballinger, Cambridge, Massachusetts.

4

Environmental Impacts of Coal Transportation

1. Introduction 73
2. Environmental Impacts Common to all Modes of Transport 74
3. Rail Transport 75
   3.1. Operational Features 75
   3.2. Human Injury and Death 75
   3.3. Fugitive Emissions 75
   3.4. Other Effects 76
4. Coal Transport by Truck 76
5. Coal Transport by Water 77
6. Slurry Pipelines 77
7. Coal Transport Impacts in Perspective 79
8. References 80

1. Introduction

The transport of coal represents one component of the complete coal cycle—from exploration and extraction of the fuel, through beneficiation (refining and processing), storage and finally its conversion to an end-use product for consumers. Like all components of the cycle, environmental impacts result from the *transport* of coal, although in many coal impact studies these have been given scant consideration.

Coal transportation is executed by rail, truck (lorry), water (on rivers, canals, lakes and intercoastal waterways) and slurry pipeline or conveyer belt. Table 4.1 gives comparative tonnages of coal transported in the USA and the average haul distance involved for each method of transport. It will be seen that most is transported by rail. However, movement over short distances usually involves trucking, and this is the fastest-growing means of transport. As coal utilization is increased it is envisaged that transport by water will increase, particularly if there is a considerably expanded international trade, and there will also be significant developments in coal movement by slurry pipeline.

Environmental impacts of coal transport occur during *loading, en route* or during *unloading*. The impacts are likely to affect "natural" systems (including agriculture, forestry, horticulture and aquaculture), buildings and installations and involve death or injury to humans in an occupational capacity, or to the public. The environmental impacts on humans have received most

TABLE 4.1. *1975 coal transport in the USA (Morris, Sevian and Crowther, 1979)*

	Tonnage (10^8 tons)[a]	Average haul distance (km)
Rail	418	480
Waterway	69	765
Truck	79	80–120
Slurry pipeline	10	400

[a] The studies on which this chapter is based quote tonnages in tons (US): 1 ton (US) = 0.9072 tonne.

attention in a series of studies conducted under contract to the United States Department of Energy (Hamilton, 1979; Morris, Novak and Hamilton, 1979; Morris, Sevian and Crowther, 1979; Szabo, 1978). It is these reports that have been utilized in producing this summary account.

2. Environmental Impacts Common to all Modes of Transport

All forms of transport of coal exhibit certain common environmental impact features. All use land for terminal handling plants or for railroad installations or pipeline throughways. Rail transport and trucks cause damage to buildings and trucking coal causes major structural damage to highways. Air pollutants are emitted from engines powering the transportation facility (train, truck, tugboat, or pump station) and also noise results. Accidents are associated with all forms of transport—usually occupational accidents causing fewer deaths but more injuries, whereas the reverse is true when the public are involved.

The transport of coal in all its forms necessarily involves fugitive dusts, even though precautionary measures are increasingly taken. It is generally estimated that 0.02% of the coal loaded is lost as fugitive dust and a similar percentage lost at unloading. Until recently, loss during transportation has not been estimated, but more work is enabling these losses now to be assessed. There is little information of the overall effect of fugitive dust emissions from coal-handling and transport *en route*, although estimates have been made that 16% and 23% of the mean particulate loading of the air in the Atlantic and Mountain regions of the USA respectively are due to coal handling. It has been estimated that, in the USSR, air dust content in the vicinity of reloading bays for coal transportation and storage may reach 100 mg m^{-3} and be even higher if low wind velocities prevail. This would lead to effects on human health, crops, other vegetation, animals, and visibility and would prove a general disamenity.

As a rule, occupational impacts occur at loading, unloading and during the movement of coal. Effects to the public mostly occur during coal movement.

3. Rail Transport

3.1. Operational Features

Coal that is to be transported by rail is generally loaded at mine sidings into wagons that will carry 55, 75 or 100 t of coal. The loading and unloading rates, depending on the methods employed, can be several thousand tons per hour. It is not unusual for trains to be assembled to include over 120 wagons, transporting over 11,000 t of coal on a 480 km haul.

3.2. Human Injury and Death

More than half of the accidental deaths and injuries involved in the transport of coal in the USA result from collisions between trains and motor vehicles at road–rail crossings. Statistics for accidental injury and death are not given according to the freight being carried. Consequently estimates must be obtained for coal transport by assigning a proportionate share based on the weight or volume of coal transported over unit distance (t-miles travelled). Accident rates vary between regions, depending on the density of automobile traffic and the driving patterns (Coleman and Stewart, 1976; Rogozen *et al.*, 1977). There is thus a considerable degree of uncertainty associated with the derived estimates.

Assumptions have been made in relation to a 1000 MW(e) power plant being fed with coal involving a 480 km coal haul, requiring 2.2×10^6 t of coal a year, equivalent to 6.6×10^8 t-miles. Public and employee death and injury estimates are shown in Table 4.2. It will be seen that there is a far higher public death:injury ratio than there is for employees.

TABLE 4.2. *Public and employee deaths and injuries per annum estimated for a 1000 MW(e) power plant*

	Per 10^9 t-miles		Per plant-year		Per train-mile	
	Deaths	Injuries	Deaths	Injuries	Deaths	Injuries
Public	1.74	3.66	1.15	2.4	0.26	0.56
Employees	0.059	4.44	0.04	2.9	0.009	0:68

3.3. Fugitive Emissions

Fugitive emissions of coal particulates and other air pollutants occur during loading, unloading and *en route*. It is now estimated that about 50% of the coal dust losses occur during journey time and 25 % at loading and 25 % at unloading. There is some loss also due to spillage. Some estimates suggest that 0.2 kg t^{-1} is emitted during loading and a similar amount during unloading. Losses in transit may be from 0.05 to 1.0 % of the total coal involved. For this reason wind guards are used, moistened coal transported or latex polymer or asphalt sprays used. Some wagons incorporate flip-top lids (Szabo, 1978).

Szabo (1978) has given estimates for atmospheric emissions from a unit train carrying 11 430 t of coal making a round trip of 985 km (Table 4.3).

There are no Federal regulations controlling the emission of particulates during coal loading and unloading, and only West Virginia has regulations that are specific to coal handling.

TABLE 4.3. *Atmospheric emissions for a unit train transporting 11,430 t of coal on a round trip of 985 km (Szabo, 1978)*

Emission	kg/trip
Particulates	345
SO_2	780
NO_x	4855
HC	2075
CO	935
Particulates during loading	2285
Particulates during unloading	2285
Fugitive emissions in transit	5700

3.4. *Other Effects*

Coal transport by rail has a number of other environmental impacts relating to noise (engine, exhaust, horn, wheel–rail interaction and brake cooling blowers). Diesel engines emit exhausts and some chemicals may be leached from the coal during transit.

Sparks from trains may cause bush fires, initially along the permanent way thoroughfare, but in dry regions these may spread to surrounding vegetation. Control measures involve spraying vegetation along the permanent way with chemicals.

4. Coal Transport by Truck

The transport of coal by truck is usually confined to short-distance haulage but is a rapidly growing method of transport. It involves two point crossings (one loaded and one empty) during which exhaust fumes are emitted, spillages occur and fugitive dust losses are experienced (probably 0.04 % of the load during combined loading and unloading and 0.05 % during transit). For houses and other inhabited buildings near the road, there is exposure to engine, exhaust, tyre and cooling fan noise, sometimes in excess of 100 dB.

It has been estimated (Morris, Novak and Hamilton, 1979) that 7 accidents occur per 10^6 vehicle-miles and there are 0.03 deaths and 0.5 injuries per accident. This is equivalent to 2.4 deaths and 40 injuries for the generation of 1 GW(e) yr^{-1}.

Road damage by haulage trucks is a major environmental cost.

5. Coal Transport by Water

The transport of coal by water involves barges, loaded by conveyer belt, and assembled for movement by tug, individual motorized barges, or coastal-water ships. The main human hazards are occupational and these are compared with other forms of coal transport in Table 4.4.

In the USA coal is moved by water on Appalachian rivers, and on the Mississippi and Ohio Rivers. Diesel tugboats, loaded by conveyer belt, transport 18,000 t. During the loading of the amount about 3500 kg of dust would escape. Unloading, by a clamshell grab into a receiving bin, might also result in 3500 kg being lost but this is reduced where receiving bins are equipped with sprays. Estimates of 0.02% over a 2-day trip are given for fugitive escapes during transit.

6. Slurry Pipelines

Although the use of slurry pipelines for coal transport is not greatly developed it is seen as a potential evolution point in coal transport.

The deaths and injuries for slurry pipeline transport have to be estimated from oil pipeline deaths and injuries, taking into account the man-hours required to operate a coal-slurry pipeline. It is regarded as an entirely occupational risk, as shown in Table 4.4.

Slurry pipelines with electrically powered pump stations 100–160 km apart are envisaged. There are no emissions to the air and, due to building insulation and remote siting, noise is not a problem. Water demand is a problem and potential environmental impact as, in the USA, it is not economical to return the water for reuse. At the power plant the water resulting from the dewatering of the slurry has to be disposed of. It is usually saline and loaded with coal "fines".

Any accidental release of coal-slurry from a pipeline has the potential for damage to crops, local vegetation, aquatic systems and aquifers but the operation of pipelines is not yet sufficiently practised to allow estimates of this risk to be given.

TABLE 4.4. *Deaths and injuries resulting from coal transportation in the USA (Hamilton, 1979)*

Mode	Accidents per t-mile				Accidents per GW(e) yr^{-1}[(b)]			
	Public death	Public injury	Occupational death	Occupational injury	Public death	Public injury	Occupational death	Occupational injury
Rail	4.0×10^{-10}	8.6×10^{-10}	1.4×10^{-11}	1.0×10^{-9}	0.41	0.87	0.014	1.1
Truck	7.0×10^{-9}	1.2×10^{-7}	7.0×10^{-9}	1.2×10^{-7}	1.2	20.0	1.2	20.0
Barge	8.8×10^{-11}	9.0×10^{-11}	2.8×10^{-10}	1.0×10^{-9}	0.14	0.15	0.46	1.6
Pipeline[a]			6.7×10^{-11}	1.1×10^{-10}			0.16	0.26

[a] Coal slurry.

[b] Assumes: 1.0 GW(e) yr^{-1} requires 3.3×10^6 t of coal; coal energy content of 12,000 Btu lb^{-1}; average haul lengths by rail = 300 miles, truck = 50 miles, barge = 450 miles and pipeline = 700 miles.

7. Coal Transport Impacts in Perspective

It is necessary to keep estimates of the environmental impacts of coal transport in perspective: in relation to other components of the coal fuel cycle; in relation to other means of electricity generation; and in relation to the hazards experienced by human beings in everyday life.

Table 4.5 gives estimates of the number of accidental deaths and injuries attributed to various modes of coal transportation in 1975, in the USA, and as estimated for growth in energy use in 1985 and 1990 according to the conventional mix of energy sources.

In terms of the total fuel cycle it is possible to attribute risk, in terms of deaths, disease and injury to coal mining, coal processing, coal transport and the generation of electricity in coal-fired power stations. These estimates are given in Table 4.6.

Estimates have compared the total risk to public health of electricity generation to populations within 80 km of plants, for the USA in 1975 (Hamilton, 1979). This information is given in Chapter 7 (Table 7.8) of this book. It has also been estimated that of the total annual deaths in the USA between the ages of 1 and 74, 0.2–1.9 % are associated with smoking and about 2.5 % with car accidents, about half of which are due to drunken drivers.

Estimates derived in the calculation of the environmental hazards of different modes of coal transport are subject to considerable inaccuracies, due to incomplete data and analysis. As coal utilization increases and coal transport with it, these uncertainties will need to be resolved.

TABLE 4.5. *Estimated annual accidental deaths and injuries due to coal transportation in the USA (Hamilton, 1979)*

Transport mode	Subject		Total		
			1975	1985	1990
Rail	Employee:	deaths	1.7–6.9	3.1–12.3	3.5–14.0
		injuries	130–560	232–996	263–1130
	Public:	deaths	51–219	91–390	103–443
		injuries	108–461	191–820	218–932
Barge	Employee:	deaths	3.3	6.7	8.2
		injuries	321	656	797
Pipeline	Employee:	deaths	0	0.16	0.38
		injuries	0	15	34
Other	Total:	deaths	72	120	130
		injuries	2700	4300	4700

TABLE 4.6. *Estimated effects (based on 1000 MW(e) plant-year working at 65% capacity) of the coal fuel cycle (Hamilton, 1979)*

Cycle component	Subject		Deaths	Disease-injury
Mining	Workers:	accidental	0.6	42
		occupational	0.02–0.4	0.5–1.0
Processing	Workers:	accidental	0.05	2.9
		occupational	—	—
Transport	Public + workers:	accidents	0.3–1.3	1.2–5.9
Electricity generation	Public:	air pollution[a]	0.6	no estimate
		air pollution[b]	6	no estimate
	Workers:	accidental	0.1	3.3

[a] Population within 80 km radius of plant.

[b] Total population.

8. References

COLEMAN, J. and STEWART, G. R. (1976). Investigation of accident data for railroad-highway grade crossings. *Railroad-Highway Crossings, Visibility and Human Factors*, pp. 60–7. Transportation Research Board, National Academy of Sciences, Washington, D.C.

HAMILTON, L. D. (1979). Health effects of electricity generation. *Symposium on Health Effects of Energy Production*, Chalk River Nuclear Laboratories, Ontario, Canada.

MORRIS, S. C., NOVAK, K. M. and HAMILTON, L. D. (1979). *Databook for the Quantitation of Health Effects from Coal Energy Systems*. Biomedical and Environmental Assessment Division, Upton, New York.

MORRIS, S. C., SEVIAN, W. A. and CROWTHER, M. A. (1979). *A Coal Transport Environmental Residuals Model for SEAS*. Biomedical and Environmental Assessment Division, Upton, New York.

ROGOZEN, M. B., MARGLER, L. W., MARTZ, M. K. and HAUSKNECHT, D. F. (1977). *Environmental Impacts of Coal Slurry Pipelines and Unit Trains*. OTA-E-60-PTZ, Office of Technology Assessment, Washington, D.C.

SZABO, M. F. (1978). *Environmental Assessment of Coal Transportation*. PEDCo Environmental Inc., Cincinnati, Ohio.

5

Coal Combustion

1. Introduction 81
2. Methods of Coal Combustion 82
  2.1. Grate Firing or Fixed Bed Coal Combustion 82
  2.2. Pulverized Fuel Combustion 83
  2.3. Fluidized Bed Combustion 84
  2.4. Coal Liquid Mixture Combustion 84
  2.5. Combined Cycles 86
  2.6. Combined Power and Process Methods for Coal Utilization 86
    2.6.1. Magnetohydrodynamic (MHD) technology 86
    2.6.2. Power-process combination options 88
3. Effluents Produced During Coal Combustion 89
  3.1. Sulphur Oxides 90
    3.1.1. Sulphur compounds and fixed bed coal combustion 91
    3.1.2. Sulphur compounds in pulverized fuel combustion systems 91
    3.1.3. Fluidized bed combustion and sulphur removal 93
  3.2. Nitrogen Oxides 94
4. Emission Control Techniques 97
  4.1. Coal Cleaning 98
  4.2. Combustion Modification 99
  4.3. Flue Gas Cleaning 100
    4.3.1. Flue gas desulphurization (FGD) 101
    4.3.2. Removal of NO_x emissions from flue gases 101
    4.3.3. Ash emission control devices 103
  4.4. Fluidized Bed Combustion (FBC) of Coal 103
5. References 104

1. Introduction

Increases in the global use of coal as a source of energy will, at least in the next decade or two, involve much of its use for the production of thermal and electric energy based on one or other methods of its combustion. The effluents produced during these processes of coal combustion will be determined both by the composition of the fuel and by the method of combustion. The elements that make up the fuel, and the oxidant (and other gases present with the oxidant) in which it is burnt, will eventually be discharged to the environment with some consequent environmental effects.

The exact nature of the effluents will be influenced to a significant degree by the combustion conditions. Whereas emissions of carbon dioxide will be largely determined by fuel characteristics, SO_2 emissions will depend on both the sulphur content of the coal and on the combustion system employed. Combustion conditions are also the determinant of the formation of other potentially toxic products (including polycyclic aromatic hydrocarbons, nitrogen oxides and trace element emissions).

Traditional techniques of coal combustion aimed at high heat release rates and as complete combustion as possible. Effluent emissions were a secondary consideration (or initially ignored completely). New coal combustion systems are often based upon physical and chemical principles that have been applied for decades in other industries. However, their development as combustion processes involves a number of new problems, particularly in relation to the reliability of the new techniques (Smoot and Smith, 1985).

The increase in the scale of coal combustion, leading to possible adverse ecological consequences, has focused attention on the application of emission control technologies to the more traditional combustion processes. The potential for the reduction of effluent emissions from more advanced combustion techniques has been one of the areas of interest. Traditional and newer coal combustion technologies are now examined.

2. Methods of Coal Combustion

Combustion is the chemical process of the interaction of two substances—fuel and the oxidant—which takes place with intensive heat release and intermittent sharp increases in temperature. The concentration of reaction products increases while at the same time the concentration of primary reacting substances falls. There are two distinct fuel combustion processes: kinetic combustion and diffusion combustion. With *kinetic combustion* fuel and oxidant are homogeneously mixed before the mixture is fed into the combustion chamber. Natural gas and the liquid fuel utilized in internal combustion engines are combusted in this way. In *diffusion combustion* the fuel and oxidant are introduced into the combustion chamber separately. The quality of combustion thus depends upon:

the distribution of fuel and oxidant throughout the combustion chamber;
feed characteristics of the oxidant (aerodynamics of the gas stream);
the chemical reaction speed.

As chemical reactions at high temperatures take place rapidly it is the first two features, determined by design features of the furnace, that have most controlling effect. In the construction and operation of large-scale furnaces it is the diffusion principle of combustion that is utilized.

2.1. Grate Firing or Fixed Bed Coal Combustion

Coal is placed on a grate and the oxidant is fed to the solid fraction from below the grate. Early systems used a flat grate with manual feeding and ash removal. Later systems use mechanical devices such as a moving grate or a spreader stoker. In addition, the oxidant stream is often blown through the layer of coal on the grate; the weight of the coal must therefore be greater than the force of aerodynamic resistance created by the coal particles, although smaller particles are combusted in a suspension above the grate. The rate of combustion, under a given oxidant stream, is proportional to the specific surface of the coal particle—it increases with a reduction in particle size. Thus larger fixed bed furnaces with a high output level are limited by two conflicting factors: optimum size range of coal particles and intensity of the oxidant feed rate.

Fixed bed coal combustion has the following disadvantages:

fractions of coal $< 3\,\mu$m in diameter cannot be utilized;
uneven temperature distribution and fuel supply characteristics limit complete combustion throughout the furnace;
automation is difficult to achieve.

For these reasons high thermal productivity is difficult to achieve and the grate ash may be relatively high in carbon and the NO_x emissions relatively low.

2.2. *Pulverized Fuel Combustion*

Pulverized fuel combustion has been in operation for more than 50 years, and most large boilers in use now employ this technique. The oxidant stream is greater than the force of gravity on the particles, and coal powder ($< 200\,\mu$m in diameter) or pulverized fuel is blown into the furnace which has a number of burners. The pulverized fuel is combusted in suspension. A small fraction of the ash falls to the bottom of the furnace (*bottom ash*) where it can be removed. However, the major part is carried away as particulates in the flue gas (*pulverized fuel ash* or *PFA*).

Usually pulverized fuel units are operated above 80–100 MW_{th} in size because of the high cost of coal preparation. Capacities in excess of 1500 MW_{th} are not uncommon.

Pulverized fuel combustion has been a preferred technique due to its relative insensitivity to coal quality, the short residence time of the fuel in the furnace, so that furnace regulation is relatively easy, and the high degree of uniform fuel combustion at a high temperature. The disadvantages relate to the necessity for coal grinding to reasonable tolerance limits, the propensity for ash fusion at the temperatures encountered in the furnace and the production of sulphur oxides and oxides of nitrogen in the effluent gas stream. Dust and NO_x emissions are higher from a pulverized fuel combustion unit than with grate firing (NO_x is 500–700 ppm compared with 200–300 ppm) due to the higher combustion temperatures. Sulphur emissions depend upon the sulphur content of the coal.

Cyclone furnaces are a modification of pulverized fuel combustion units and increase performance as well as allowing for enhanced ash removal. Electrostatic precipitators and bag filters are utilized, as well as cyclonic devices, to obtain a high degree of pulverized fuel ash removal.

2.3. *Fluidized Bed Combustion*

The fluidized bed combustion (FBC) process makes use of an oxidant feeder rate to a fixed grate that overcomes the force of gravity of the fuel particles so that they move in a reciprocating fashion forming a "fluidized bed" of fuel suspended in the oxidant stream. This increases residence time of the larger coal particles in the reactor zone and improves the chance of complete combustion. The lightest ash particles may be carried away in the effluent stream and the heavier ones pass out of the bed.

There are two types of FBC applicable to solid fuels, namely bubbling and circulating beds. There are both atmospheric and pressurized versions of the two types, although none is at the commercial stage of development. Fluidized bed units are commonly used in process industries as it is an appropriate technology where chemical reactions between a fluid and solid particles are to be carried out. Many applications of the technique are found in the metallurgical and petrochemical industry. Also lignite gasification processes, where the lignite is reacted with steam and oxygen to produce a combustible gas (e.g. Winkler process), utilize fluidized beds. For purely combustion processes, FBC is used mainly for industrial boilers and furnace driers. Few units have been installed that are large enough to meet base load applications of the electrical utilities and there are questions relating to their availability.

The special characteristics that make the fluidized bed technique so useful for coal combustion are:

the high content of inerts in the bed (90–95 %) make the heat output less sensitive to fluctuations in fuel quality;

the temperature profile in the furnace is smooth and disturbances in performance are minimized;

the thermal properties of the bed make it possible to maintain a low combustion temperature (800–900 °C), but at the same time a high combustion efficiency; the recent use of submersible heating surfaces has enhanced this feature; these low temperatures result in low NO_x yield and the possibility of adding limestones (carbonate materials) to the coal feed in order to bind sulphur against release to the atmosphere;

the high heat transfer coefficient between the combustion gases and cooling surfaces allows units to be compact (even more so when the pressurized technique is used).

In the early stages of the application of this technique it faced a number of problems mainly concerning fuel input and ash removal.

2.4. *Coal Liquid Mixture Combustion*

A new technique for coal utilization now being developed is coal liquid mixture combustion. These are suspensions of coal dust in liquid, that is coal:liquid mixtures (CLM). The coal is finely ground and mixed with water, liquid oil or alcohols (such as methanol). A coal oil mixture (COM), coal oil water mixture (COW), coal water mixture (CWM), coal methanol mixture (CMM) or a mixture of coal methanol water (CMW) have all been tested. Morrison (1983) gives a comprehensive review of the use of coal liquid mixtures as alternative fuels for oil-fired boilers.

The commercially most interesting mixtures are CWM having a composition of about 70 % coal, 30 % water and less than 1 % chemical additives. Slurries with 40 % coal and 60 % oil or methanol have been investigated, but have not found widespread application.

The manufacturing process of CWM is relatively simple. Coal is pulverized to particle size $< 100\ \mu m$. The coal is then cleaned through a froth flotation or oil agglomeration step and finally the slurry is stabilized with additives and water. A suspension that does not demix readily, possesses a relatively low viscosity and has a relatively high concentration of coal is required. The additives are thus surfactants that stabilize the slurry, decrease viscosity and, in some cases, act as combustion improvers. This may allow the slurry to remain stable in storage over a period of weeks, but it is usual to use pumps or other equipment to keep the slurry homogeneous by activation.

A coal water mixture can be fired in converted oil-fired boilers. The burners have to be replaced by special burners, sometimes with a ceramic ignition chamber. The bottom of the boiler may require modification to allow ash removal, and soot-blowers and electrostatic precipitators or bag-house filters will require fitting. A converted oil-fired boiler will lose some capacity, the derating depending on furnace volume and shape, spacing between superheater tubes and coal quality. Derating may vary between 10–40 % of the original oil-fired capacity.

The converted boiler has a reduced combustion process temperature and the emissions are therefore less than from traditional coal combustion methods. Lower particulate, trace metal and SO_2 emissions result and NO_x emissions of 165–270 ppm have been quoted (Stigsson, Johansson and Carlson, 1984) from coal water mixtures containing 65–70 % solids. They also give SO_2 emission data of 210–475 ppm, the lower concentration resulting from runs with limestone ($CaCO_3$) additions to the fuel with an original sulphur content of 0.31 %.

The additional advantages of coal water mixture use include reduced transport costs, simplified handling with consequent reductions in noise and dust levels, ease of fuel regulation to the furnace chamber and relatively simple boiler modifications. It appears that coal slurry combustion will be particularly suited for smaller sized boilers for use in urban areas. A small number of boilers are already operating utilizing fuel with brand names such as "Fluidcarbon". Use of this CWM fuel has the most extensive combustion experience on record.

2.5. Combined Cycles

The combined cycle (CC) technique is a potential, future alternative to pulverized coal combustion, flue gas cleaning and steam turbines. The environmental effects of the two alternatives appear to be equivalent, but the CC technology may prove to be more economical, and flexible in the choice of fuel quality. With the greater complexity involved, however, problems of reliability may arise.

There are a number of different CC techniques being considered, all of which combine the use of gas turbines and steam turbines. Using pressurized versions of fluidized bed combustion, after clean-up the pressurized flue gases are expanded through a gas turbine and used to generate electricity and pressurize air taken in for combustion purposes. Use of steam turbines limits operating temperatures to 550 °C because of the properties of the materials from which they are constructed. However, the combination of gas and steam turbines increases the maximum temperature that can be employed quite considerably. This in turn increases the efficiency of electricity generation, and efficiency a little above 40% can be obtained.

Combined cycles are in use commercially for electricity generation but using oil or natural gas. For coal to be used, dust cleaning and improved construction materials need to be developed. Pressurized coal gasification, with gas cleaning, followed by gas combustion and CC, offers another promising line of development. This also has the greatest potential for emission control as sulphur and nitrogen compounds, and dust, will be removed before combustion.

2.6. Combined Power and Process Methods for Coal Utilization

Combined cycle techniques are not the only options open for instituting complex power and process systems; however, energy efficiency is not increased merely by the amalgamation of a power plant with some fuel processing technology. Rather, the aims are to meet several objectives using only one process operation, to increase the range of useful end products and to optimize the utilization of heat from production processes including, if necessary, fundamentally altering the method of electricity generation.

2.6.1. Magnetohydrodynamic (MHD) technology

A good example of optimizing the utilization of heat and adapting electricity generation methods is the combination of extracting nitrogen from the atmosphere, which requires a high temperature, and the use of magnetohydrodynamic (MHD) methods for electricity generation. This is also a high temperature process (Fig. 5.1). The coal is subjected to high-speed pyrolysis at a temperature of about 900 K. Gases, tar and semi-coked coal result. The light

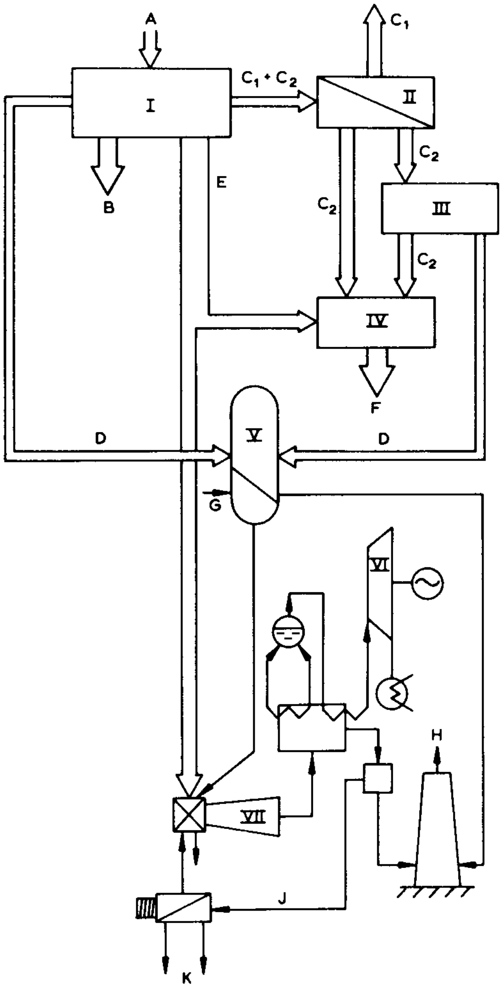


FIG. 5.1. Scheme for power and process utilization of coal incorporating a magnetohydrodynamic (MHD) power plant.
I. Pyrolysis installation. II. Fractionation of pyrolysis products. III. Thermal contact coking. IV. Briquette manufacture. V. Air preheater. VI. Steam turbine plant. VII. Magnetohydrodynamic. VIII. H_2S and HNO_3 removal chamber. A. Coal. B. Ash and slag. C_1. Light fraction of pyrolysis tar. C_2. Heavy fraction pyrolysis tar. D. Gases from pyrolysis and coking. E. Semi-coke. F. Domestic fuel. G. Air. H. Flue gases. J. Additive. K. H_2S and HNO_3.

fraction of tar can be used as a chemical feedstock or as synthetic liquid fuel; the gas is used to preheat air taken into the MHD plant; the semi-coke is used as a fuel for the MHD generator, but may also be mixed with heavy tar oil fractions to make coal briquettes.

In MHD the combustion gases are ionized in the combustion chamber by adding potassium carbonate (K_2CO_3), and a high conductivity plasma is produced. The hot plasma is passed through a strong magnetic field at high speed. The movement of the plasma (a conductor) through this field creates a current which is taken off by electrodes inside the channel. The K_2CO_3 removes SO_2 and NO_x from the flue gases. The potassium salts that result are collected and recycled. Elemental sulphur and nitric acid are by-products.

The energy efficiency of MHD power plants is in the order of 50 % and their net efficiency, due to engineering and materials limitations, about 45 %.

2.6.2. Power-process combination options

The new Turgai coalfield in the Kazakhstan Soviet Socialist Republic has vast resources of high sulphur brown coal. Some characteristics of this coal are given in Table 5.1. There has been considerable discussion about the best way to exploit this resource (Maslennikov *et al.*, 1983; Maslennikov and Shterenberg, 1983). The possibilities considered have included the following:

(i) steam-turbined power station (8 x 500 MW capacity) with direct coal cumbustion and high stacks (320 m);
(ii) Similar to the above with FGD fitted (stacks 180 m high);
(iii) combined cycle steam-gas power plant;
(iv) MHD power plant.

Comparisons are made in Tables 5.2 and 5.3. It is claimed that the cost of electro-energy in power and process complexes can be significantly lower than with the standard gas-turbine power plant and also solve some of the environmental problems of effluent emissions. The MHD process is largely in the development stage and problems concerning identification of suitable construction materials still have to be overcome.

TABLE 5.1. *Characteristics of Turgai brown coal from the Kazakhstan Soviet Socialist Republics*

	%
C	21.3
H	2.3
S	2.8
N	0.6
O	9
Wp	37
Ap	17
Qp (mg kg^{-1})	13.2

TABLE 5.2. *Technical and economic data relating to options for the use of Turgai brown coal in power plants*

	Steam turbine power plant	Steam and gas turbine power plant	MHD power plant
Capacity (MW)	4000	4000	4000
Unit capacity (MW)	500	500	1000
Operating conditions:			
Gas turbine K	–	1600	–
MPa	–	3.0	–
Steam turbine K	813	743	813
MPa	23.5	12.7	23.5
Preheating oxidant K	–	–	2273
Net efficiency of power plant (%)	39.5	49.5	50
Capital cost	100	59	130
Additional costs (%)			
FGD	27	–	–
Coal gasification + gas cleaning	–	48	–
NO_x removal and HNO_3 manufacture	–	–	8
Potassium salt separation	–	–	3
S extraction	–	–	6

3. Effluents Produced During Coal Combustion

Coal combustion generates a range of gaseous and liquid effluents as well as solid wastes. As an example the effluents from a coal-fired power plant generating 1000 MW(e) yr^{-1} (8.76×10^9kWh yr^{-1}) are given in Table 5.4. They are calculated from data given by Wilson and Jones (1974) and assume the power plant burns 3×10^6 t coal with 2% sulphur content, an energy content of 2.74×10^7 kJ t^{-1} and the thermal efficiency of the power plant is 38%; a fly ash removal of 99% and no flue gas desulphurization is assumed. Such a plant would have an installed capacity greater than 1000 MW(e) (a power plant with a generating capacity of 1000 MW(e) operating at 0.75 capacity would produce 6.75×10^6 kWh yr^{-1}). Brewer (1976) estimated trace metal and radioactivity emissions from such a coal-fired power station as 0.67 *t* and about 1 GBq respectively.

Particulates emitted will be mainly in the $< 1\ \mu m$ diameter range as particulate control devices have a higher efficiency of removal of the larger particulates. Volatilized minerals will leave in the flue gases as vapour.

The hydrocarbons in the flue gases are largely polycyclic aromatic hydrocarbons (PAHs). The environmental significance of these compounds is discussed in detail in Chapter 9. Many PAH compounds, such as benzo(*a*)pyrene, are carcinogenic and, therefore, in spite of their low concentrations in effluents, they need to be given serious attention. Increasing combustion temperatures, and oxidant level, cause benzo(*a*)pyrene levels to fall sharply. However, this operation, in its turn, increases nitrogen oxide

TABLE 5.3. *Technical and economic data relating to options for the use of Turgai brown coal in power and process complexes*

	Steam turbine power plant (320 m stacks)	Steam turbine power plant + FGD	Combined cycle gas turbine and coal gasification	MHD + coal pyrolysis
Capacity (MW)	3960	4000	4062	4069
steam turbine	3960	3960	–	1739
steam + gas	–	–	4062	–
MHD	–	–	–	2330
Fuel consumption (mtce yr^{-1})	7.60	7.72	6.85	12
Production:				
electricity (kWh)	22.8	22.8	22.8	22.8
light tar (mtce yr^{-1})	–	–	–	1.54
coal briquettes (mtce yr^{-1})	–	–	–	4.4
sulphur ($t \times 10^3\ yr^{-1}$)	–	–	382	220
HNO_3 ($t \times 10^3\ yr^{-1}$)	–	–	–	875
ammonia ($t \times 10^3\ yr^{-1}$)	–	–	69	–
Gaseous emissions:				
SO_2 ($t \times 10^3\ yr^{-1}$)	950	95	95	47
NO_x ($t \times 10^3\ yr^{-1}$)	50	50	8	25
Capital cost (K as %)	100	127	107	173
Annual cost (%) (0.24K + cost of fuel)	100	116	104	170
Relative value (annual cost = 100) of by-products:	–	–	9	78.4
tar	–	–	–	30
briquettes	–	–	–	34
sulphur	–	–	7.5	4.4
HNO_3	–	–	–	
ammonia	–	–	1.5	13
Electricity cost	100	116	95	89

concentrations. Hence it may be that when excess oxidant level is reduced from 10% to 2%, nitrogen oxide concentrations may be lowered by 20–30%. However, benzo(*a*)pyrene levels in the airborne effluent rise by a factor of four.

Most interest is focused on sulphur and nitrogen oxide pollutant emissions at present.

3.1. Sulphur Oxides

When coal containing sulphur is burnt, sulphur compounds will be distributed between the gaseous effluent and solid wastes. The exact compartmentalization will depend upon combustion temperature, oxidant

TABLE 5.4. *Annual effluent production (tonnes) from a 1000 MW(e) coal-fired power plant (UNEP, 1981)*

Airborne effluents:	
Particulates	3×10^3
Sulphur dioxides	11×10^4
Nitrogen oxides	2.7×10^4
Carbon monoxide	2×10^3
Hydrocarbons	400
Liquid effluents:	
Organic material	66.2
Sulphuric acid	82.5
Chloride	26.3
Phosphate	41.7
Boron	331
Suspended solids	497
Solid wastes:	
Bottom ash + recovered fly ash	3.6×10^5

supply level (in relation to the stoichiometric requirement for complete fuel oxidation) and the nature of the ash that forms. Figures 5.2, 5.3, 5.4 and 5.5 show how, when thermodynamic equilibrium is fulfilled, temperature affects the distribution of sulphur between solid and gaseous phases for a coal (2.8 %s) with a high ash content and how this is modified by properties of the coal ash that forms. At temperatures above 1800–1900 K practically all sulphur compounds are present in the gaseous phase. Below 1300–1400 K they tend to be present in the solid phase as sulphides if the excess oxidant level is relatively low (Fig. 5.2) and as sulphates if it is higher (see Figs. 5.3 and 5.4).

3.1.1. Sulphur compounds and fixed bed coal combustion

This type of combustion utilizes the coal in relatively large particles. The gaseous products of incomplete combustion have little contact with the coal particles or the ash. Hence, the likelihood of gaseous sulphur compounds combining with ash material is slight. Ash remaining on the grate may retain sulphides, but it is likely, depending on grate temperatures, that these will form sulphates or oxides. In the zone above the coal bed gaseous sulphur compounds oxidize to SO_2.

3.1.2. Sulphur compounds in pulverized fuel combustion systems

In furnaces where pulverized coal is combusted the temperature reaches 2000 K in the "flare ring", but is considerably lower in the convective streams within the furnace (1400 K). All sulphur is converted to SO_2 in the "flare

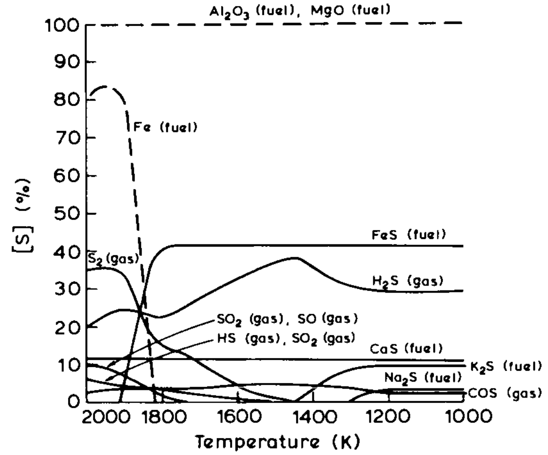


FIG. 5.2. Relative distribution of sulphur in solid (fuel) and gaseous phases and elements in the ash in combustion products of Donetsk coal where the oxidant coefficient a = 0.5; mineral S concentration or element concentration in fuel or gas is expressed relative to total.

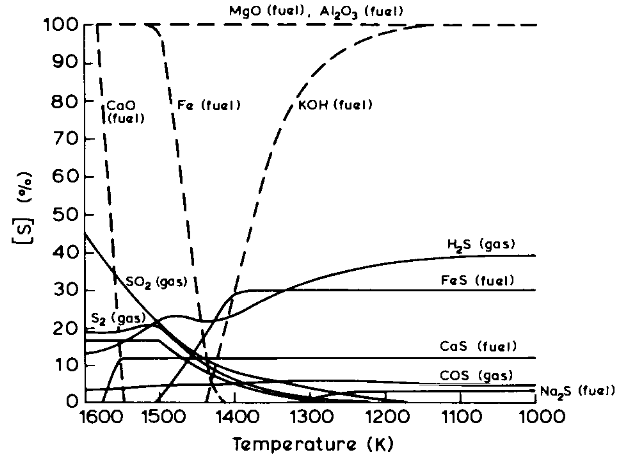


FIG. 5.3. Relative distribution on sulphur in solid (fuel) and gaseous phases and elements in the ash in combustion products of Donetsk coal (a = 0.8).

ring", but this could interact with the fly ash in the upper section of the furnace where the temperature is reduced. However, at 1400 K the speed of the reaction between SO_2 gas and ash is reduced (Fig. 5.5). Only where coal contains a relatively high content of alkali or alkaline earth metal (mainly Ca) does the sulphur form sulphates (Fig. 5.6). For most coals the sulphur associated with ash constitutes only 1% of the total weight of coal.

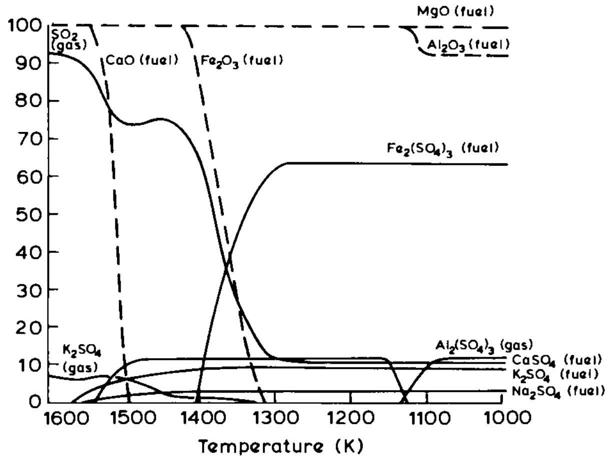


FIG. 5.4. Relative distribution of sulphur in solid (fuel) and gaseous phases and elements in the ash in combustion products of Donetsk coal (a = 1.1).

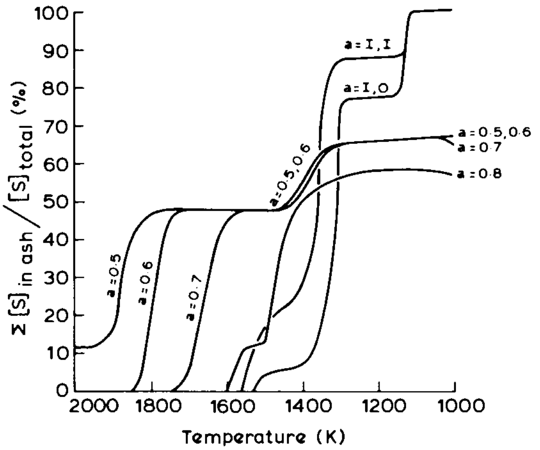


FIG. 5.5. Percentage of total fuel sulphur remaining in the ash from the combustion of Donetsk coal at different oxidant coefficients (a) 0.5–1.1 under thermodynamic equilibrium.

3.1.3. Fluidized bed combustion and sulphur removal

It is in fluidized bed combustion systems that the most favourable conditions exist for sulphur compounds fusing with the ash. A system with relatively low combustion temperatures (1050–1250 K), a high mass transfer ratio with the solid phase, and a relatively long contact time between combustion products and the solid phase, can remove well above 95 % of the SO_2 from the system when a $CaCO_3$ admixture is used with the feed (Figs. 5.6–5.9).

3.2. Nitrogen Oxides

The formation of nitrogen oxides (mainly NO and NO_2) during fuel combustion results from both the oxidation of atmospheric nitrogen at high

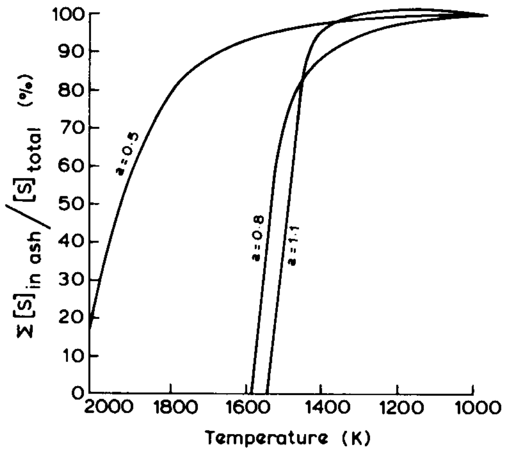


FIG. 5.6. Percentage of total fuel sulphur remaining in the ash (a = 0.5–1.1) from the combustion of Donetsk coal mixed with a calcium compound to give 5 % Ca content.

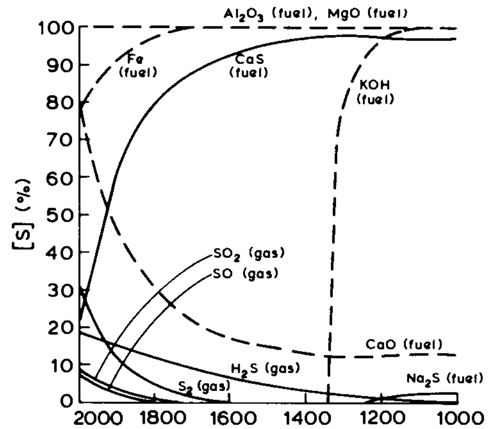


FIG. 5.7. Relative distribution of sulphur in solid and gaseous phases and elements in ash (a = 0.5) with addition of 5 % calcium (cf. Fig. 5.2).

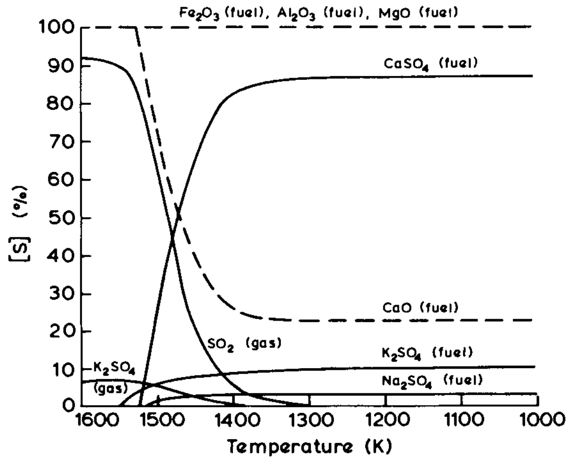


FIG. 5.8. Relative distribution of sulphur in solid and gaseous phases and elements in ash (a = 0.8) with addition of 5% calcium (cf. Fig. 5.3).

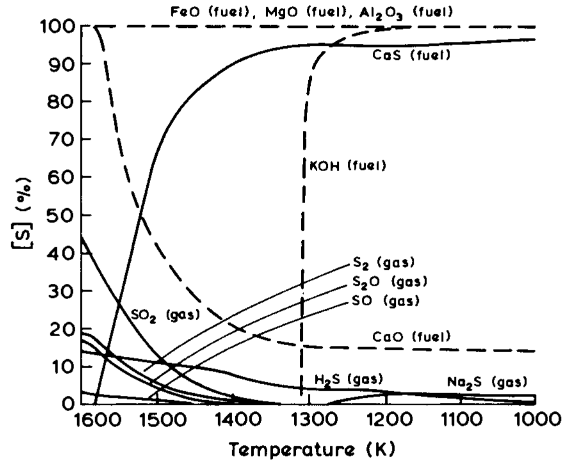


FIG. 5.9. Relative distribution in solid and gaseous phases and elements in ash (a = 1.1) with addition of 5% calcium (cf. Fig. 5.4).

temperatures (thermal NO_x) and oxidation of the nitrogen compounds contained in the coal (fuel NO_x). An approximate yield ratio is thought to be 20 : 80 (Morrison, 1980). Oxidation of fuel nitrogen involves decomposition of organic nitrogen compounds to form NH, NH_2, NH_3 and CN radical groups and then their transformation to molecular nitrogen or oxides.

Some nitrogen in the fuel is released early on in the combustion process with the volatiles; some nitrogen remains in the char and is released more slowly. Volatile NO_x emissions account for 60–80% of the fuel NO_x. Fuel NO_x generation is temperature independent over a wide range, but is influenced by the fuel : air ratio in the flame (Figs. 5.10 and 5.11). The formation of thermal NO_x is temperature dependent.

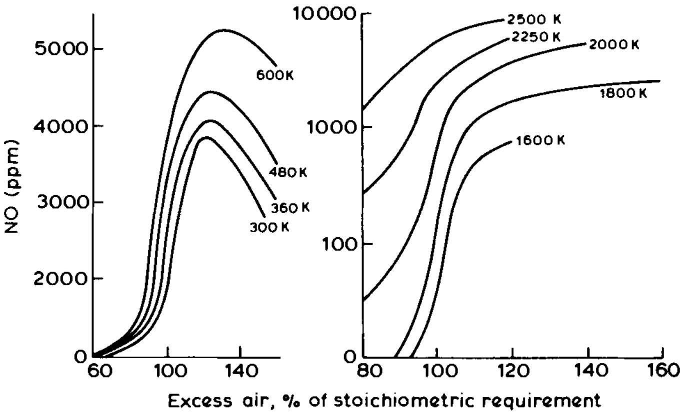


FIG. 5.10. Thermodynamic equilibrium nitric oxide (NO) content in the combustion products of natural gas in relation to combustion temperature (K) and oxidant supply.

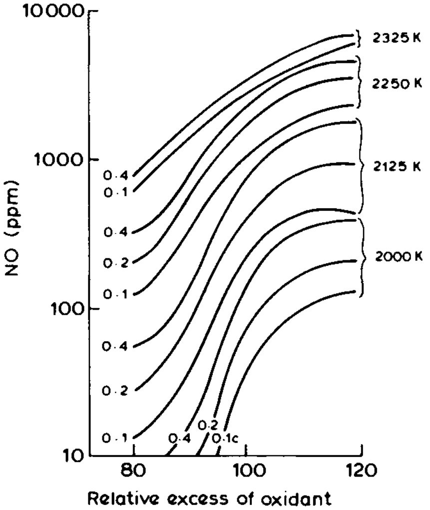


FIG. 5.11. Kinetic curves for the oxidation of atmospheric nitrogen to NO during combustion of natural gas in relation to combustion temperature (K) and relative amount of oxidant.

A number of features of coal combustion affect both temperature and supply of oxidant. The manipulation of these provides the possibility of NO_x emission control. Thus, when pulverized coal is combusted in conventional furnaces air is preheated. This influences the temperature of the flame and therefore thermal NO_x production. Excess air is normally provided in a furnace. This not only determines the fuel:air ratio but also flame temperature, thus affecting NO_x emissions. Furnace size affects temperature relationships, but this varies with boiler design. Burner design has the largest effect on NO_x emissions, and Morrison (1980) lists method of fuel injection, burner throat velocity, burner exit geometry, distribution of combustion air and rotation of the air stream as being of particular significance. Firing patterns greatly influence NO_x emissions, with tangential patterns producing the lowest emissions. Fuel nitrogen content (Fig. 5.12), along with other features of coal composition (type of N compound and coal rank), may influence fuel NO_x production.

As interest in NO_x effects on health, materials and its ecological impacts has developed (Smith, 1980), the control and removal of nitrogen oxides have been the subject of many studies on abatement procedures in relation to coal combustion (Yaverbaum, 1979; Morrison, 1980). The contribution of nitrogen oxides to acid depositions and their role in a number of environmentally significant atmospheric chemical reactions suggests that this interest will continue to grow.

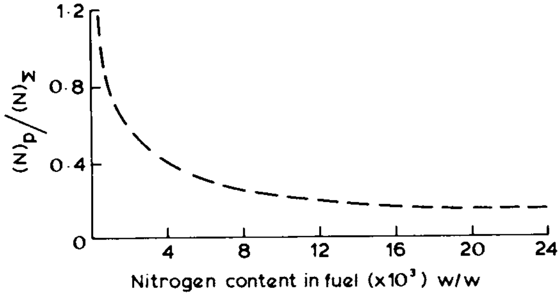


FIG. 5.12. Relation between the nitrogen content of fuel and the proportion of nitrogen content converted to nitrogen oxides during the combustion process; Np–N converted to NO_x, N_Σ–total N content in fuel.

4. Emission Control Techniques

Emission control technologies associated with coal combustion essentially tackle the problem in three ways: by removing or lowering the concentration of substances responsible for producing hazardous discharges *before the fuel is burnt*; *modifying the combustion process* itself to reduce hazardous compound generation; *extracting hazardous compounds* after combustion, *from flue gases*, before they are discharged into the atmosphere. A fourth option, sometimes available, is to use coal naturally low in sulphur.

4.1. Coal Cleaning

Coal cleaning aims at reducing both the sulphur content and the mineral trace elements in the organic coal material and hence their emission to the atmosphere. At the same time the resulting coal has a higher heating value and there is a lower ash load on the boiler. The emissions of sulphur and trace elements, as well as the amount of ash and particulates, are linked to the composition of coal.

The main forms of sulphur in coal are

(i) iron pyrites (FeS_2); this mineral formed during organic accumulation under reducing conditions is not part of the "coal molecule"; it forms in cleats and may be relatively stable (Caruccio, 1975);
(ii) organic sulphur; this is chemically part of the coal molecule;
(iii) sulphates and sulphides; these usually comprise only very small proportions of total sulphur content.

Sulphur may be removed during coal cleaning by physical or chemical means, or a combination of both. *Physical* coal cleaning methods utilize three basic principles:

(i) *density differences*—iron pyrites has a higher density than the organic content of coal; if the coal is crushed and pulverized it may be mixed with a liquid with a specific gravity between that of the organic and mineral components; separation is usually be settling devices or cyclonic separations;
(ii) *surface properties*—organic material in coal is hydrophobic, but ash is hydrophilic; by froth flotation, whereby air is blown through a suspension of coal and water, the ash will sink to the bottom and a coal-froth will float on the surface; unfortunately iron pyrites is also hydrophobic; organic fractions of coal are oleophilic whereas ash is oleophobic and thus an oil-coal mixture may be separated off;
(iii) *magnetism*—iron pyrites in coal is paramagnetic, while the organic fraction of coal is diamagnetic; a strong magnetic field is used to separate the minerals from coal held in a suspension of water; this process is known as High Gradient Magnetic Separation.

The methods described above are able to achieve ash levels of 2–4 % and bring about reductions of up to 80 % of the pyritic sulphur.

If coal has a substantial organic sulphur content or if ash levels of less than 10 % are required, then physical methods must be replaced or supplemented by *chemical* ones. Coal may be treated with aqueous solvents (alkaline or acid solutions) or by encouraging limited hydrogenation. Both treatments aim at breaking the chemical bonds between the sulphur and the rest of the coal molecule.

A chemical method of removing iron pyrites (the Mayer method) utilizes a solution of iron sulphate to finally precipitate elemental sulphur.

The physical methods of coal cleaning are not sufficiently efficient or the chemical methods well-developed enough to make coal cleaning a frequently employed option in limiting hazardous environmental discharges. There is also the associated problem of the disposal of the wastes which are low in organic content but high in sulphur and other mineral components.

4.2. Combustion Modification

A number of methods have been developed for pulverized coal burners to reduce emissions of both sulphur and nitrogen oxides. The methods are based on the modification of the combustion conditions with or without the simultaneous introduction of a gas "absorbing" material. The use of limestone ($CaCO_3$) or dolomitic limestone ($CaCO_3.MgCO_3$), introduced with the feed, in traditional combustion processes is not particularly effective, however, as the high temperatures involved give only a low efficiency of absorption.

NO_x emission reduction by burner modification is under development. The aim is to decrease the coal combustion temperature and oxygen concentration by recirculating cooled flue gases, or by a stepwise introduction of air to the flame which then becomes more widely spread and thus cooled. The injection of water into the combustion zone and two-stage combustion with intermediate heat removal have also been tested. The lower combustion temperature decreases the rate of oxidation of the nitrogen in the air (and incidentally because of the lower combustion temperature increases sulphur absorption by carbonate minerals). Unfortunately, the lower temperature also risks incomplete coal combustion and higher PAH emissions. This results also if the excess oxidant supply is lowered in the combustion zone. Utilizing these measures with pulverized coal combustion has only resulted in an NO_x reduction of between 30–40%.

An original method suggested to suppress NO_x formation is outlined in Fig. 5.13. It is based on a two-stage fuel combustion process used, for example, in magnetohydrodynamic generators. In the first stage of fuel combustion only 5–10% of the stoichiometric requirement of the oxidant is supplied and the products of incomplete combustion are held at temperatures of 2000–2300 K so that the nitrogen oxides which have formed decompose. Combustion is completed in a second stage, at below 1800K so that secondary nitrogen oxides are not generated.

The oxidation of nitrogen compounds in coal can be contained by primary fuel gasification wherein the nitrogenous compounds are converted either to

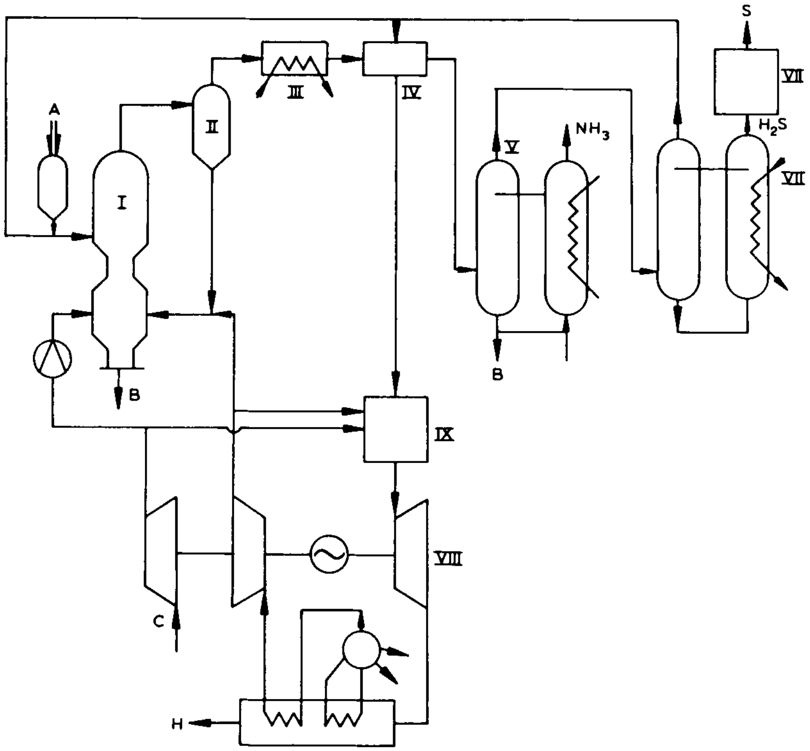


FIG. 5.13. Combined steam and gas turbine power plant incorporating coal gasification; I – gasification reactor, II – cyclonic furnace, III – heat exchanger, IV – gas heat exchanger, V – ash and NH_3 removal, VI – H_2S removal, VII – Klaus installation, VIII – steam and gas plant, IX – combustion chamber, A – fuel, B – ash, C – air, H – flue gases.

molecular nitrogen or ammonia. These are extracted from the gasification products and the cleaned gas is utilized in a second combustion stage. The most radical modification in order to reduce emissions is achieved with FBC (see Section 4.4).

4.3. Flue Gas Cleaning

Flue gas cleaning can easily reduce sulphur oxides emissions by over 90 %, NO_x emissions by more than 80 %, and remove over 99 % of dust. The techniques for sulphur and dust removal are frequently applied commercially. Techniques for NO_x removal are less frequently used (Fig. 5.14).

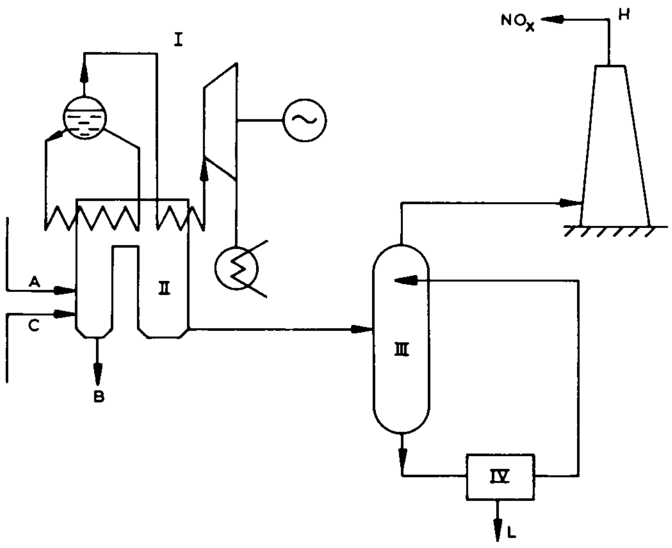


FIG. 5.14. Steam turbine power plant incorporating flue gas desulphurization; I – steam turbine, II – steam boiler, III – scrubber, IV – slurry removal, A – fuel, C – air, H – flue gases, L – slurry.

4.3.1. Flue gas desulphurization (FGD)

There are three main ways of treating flue gases to remove sulphur oxides, the first method being used most frequently:

(i) *non-regenerative processes*, whereby the sulphur dioxide gas is neutralized with $CaCO_3$ or $Ca(OH)_2$ and a slurry of sulphites and sulphates is formed (Fig. 5.15);

(ii) *regenerative cleaning methods*, where the SO_2 is extracted and concentrated, or sulphuric acid, ammonium sulphite or ammonium bisulphite is formed utilizing a sorbing agent (Fig. 5.16);

(iii) *catalytic methods*, based on the oxidation of SO_2 to SO_3, in the presence of a catalyst, and the production of dilute sulphuric acid (Fig. 5.17).

Although FGD systems are well established, there are a number of problems associated with waste disposal, corrosion and increased cost. FGD installation may increase capital costs of a power station by 30 % and also add to the recurrent costs of electricity generation.

4.3.2. Removal of NO_x emissions from flue gases

There are a number of methods for the extraction of nitrogen oxides from flue gases and the production of nitric acid. However, due to the relatively low concentrations of oxides and the large volume of flue gas to be treated such

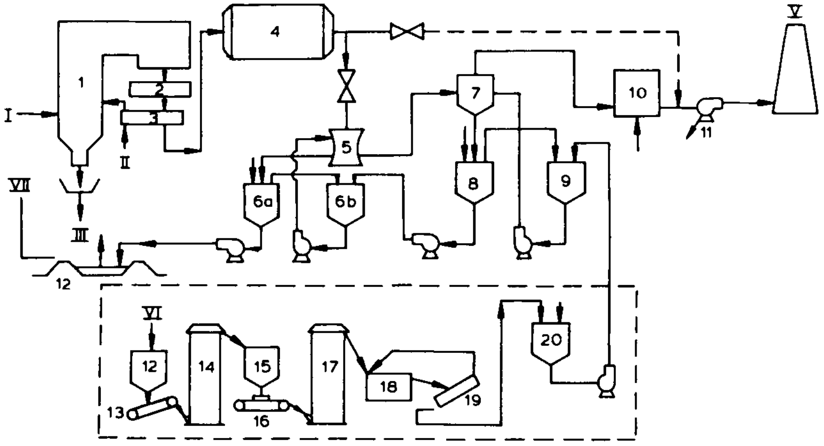


FIG. 5.15. Calcareous process scrubber for flue gas desulphurization on a coal steam turbine power plant; 1 – boiler assembly, 2 – economizer, 3 – air preheater, 4 – electrostatic precipitators, 5 – Venturi scrubber, 6a and 6b – neutralizing tanks, 7 – spray tower, 8 – neutralizing tank, 9 – holding container, 10 – flue gas preheater, 11 – flue gas extractors, 12 – bunker, 13 – conveyor, 18 – wet pebble mill, 19 – classifier, 20 – neutralizing tank, 21 – sedimentation tank, I – pulverized coal, II – air, III – ash, IV – steam, V – flue gases emitted after cleaning, VI – limestone, VII – return water.

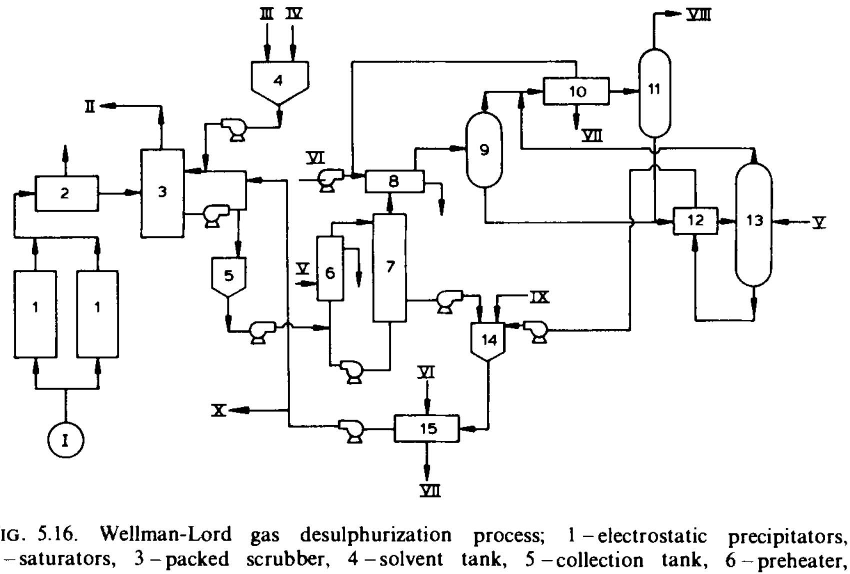


FIG. 5.16. Wellman-Lord gas desulphurization process; 1 – electrostatic precipitators, 2 – saturators, 3 – packed scrubber, 4 – solvent tank, 5 – collection tank, 6 – preheater, 7 – vaporizer-crystallizer, 8 – condenser, 9 – separator drum, 10 – condensor, 11 – separator drum, 12 – heat exchanger, 13 – SO_2 distillation, 14 – solvent tank, 15 – heat exchanger, I – flue gases from boiler assembly for cleaning, II – flue gases after cleaning, III – water, IV – NaOH, V – steam, VI – cooling water, VII – disposal of cleaning water, VIII – gas for H_2SO_4 production, IX – water from purifying system, X – steam.

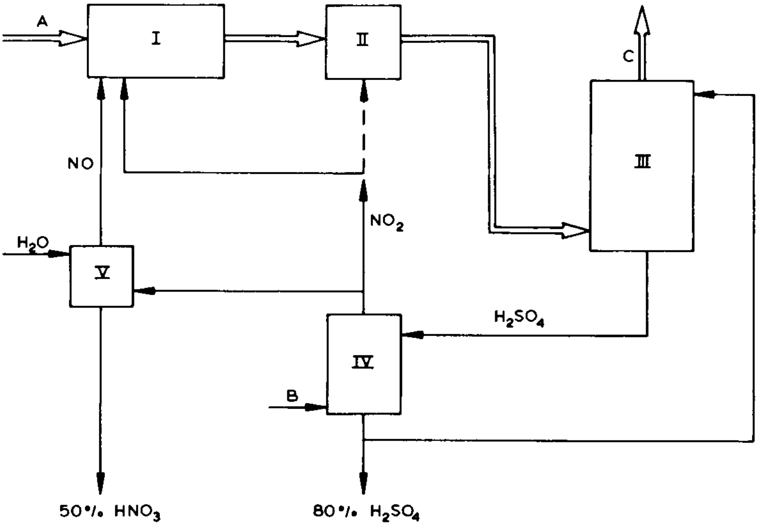


FIG. 5.17. Process for cleaning flue gases of SO_2 and NO_x; I – chamber, II – gas mixer; III – absorber; IV – nitrosulphuric acid producing apparatus, A – combustion products, B – air, C – cleaned combustion products.

methods are not practicable. In Japan an ammonia injection system (known as SCR) allows nitrogen oxides to be catalytically reacted with ammonia to form nitrogen gas.

4.3.3. *Ash emission control devices*

Finely dispersed dust from coal combustion can be efficiently extracted from flue gases before the gases are fed into the stack. The main removal devices (apart from the wet scrubbers that are installed as part of the sulphur dioxide control process) are:

(i) *electrostatic precipitators* that trap 98–99% of the particulates by electrically charging the particles and attracting them to an oppositely charge collecting plate;
(ii) *cyclonic collectors* relying on centrifugal force;
(iii) *bag-house fabric filters* which are most efficient in retaining the finer respirable particles; these filter out some volatile metals present in the flue gas.

4.4. Fluidized Bed Combustion (FBC) of Coal

As has already been indicated, one of the most promising trends in reducing emissions from coal burning is by FBC. There are two versions of fluidized bed combustion: bubbling fluidized bed and circulating fluidized bed (CFB), the latter being a "second generation" process. Additions of limestone

($CaCO_3$) or dolomite ($CaCO_3.MgCO_3$), in a powdered form, allow sulphur dioxide to be extracted:

$$CaCO_3 + O_2 + SO_2 \longrightarrow CaSO_4 + CO_2.$$

The temperature should be less than 1250 K and tubular heat exchange surfaces may be introduced into the fluidized bed to remove heat from the reactor zone. The bed temperature is thus kept between 1050–1250 K. Lowering the temperature also reduces nitrogen oxide production. Thus both sulphur and nitrogen oxide emissions can be reduced well below stipulated limits.

The constant movement of coal particles increases the heat transfer ratio and the process works well with low-grade (and low calorific content) coal.

One of the largest FBC boilers in operation is at the Rivesville power station in West Virginia, USA. Coal combustion averages 98.5 % and SO_2 removal from flue gases average 85–96 % overall efficiency. It is mainly because of the environmental advantages that the technique has had its breakthrough on the market for small- and medium-scale heat and power production.

The FBC technique is still under rapid development and larger units are being taken into operation. It is expected that soon it will be applied also to large-scale power production.

5. References

BREWER, S. T. (1976). Quantification and comparison of external costs of nuclear and fossil electrical power systems. *Energy and the Environment—Cost-Benefit Analysis* (ed. by R. A. Karam and K. Z. Morgan). Energy, Suppl. 1, 506–30. Pergamon, New York.

CARUCCIO, F. T. (1975). Estimating the acid potential of coal mine refuse. *The Ecology of Resource Degradation and Renewal* (ed. by M. J. Chadwick and G. T. Goodman). Blackwell, Oxford.

MASLENNIKOV, V. M. and SHTERENBERG, V. Y. (1983). Technical and economic comparative analysis of different methods for complex power and process fuel utilization. *UN-ECE Seminar on Integrated Utilization of Low-calorie Fuels.* Moscow.

MASLENNIKOV, V. M., VYSKUBENKO, Y. A., SHTERENBERG, V. Y., SMITHSON, G. R., ROBSON, F. L., LEMON, A. V. and LOCKON, V. T. (1983). *Steam and Gas Power Plants with Intercyclic Fuel Gasification and the Ecological Problems of Power Engineering.* State Scientific Publishing House, Moscow.

MORRISON, G. F. (1980). *Nitrogen Oxide from Coal Combustion—Abatement and Control.* Report ICTIS/TR 11. IEA, London.

MORRISON, G. F. (1983). *Combustion of Coal Liquid Mixtures*, Report ICTIS/TR 24. IEA, London.

SMITH, I. (1980). *Nitrogen Oxides from Coal Combustion—Environmental Effects.* Report ICTIS/TR 10. IEA, London.

SMOOT, D. L. and SMITH, P. J. (1985). *Coal Combustion and Gasification.* Plenum, New York.

STIGSSON, L. L., JOHANSSON, C. A. and CARLSON, R. G. (1984). Commercial production and utilization of fluidcarbon fuel. *6th International Symposium on Coal Slurry Combustion and Technology.* Orlando, Florida, USA.

UNEP (1981). *Environmental Impacts of Production and Use of Energy*, Vol. 1 (Study Director E. el-Hinnawi). Tycooly International, Dublin.

WILSON, R. and JONES, W. J. (1974). *Energy, Ecology and the Environment.* Academic Press, New York.

YAVERBAUM, L. H. (1979). *Nitrogen Oxides Control and Removal: Recent Developments.* Noyes Data Corporation, Park Ridge, N. J.

6

Coal Conversion Technologies

1. Introduction 105
2. Coal Conversion Process Routes 108
3. Direct Liquefaction 112
   3.1. Fundamental Considerations 112
      3.1.1. Chemical structure of coal 112
      3.1.2. Characterization of products 114
      3.1.3. Kinetics of coal hydrogenation 116
   3.2. Technical Developments 117
4. Gasification 129
   4.1. Basic Principles 131
   4.2. Commercial Processes 132
   4.3. Application and Improvement of Conventional Processes 138
      4.3.1. Moving-bed gasification 138
      4.3.2. Fluidized bed gasifier 142
      4.3.3. Entrained phase gasifier 143
   4.4 New Processes under Development 144
      4.4.1. Gasification by nuclear process heat 144
      4.4.2. Catalytic gasification 144
      4.4.3. Other processes 146
5. Pyrolysis-based Conversion Processes 147
6. Emissions from Gasification and Liquefaction Plants 149
7. References 150

1. Introduction

The purpose of coal conversion is the production of alternative fuels (including motor fuels) or the generation of a range of chemicals from coal by process routes such as gasification, direct and indirect liquefaction, hydropyrolysis and plasmapyrolysis. The application of these techniques is essential if the large reserves of coal are to be substituted for less plentiful fuels. The processes by which such conversion can be brought about are gasification and direct and indirect liquefaction. Definitions of these processes are given in Table 6.1. From Fig. 6.1 it can be seen that these processes involve an increase in the H:C atomic ratio (or hydrogenation). To achieve this, the necessary energy is ordinarily derived from oxidizing a portion of the incoming coal. Hydrogen is supplied either through direct or indirect utilization of the hydrogen atoms in water or through use of a hydrogen donor solvent.

Gasification and indirect liquefaction are undertaken industrially for the production of motor fuels and chemical feedstocks, such as NH_3 for fertilizer

TABLE 6.1. *Definition of coal gasification and liquefaction*

Gasification	Complete conversion of the organic matter of coal into gases (used for reduction, chemical synthesis and combustion)
Liquefaction	Conversion of as much as possible of the organic matter of coal into liquid products (used as fuels, gasoline, chemicals)
Process route 1	Direct liquefaction Hydrogenation of coal at high pressure
Process route 2	Indirect liquefaction Gasification of coal and consecutive catalytic conversion of the synthesis gas

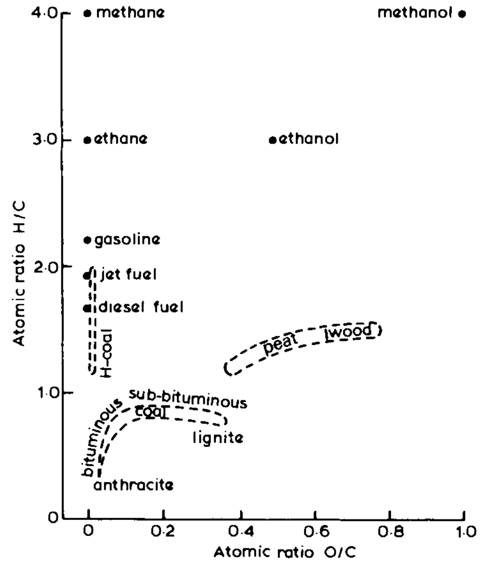


FIG. 6.1. The atomic compositions of several raw materials and finished products involved in coal conversion processes.

production. New improved processes are under development, some having reached the demonstration phase. The techniques of direct liquefaction of the 1930s and 1940s have been improved, and some have reached the demonstration plant phase. Commercial-sized plants, such as the projected SRC II, have been cancelled, since at present the products are not competitive with those obtained from mineral oil. Hydropyrolysis involves a "creaming off" of gases and liquids from coal by pressurized pyrolysis in a hydrogen atmosphere. The products are methane and aromatic liquids. The residual char can be used as feedstock for combustion and gasification. Plasmapyrolysis is undertaken to convert coal into acetylene or reduction gas at very high temperatures and at atmospheric or low pressures. Its feasibility has been proven in laboratory or pilot scale plants.

Since the spate of activity during the late 1970s in the development and commercialization of coal liquefaction and gasification plants (Chadwick and Lindman, 1982), the worldwide economic recession has considerably reduced the interest and investment in such projects (Hansen, 1981). Due to the large investment requirements of most synthetic fuel projects, some kind of sponsorship from governmental authorities is normally necessary. The Synthetic Fuels Corporation (SFC) was the government agency for supporting such projects in the USA, normally by loan guarantees and price supports. A recent vote in Congress ended funding for SFC and the DOE refused extra funds for the Great Plains Gasification project. Among those projects previously found ineligible for further funding were the SRC II project (up to then backed by the USA, the Federal Republic of Germany and Japan), the EXXON Donor Solvent Liquefaction Process and the Cogas gasification project, developed jointly by the FMC Corporation of the USA and the UK NCB (Conn, 1981; C&EN, 1981a; Schindler, 1982). In other countries, too, coal processing developments have undergone restrictions. There has been the cancellation of two large gasification projects in the Netherlands and the Federal Republic of Germany by Shell Nederland, and British Petroleum has withdrawn from a gasification pilot project in the United Kingdom (C&EN, 1982b).

Despite the adverse economic climate, certain projects remain viable. A number of ventures are being pursued in the Federal Republic of Germany (Speich, 1981; Cornils *et al.*, 1981; Theis and Nitschke, 1982; C&EN, 1981b), and continued developments in South Africa (Sterbenz, 1980), China (C&EN, 1981c) and the UK have recently been reported.

In the USA, the Texaco and Lurgi gasification processes and the H-coal liquefaction project appear to have governmental and industrial support and continued development is expected (C&EN, 1982a, 1982c, 1982d; HP, 1982; CEP, 1982; Kuhn, 1981). In addition, the U-gas process is likely to be pursued (C&EN, 1982a). In particular, a Lurgi plant for the production of 3.89×10^6 Nm^3 day^{-1} from 14,000 tons of coal is under construction by the ANG Gasification Company in North Dakota, USA, and is nearing completion (Koh, 1982). The Texaco process was successfully run over 175 hours for synthesis gas production in a 200-ton coal-to-ammonia plant owned by the Tennessee Valley Authority (HP, 1982), and a similar 165 ton day^{-1} plant has been operated since 1978 by Ruhrchemie in Oberhausen in the Federal Republic of Germany (CEP, 1982; Konkol *et al.*, 1982). In the Mojave Desert, coal is used to produce gas which, once desulphurized, converts water to steam and is burnt in a gas turbine to produce more steam to produce electricity. This Cool Water Coal Gasification Combined Cycle Power Plant has operated for over 8,000 hours. The H-coal process, for the production of synthesis crude oil, recorded a 131-day test run with 70% of this time at the 220 ton day^{-1} design capacity (C&EN, 1982c). Other Lurgi, Winkler and Koppers-Totzek (Shell-Koppers) processes continue in operation (Speich, 1981; Theis and Nitschke, 1982; C&EN, 1981b).

In situ coal-to-gas conversion, although not as well developed as surface methods, continues to be of interest and an experimental plant is being developed near Newark, UK. Data on dispersion of pollutants from the gasification reaction into subsurface water are now becoming available (Wang, Mead and Struermer, 1982; Struermer and Morris, 1982; Thurnau and Bates, 1980).

In this chapter the development and application of coal conversion processes over the last 2 or 3 years, in this changed economic climate, are discussed.

First, products from coal conversion processes and different process routes are described (section 2). Section 3 deals with direct coal liquefaction, proceeding from coal structure and the basic principles of the hydrogenation reaction. Section 4 deals with general progress made in coal gasification. The development of pyrolysis processes is briefly explained in section 5. Environmental problems, emissions from gasification and liquefaction plants and the costs of pollution control are discussed in section 6.

2. Coal Conversion Process Routes

The essential feature of coal conversion by gasification and direct or indirect liquefaction is the conversion of coal into higher-grade products (Fig. 6.1) such as fuel gases, motor fuels and chemicals which, at present, are mainly produced from oil and natural gas. Coal gasification and liquefaction processes are complex and a large variety of products can be obtained.

The main product routes for raw gas from coal gasification are shown in Fig. 6.2 (Jüntgen, 1982a). Simple purification and treatment of raw gas yield a hydrogen-enhanced reduction gas and a lean gas for combustion. By additional methanation, town gas and substitute natural gas (SNG) can be produced. An important process route, run on an industrial scale in South Africa, is the Fischer-Tropsch synthesis which yields gasoline, diesel fuel and chemicals. Another industrial scale process is methanol synthesis. Methanol is used as a solvent, as motor fuel, and is an important feedstock for chemical industries (e.g. for the production of lead free anti-knock agents such as MTBE, acetic anhydride and polyvinyl acetate). A future possibility is the large-scale production of motor fuel. Further processing of methanol by the Mobil process gives knock-proof gasoline and, under different reaction conditions, chemical products such as olefines. These technologies are under development. Synthesis gas is also the basis for NH_3-synthesis and so-called oxo-synthesis in which aldehydes are produced from olefines.

As shown in Fig. 6.3, coal from hydrogenation yields phenols, BTX-aromatics, olefines and higher nuclei aromatics, premium gasoline and diesel

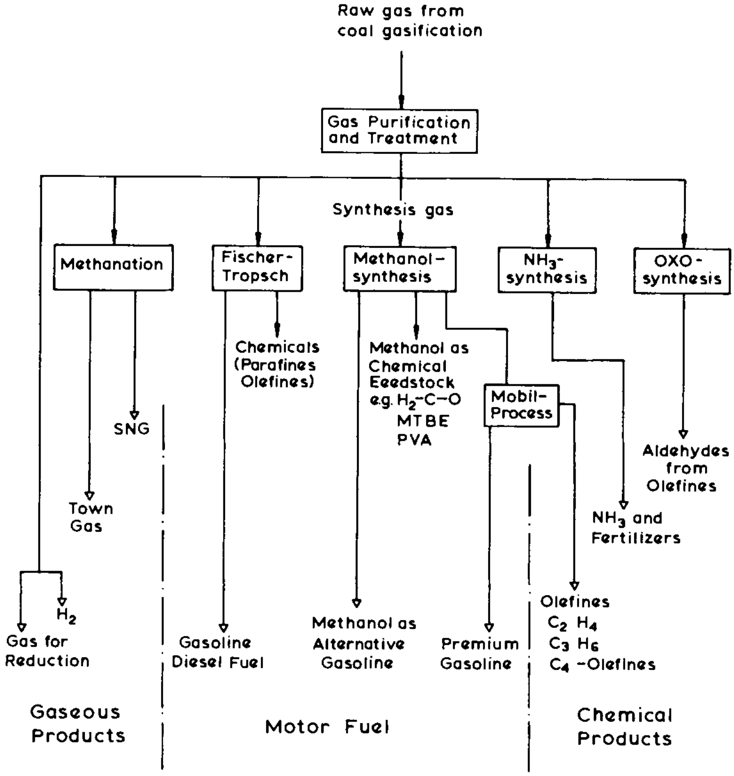


FIG. 6.2. Gasification and indirect liquefaction: production of fuel gases, gasoline and chemicals.

fuel (Jüntgen, 1982a). The first step of coal-oil processing is distillation by which light oil, middle oil and heavy oil are produced. The processing of these fractions utilizes techniques well known in mineral oil refining. The composition of the coal-oil fractions, however, differs from that of mineral oil fractions; they generally contain more aromatic compounds and as a result the process conditions have to be altered in terms of higher temperature, pressure and a longer residence time. Alternatively, the development of new and more efficient catalysts will be required. Most of the processes referred to here are run for removal of nitrogen, sulphur and oxygen, hydrocracking and octane number increase (Jacobssen and Gallei, 1981). In addition, phenols and BTX-aromatics can be extracted from light oil or higher nucleid aromatics from middle oil. The production of diesel fuel from middle oil requires extensive hydrogenation of aromatic compounds (Graser and Jankowski, 1982). The same applies to pretreatment, prior to steam cracking, of light oil to produce olefines (Hillebrand *et al.*, 1981).

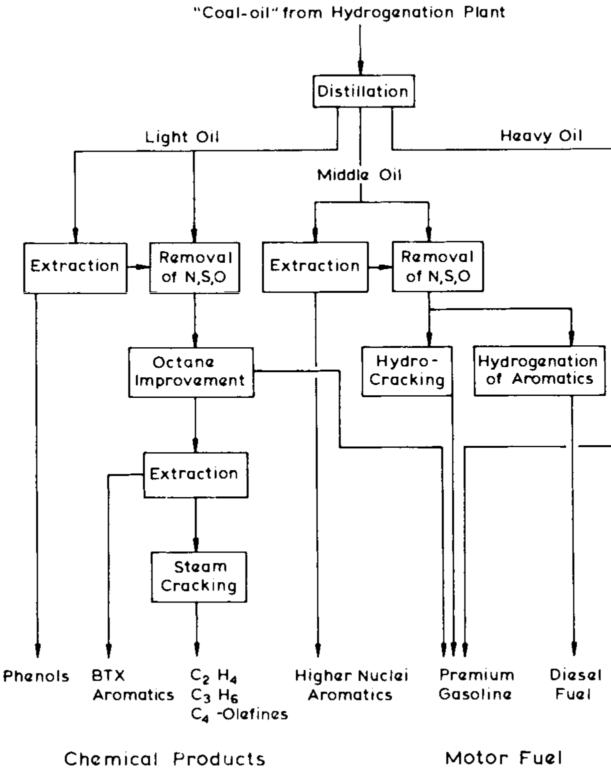


FIG. 6.3. Direct liquefaction: production of fuel oil, gasoline and chemicals.

A number of important comparisons can be made between direct and indirect liquefaction with regard to the production of different motor fuels (premium gasoline, gasoline, methanol and diesel fuel). A feasibility study of individual process routes for production on an industrial scale needs to consider the specific product yield related to coal input, the quality of motor fuel and production costs (Jüntgen, Nitschke and Wiegand, 1982). Each route has advantages, disadvantages and development needs. These are listed in Table 6.2. The relative importance of these factors can be expected to vary from one country to another, depending on the type of coal available, the quality of motor fuel desired and on raw material and investment costs. Even within a single country different methods have been used to assess the value of the motor fuel produced.

The quantity of coal which would be required to replace oil for the production of chemicals in the Federal Republic of Germany is between 32.3 and 43 × 10^6 t of coal per annum (Schulze and Schmidt, 1981). This would be required to replace the current oil consumption of 26.6 × 10^6 t per annum. The conversion rate, in terms of coal, of chemical production per unit weight

TABLE 6.2. *Advantages, problems and development aims of alternative motor fuel production processes (Jüntgen, Nitschke and Wiegand, 1982)*

	Advantages	Problems	Development Tasks
Gasoline:			
Direct hydrogenation	High efficiency; similar to petroleum	Coal suitability limited to brown coal or low rank bituminous coal low in ash	Refining of process; optimization of fuel quality; market penetration
Mobil synthesis	All coals usable; "Super" petrol grade achieved	Lower efficiency of production	Transfer to technical level; catalysts with higher stability
Fischer-Tropsch synthesis	All coals usable; "Normal" petrol grade achieved	Lower efficiency of production; higher production costs; wide product range	–
Methanol synthesis	All coals usable; high MON hence high engine efficiency; environmentally acceptable	Large storage tanks needed; distribution more costly	Engine development; market penetration
Diesel Fuel:			
Direct hydrogenation	High efficiency	Coal suitability limited; high "Cetane" values not obtainable (cf. F-T synthesis)	Production of a product with sufficient "Cetane" values
Fischer-Tropsch synthesis	All coals usable; high "Cetane" values obtainable; environmentally acceptable	Efficiency of production lower than direct hydrogenation but higher than gasoline production from F-T synthesis	Conversion of "maximum diesel oil" concept into large-scale technology

of feedstock corresponds, therefore, to 62–82 % of oil. A British study on the use of coal-derived liquids as a source of chemical feedstock has shown that most of these could be made from coal; however, considerable technical problems still have to be solved (Owen, 1982). It should be stressed that an alternative route for the conversion of coal into chemicals is via coal-based acetylene production (Passler, 1982).

3. Direct Liquefaction

3.1. Fundamental Considerations

3.1.1. Chemical structure of coal

Direct liquefaction of coal involves hydrogenation in the presence of hydrogen donor solvents and molecular hydrogen at high pressure and relatively low temperature. The hydrogenation mechanisms depend on the chemical structure of coal. This has long been under investigation. A synopsis of the main results and statistical constitution analyses, as well as information on the changes which occur during carbonization, is given by van Krevelen (1961). Knowledge of the chemical reactions and the constitution of coal show that coal is of macromolecular character (Wender *et al.*, 1981). The basic coal structure can be regarded as a three-dimensional polymeric network. This network is made up of hypothetic monomeric "basic units" having a mean molecular weight of about 400, connected by relatively weak linkages. The chemical structure of the basic unit changes, the aromatic part increasing as a function of coalification. The network forms an extended pore structure mainly consisting of micropores. Due to well-known molecular sieve effects, extractable compounds of lower molecular weight are trapped in the micropores and can be recovered by extraction with supercritical gases. Coalification products of vegetal origin, such as isoprenoid and straight chain alkanes, are trapped in the micropores.

Figure 6.4 shows an example of the constitution of the basic unit of high volatile bituminous coal, 83 % of which is carbon. Aromatic and hydroaromatic molecules with two to four aromatic nuclei are connected by five bridges—one of these is an aromatic ether bridge: the other four are aliphatic hydrocarbon bridge structures. The model molecule is connected by two further linkages to the rest of the coal structure and is, therefore, part of a larger macromolecule. The oxygen atoms are distributed as phenolic groups, as an aromatic ether bridge and as heterocyclic or alipathic ether bridges. Nitrogen and sulphur are bonded within the aromatic structure.

The strength of the different chemical bonds largely controls the behaviour of the coal molecule during chemical reactions. Various bonding forces are evident in this heterogeneous structure of the basic units. The bonding energies of specific aromatic and aliphatic hydrocarbons, which constitute the model of the coal molecule, are listed in Fig. 6.5.

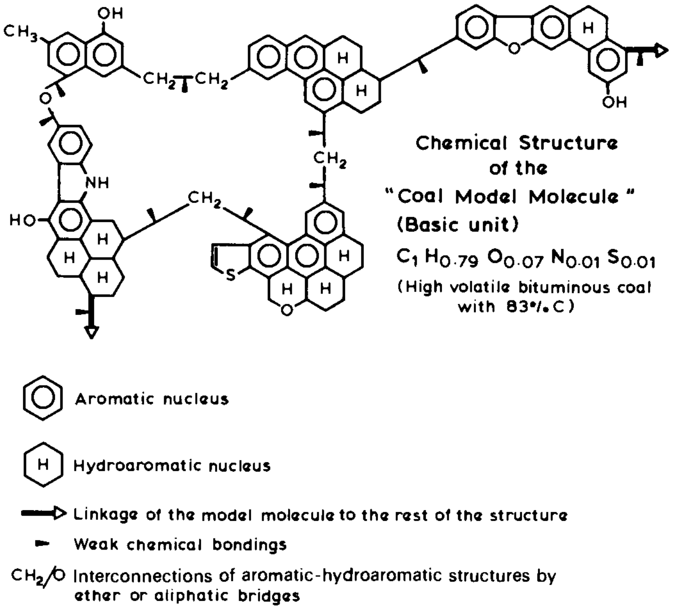


FIG. 6.4. Chemical structure of the "Coal Model Molecule".

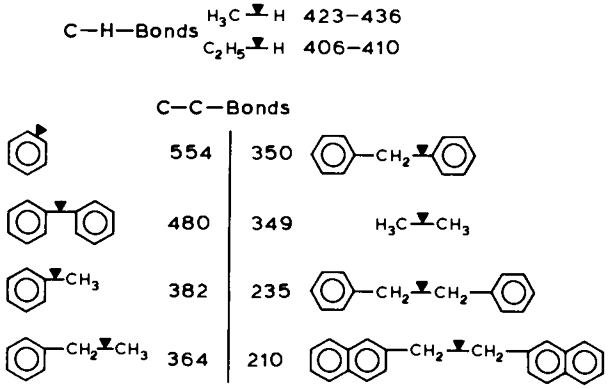


FIG. 6.5. Values of bond energy of hydrocarbons (kJ mol^{-1}).

Aromatic C–C bonds and C–H bonds are very strong. Furthermore, the C–C bonds in aliphatic hydrocarbon bridges become weaker the more the CH_2 group is substituted by aromatic compounds. It can, therefore, be assumed that the bridges between the single basic units of the macromolecule of coal and the bridges between the aromatic-hydro-aromatic compounds within a basic unit are less stable than the other chemical bonds.

The chemical reactions imply that the coal molecule is being broken down firstly into basic units and subsequently with increasing temperature into aromatic-hydro-aromatic structures. The aliphatic fragments formed during this process can react to produce small aliphatic hydrocarbons, whilst in parallel to these reactions the vaporization of molecules occluded in the micropores takes place. Further behaviour of the aromatic-hydro-aromatic clusters depends on the presence of reacting species. In the presence of hydrogen donor solvents and molecular hydrogen, the original aromatic-hydro-aromatic compounds of the base unit can take up hydrogen with simultaneous stabilization. More details of this mechanism are shown in Fig. 6.6. The first step of this reaction is the breaking of weak chemical bonds to form radical fragments. The next step is a quick reaction between hydrogen and the radical fragments producing so-called preasphaltenes, asphaltenes and hydro-aromatic compounds with molecular weights between 300 and 1000. In the following slower reaction the asphaltenes are converted to smaller molecular substances by hydrocracking and hydrogenation (Whitehurst, Mitchell and Farcasiu, 1980).

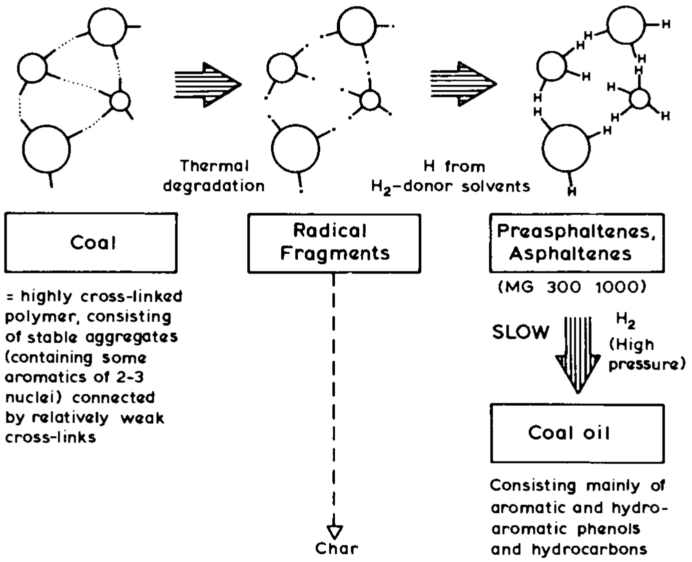


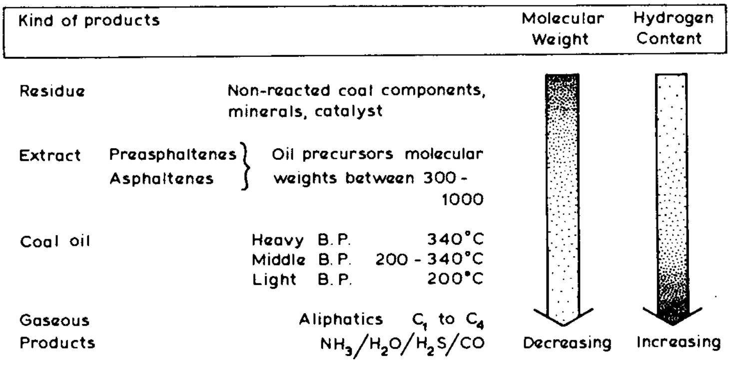
FIG. 6.6. Reaction steps of direct hydrogenation of coal (Whitehurst, Mitchell and Farcasiu, 1980).

3.1.2. *Characterization of products*

Further knowledge of the hydrogenation mechanism is derived from a characterization of the products, varying as a function of process conditions.

As shown in Fig. 6.7, products with decreasing molecular weight and simultaneously increasing hydrogen content are extracts consisting of pre-asphaltenes and asphaltenes. These are black solids at room temperature, but at higher temperatures release oil fractions and gaseous aliphatics with one to four carbon atoms. Furthermore, H_2S, NH_3, H_2O and CO are formed by the reaction of hydrogen with part of the oxygen, nitrogen and sulphur contained in the coal. A solid residue remains, consisting of non-reacted coal-components, coal-minerals and catalysts. The product distribution depends on the process conditions (residence time, temperature and hydrogen pressure). The type of coal and catalyst used is also important. A significant overall process parameter is the hydrogen consumed during the reaction, in terms of hydrogen consumption versus carbon input. The proportion of oils and gaseous products increases with increasing hydrogen consumption. Methods of product separation and characterization are listed in Table 6.3.

A new German technology utilizes process conditions involving a high hydrogen consumption resulting in a high yield of coal-oils. Analytical data on coal-oil are shown in Fig. 6. 8. The main constituents of the light coal-oils are BTX-aromatics, phenols and derivatives of pyridin and also a large proportion of paraffins. The main components of middle oil are naphthols, di- and trivalent phenols, hydro-aromatics and aromatics with three nuclei. From Fig. 6.9 it may be seen that the proportions of HPLC fractions are different for coal-oils obtained from bituminous coals than those from lignite. Coal-oil produced from the latter contains more aliphatics and less aromatics with three nuclei.



Kind of products			Molecular Weight	Hydrogen Content
Residue		Non-reacted coal components, minerals, catalyst		
Extract	Preasphaltenes Asphaltenes	Oil precursors molecular weights between 300 - 1000		
Coal oil		Heavy B.P. 340°C Middle B.P. 200 - 340°C Light B.P. 200°C		
Gaseous Products		Aliphatics C_1 to C_4 $NH_3/H_2O/H_2S/CO$	Decreasing	Increasing

Product distribution depends on:

- Residence time, temperature, H-pressure, catalyst, coal

Important parameter:

- kg hydrogen consumption /kg carbon in coal

FIG. 6.7. Products of coal hydrogenation.

TABLE 6.3. *Product separation and characterization methods*

(a) FRACTIONATED EXTRACTION USING SOLVENTS OF DIFFERENT DISSOLVING POWER

(mainly used for the characterization of products rich in extract)

Suitable solvents for dissolution:

preasphaltenes:	*asphaltenes*:	*oil*:
cresol	benzene	n-hexane
pyridine		n-pentane
tetrahydrofuran		

Characterization of extracts:
- molecular weight
- ultimate analysis
- C:H distribution

(b) DISTILLATION IN COMBINATION WITH OTHER METHODS

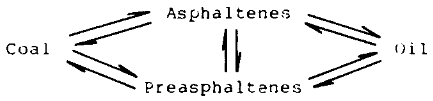
Separation of residue and extract from oil by flash distillation

Separation of oil fraction in heavy, middle and light oils by atmospheric distillation

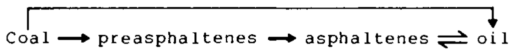
Separation of single oil fractions by
- alkaline extraction (phenols)
- acid extraction (pyridine bases)
- high-performance liquid chromatography (HPLC)
- separating according to ratio hydro-aromatic/aromatic
- gas-chromatography (GC) separating according to TLC molecular weight

3.1.3. *Kinetics of coal hydrogenation*

The kinetics of coal hydrogenation will not be discussed in any detail, as comprehensive accounts are available (Hill *et al.*, 1966; Gurran, Shruck and Gorin, 1967; Wiser, 1968; Gorin *et al.*, 1971; Jüntgen, 1982b; Abusleme, Badilla-Ohlbaum and Neuburg, 1983). Overall the reactions may be depicted as:

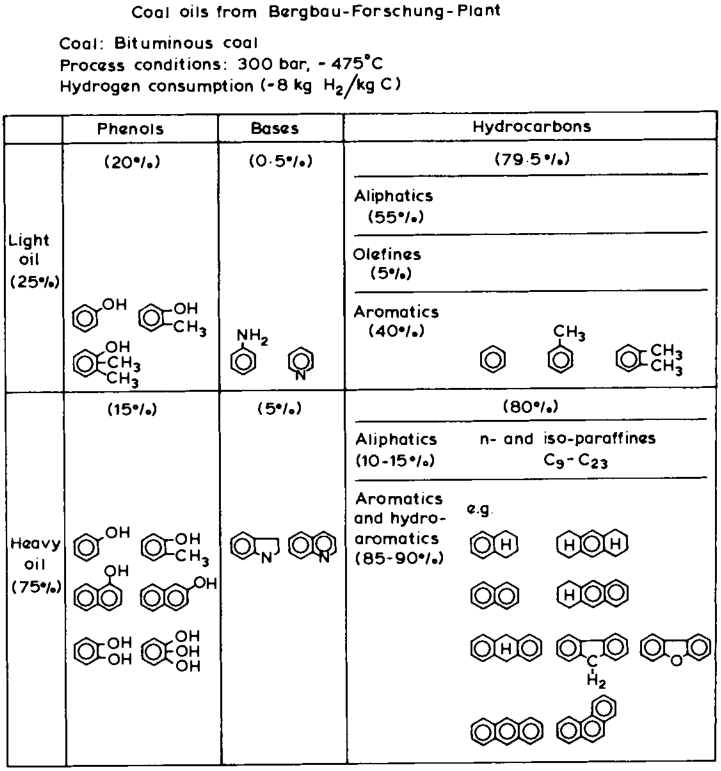


The evaluation of experimental batch data suggests that the reaction pathway depends upon the temperature conditions and the type of coal utilized. The rate of coal conversion and hydrogen transfer is controlled by the rate of thermal decomposition of the coal. Different rates arise from the rupture of bonds of varying strength in the coal matrix. A direct reaction pathway from coal to oil can be interpreted as a desorption of substances occluded in the micropore structure of the coal matrix.



At temperatures of about 450°C and above, the reaction mechanism seems to change; there is then no direct pathway from coal to oil, but only from coal to asphaltenes. The oils are capable of being converted into higher condensation products (asphaltenes 2).

$$\begin{tikzcd}
\text{Coal} \arrow[r] \arrow[rr, bend left] & \text{preasphaltenes} \arrow[r] & \text{asphaltenes} \arrow[r] & \text{oil} \arrow[r] & \text{asphaltenes 2}
\end{tikzcd}$$



Coal oils from Bergbau-Forschung-Plant

Coal: Bituminous coal
Process conditions: 300 bar, ~475°C
Hydrogen consumption (~8 kg H_2/kg C)

	Phenols	Bases	Hydrocarbons
Light oil (25%)	(20%)	(0.5%)	(79.5%) Aliphatics (55%) Olefines (5%) Aromatics (40%)
Heavy oil (75%)	(15%)	(5%)	(80%) Aliphatics (10-15%) n- and iso-paraffines C_9-C_{23} Aromatics and hydro-aromatics (85-90%) e.g.

FIG. 6.8. Characterization of light and middle coal-oils (Liphard *et al.*, 1981).

3.2. *Technical Developments*

The main technical developments in the Federal Republic of Germany have been concerned with the improvement of the well-known Bergius-Pier process (Kronig, 1978). The overall coal hydrogenation reaction, under high pressure, is described in Fig. 6.10. The coal-oil yield, under given reaction conditions (pressure, temperature, residence time and catalyst type and quantity), is

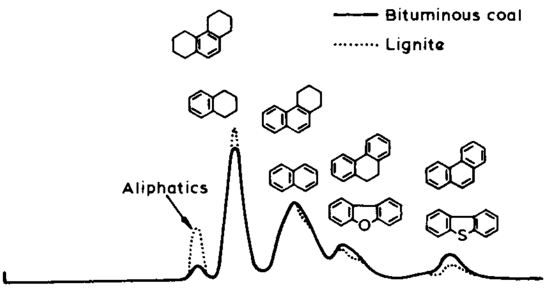


FIG. 6.9. Typical HPLC chromatograms of neutral coal-middle-oils from bituminous coal and lignite.

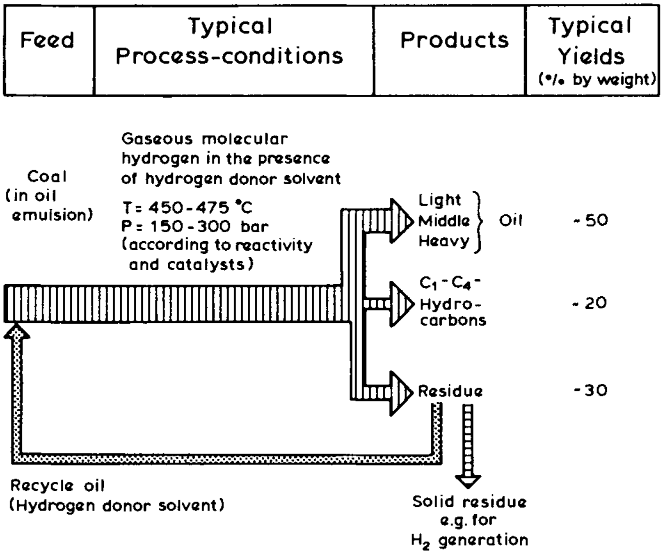


FIG. 6.10. Overall coal hydrogenation reaction.

dependent on the type of coal under investigation. Bergbau-Forschung runs a small 250 kg day^{-1} pilot unit for investigating the behaviour of different coals under conditions similar to those prevailing in industrial hydrogenation plants. An important result concerning the reactivity of different coals under comparable process conditions is shown in Fig. 6.11. It can be seen that the yield of oil products, corresponding to coal reactivity, increases with increasing rank, reaches a maximum in the range of high volatile bituminous coal and then decreases. A simplified flow sheet of a coal liquefaction plant, utilizing this technology for the production of gasoline, is shown in Fig. 6.12.

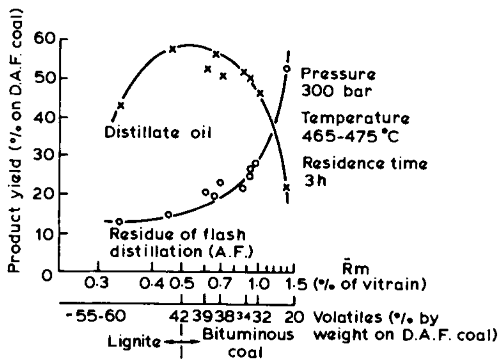


FIG. 6.11. Reactivity of coals vs. H_2 in the presence of H donor solvent (Strobel *et al.*, 1981).

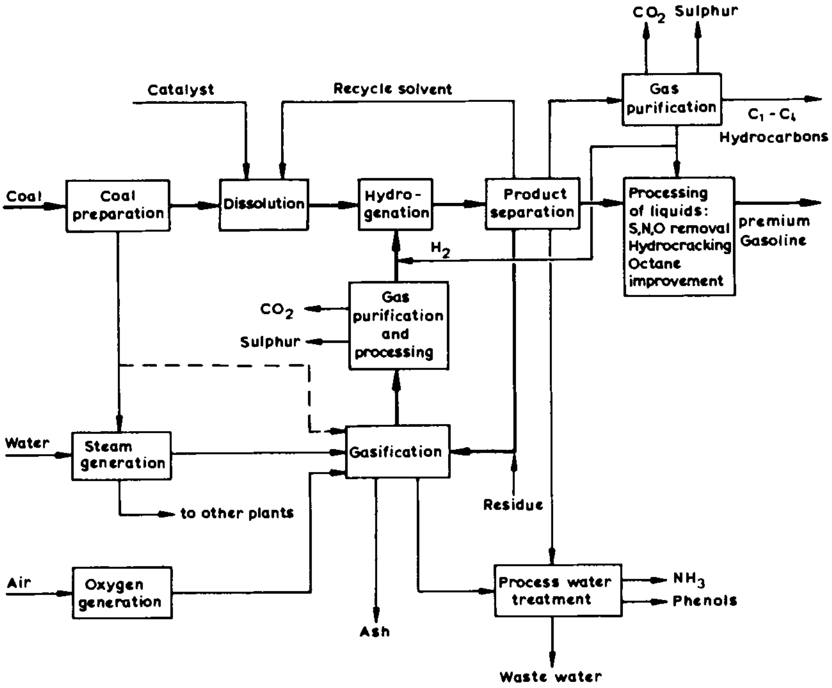


FIG. 6.12. Simplified coal liquefaction plant flow sheet.

During hydrogenation, performed in the presence of oil acting as the hydrogen donor, finely ground coal is dissolved in a recycled solvent. Catalyst is added to this mixture and the coal-oil slurry is fed, together with hydrogen, into the high-pressure hydrogenation reactor. The primary product separation yields solvent for recycling, solid residue, C_1 to C_4 gases and coal-oil. The solid residue is then gasified with steam and oxygen for the production of hydrogen. The product gas is purified, the hydrogen is recycled and the hydrocarbons can be used as fuel gas of high calorific value. The hydrogenation reaction is performed in an extended, upright tubular reactor constructed from high-alloy steel (Fig. 6.13). The coal slurry and hydrogen mixture is fed from the bottom into the reactor. The reaction products are withdrawn from the top of the reactor. Inlets are provided for quenching the exothermic hydrogenation reaction with cold hydrogen. Three reactors are run in line. In the 1940s industrial plants used reactors with an internal diameter of 1 m and a height of 18 m (Honing and Donath, 1958).

The reactor used in the 200 t coal day^{-1} pilot plant of Ruhrkohle AG and Veba AG at Bottrop exhibits the same height:diameter ratio and scaling up is intended for future industrial plants (Wolowski, 1982). The plant was commissioned in 1981 and by June 1982 had recorded about 2000 hours of operation. The design parameters stem from the pilot plant of Bergbau-Ferschung and most were confirmed throughout its operation (Wolowski and Hosang, 1982). Design studies are now under way for commercial plants with coal throughputs of 6×10^6 t yr^{-1} (Langhoff, Durrfeld and Wolowski, 1981) and about 1×10^6 t yr^{-1} (Wolowski, 1982) but falling oil prices have halted plans for such commercial plants. A second pilot plant for coal hydrogenation (capacity 6 t coal day^{-1}) has been constructed and operated by Saarbergwerke AG. The objective of this plant is to obtain operational experience as a prelude to a 2×10^6 t yr^{-1} industrial plant (Würfel, 1981). For hydrogenation of lignite a small pilot plant (throughput 0.5 t lignite day^{-1})

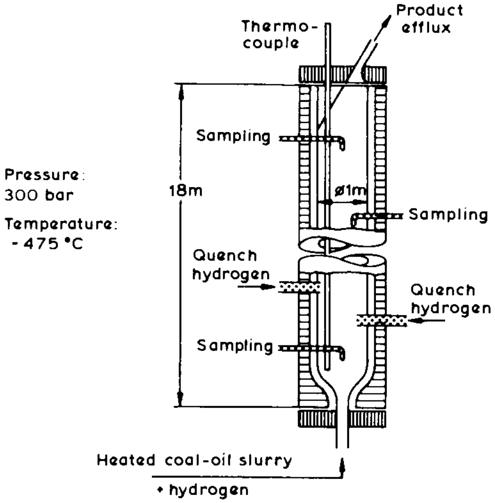


FIG. 6.13. Reactor for coal hydrogenation.

based on the Bergius-Pier technology is being run by Rheinbraun. Improvements compared with an older technique are summarized in Table 6.4. The development schedule for the design and construction of commercial plants is shown in Table 6.5.

TABLE 6.4. *Comparison of conditions and results for the hydrogenation of lignites (Teggers, Lenz and Schrader, 1981)*

	Old techniques (1943)	New techniques (1980)
Process pressure (bar)	710	300
H_2 partial pressure (bar)	510	270
Temperature of reaction (°C)	478	460
Residue treatment	Centrifugation and carbonization	Distillation and gasification
Coal throughput ($t\,m^{-3}\,h^{-1}$)	0.4	0.6
H_2 consumption (% by weight)	6.5	6.0

TABLE 6.5. *Stages of development of lignite hydrogenation in the Federal Republic of Germany (Teggers, Lenz and Schrader, 1981)*

Status of plant construction	Coal feed[a] ($t\,yr^{-1}$)	Production ($t\,yr^{-1}$)
Commercial-scale plants 1941–4	3.75×10^6	250×10^3
Plant development unit[b]	75	18
Pilot plant[b]	120×10^3	22×10^3
Demonstration plant	3.5×10^6	400×10^3

[a] raw brown coal with 60% moisture.
[b] external supply of energy + H_2.

The basis of the EDS process (Epperly and Taunton, 1979; Epperly, Plumlee and Wade, 1981) is the use of an externally hydrogenated donor solvent, produced by a catalytic hydrogenation of part of the coal-derived liquid products. Options available with this process are shown in Fig. 6.14. Figure 6.15 describes the EDS coal liquefaction pilot plants. The larger one of the two was commissioned in 1980. During 1980 and 1981 the plant was run for about 4000 hours. Further runs were carried out in 1982 with the recycling of liquefaction bottoms. The behaviour of six different coals during hydrogenation (lignites, sub-bituminous and bituminous from the USA and Australia) was investigated. Typical yields of liquefaction products (with more than three carbon items) are shown in Fig. 6.16, from which it may be seen that product yields can be improved by bottom recycling. The external

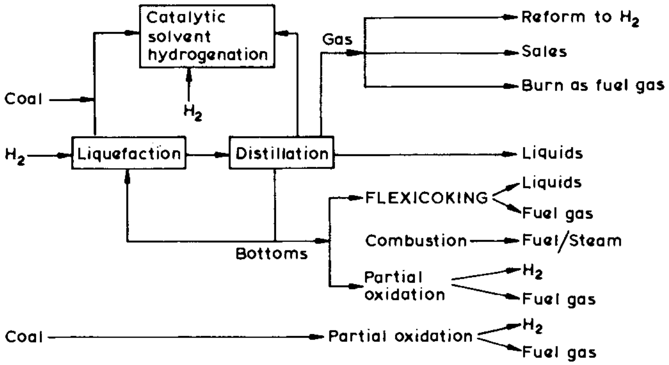


FIG. 6.14. Options available with the EDS process (Taunton, Trachte and Williams, 1981).

Reproduced from Smith, A. and Jones, B. FUEL, *1984*, 25, 99–108, by permission of the publishers, Butterworth & Co (Publishers) Ltd. ©.

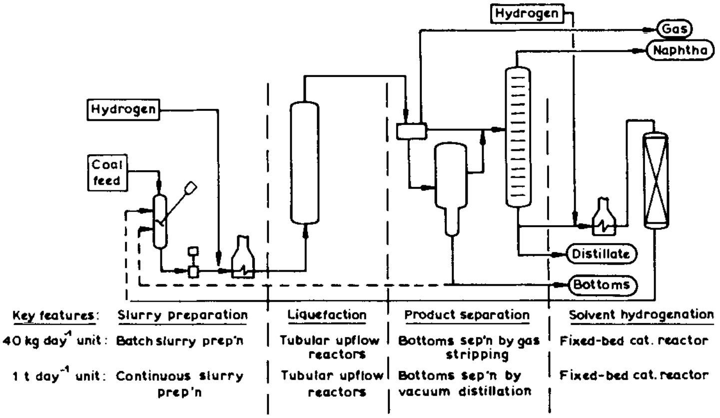


FIG. 6.15. EDS coal liquefaction pilot plants (Taunton, Trachte and Williams, 1981).

Reproduced from Smith, A. and Jones, B. FUEL, *1984*, 25, 99–108, by permission of the publishers, Butterworth & Co (Publishers) Ltd. ©.

and separate hydrogenation step allows precise adjustment of solvent donor hydrogen in order to achieve the maximum conversion of any coal. The ability of the solvent to transfer hydrogen to the coal slurry is defined by the so-called solvent quality index (SQI). It has been shown that for maximum conversion of less reactive coals solvents with a high SQI are required. A further result obtained from the operation of the plant is that processing of lower-rank coals (lignite and sub-bituminous coals) is generally more difficult than of higher-rank bituminous coals. This is thought to be due to higher oxygen contents in the coal matrix and/or finely distributed calcium (Taunton, Trachte and Williams, 1981).

An H-coal process demonstration plant (coal throughput 160 t yr^{-1}) was brought into operation in May 1980. The process is based on the use of a

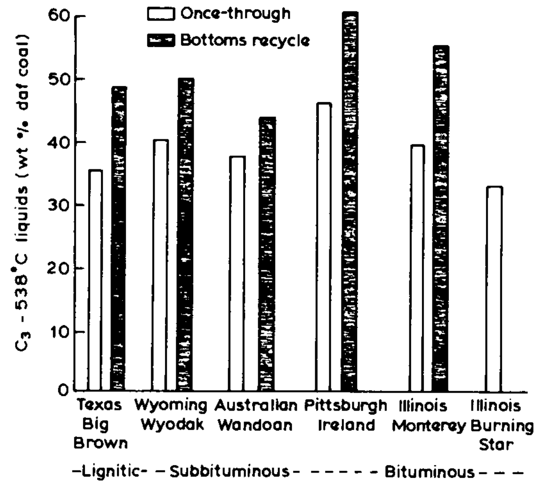


FIG. 6.16. Increased liquid yields obtained with bottoms recycle compared with once-through operation (Taunton, Trachte and Williams, 1981).

Reproduced from Smith, A. and Jones, B. FUEL, *1984*, 25, 99–108, by permission of the publishers, Butterworth & Co (Publishers) Ltd.©.

granular catalyst in a so-called ebullated bed. The objective is to produce a coal-oil called syncrude. Test runs were successful with different US coals (Langhoff, Durrfeld and Wolowski, 1981).

The main developments in coal liquefaction technologies in the USSR in recent years have been directed towards attaining coal hydrogenation under decreased pressure (10 MPa rather than 30–60 MPa). Lower pressure systems not only reduce costs of construction but makes fuller use of a wide range of existing machine building techniques.

Decreased pressure hydrogenation is achieved by introducing liquid hydrogen donors—either of coal or oil origin—and by the use of more active catalysts. This then requires improved methods of catalyst regeneration and methods of processing the slurry that is the coal residue from liquefaction processes. Developmental tests need to take account of a wide range of coal characteristics and coals ranging from high rank bituminous with low (2.6 %) ash content, bituminous coals with higher ash contents (14.4 %) and low-ranking coals with a low ash content (5 %) and others with high moisture contents (21–40 %) have all undergone investigation.

The main steps in the production of synthetic liquid fuel from coal in the USSR are outlined in Fig. 6.17. At present (1985) this plant is operating at 1000 t day^{-1}.

Moisture in coal prevents solvent penetration, lowers the partial pressure in the steam-coal paste reaction mixture and increases the volume of process waste waters. For these reasons effective methods of predrying pulverized coal have been sought without increasing coal oxidation.

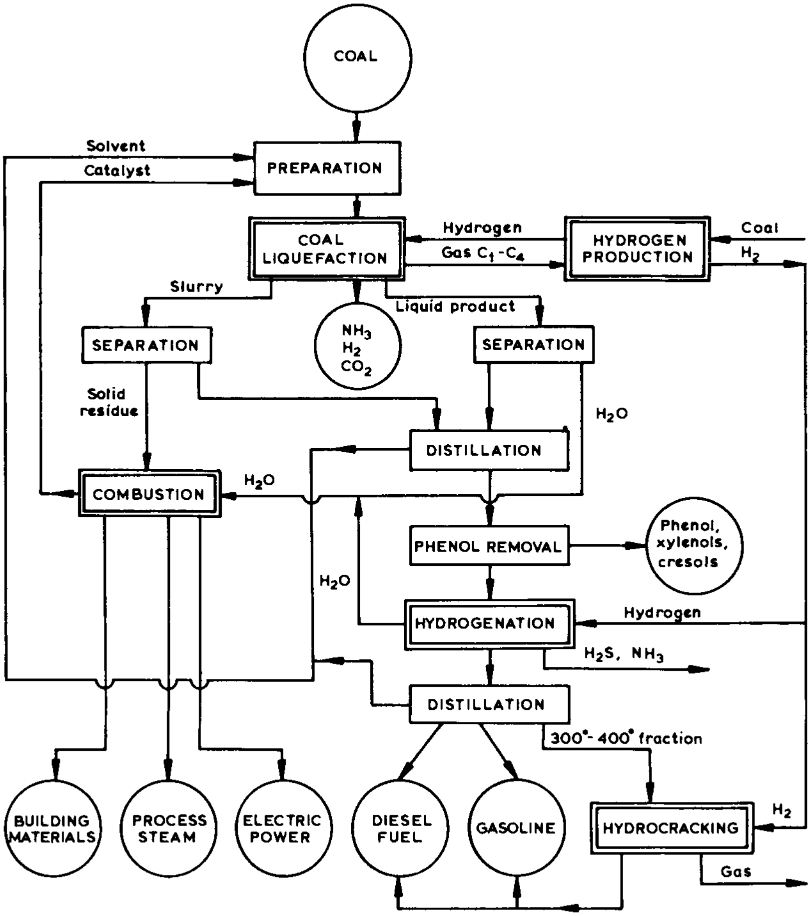


FIG. 6.17. Synthetic liquid fuel production from coal.

Thermal treatment of the coal in vortex chambers has increased the effectiveness of water removal (Zverev *et al.*, 1981; Gagarin, Smirnova and Markina, 1979). A system using two or three vortex chambers with an overall capacity of up to 10 t of coal h^{-1} is shown in Fig. 6.18. It enables brown coal to be processed to produce a feedstock with predetermined moisture characteristics. Table 6.6 shows the results of a four-stage treatment of such a coal.

Molybdenum compounds and bivalent metals are most effective catalysts for the production of low-molecular oils at lower process pressures (Krichko, Lebedev and Farberov, 1978). It has been found that the method of catalyst addition influences the hydrogenation. Combination with dry coal by vibropulverization or as an emulsion with elemental sulphur gives the best results.

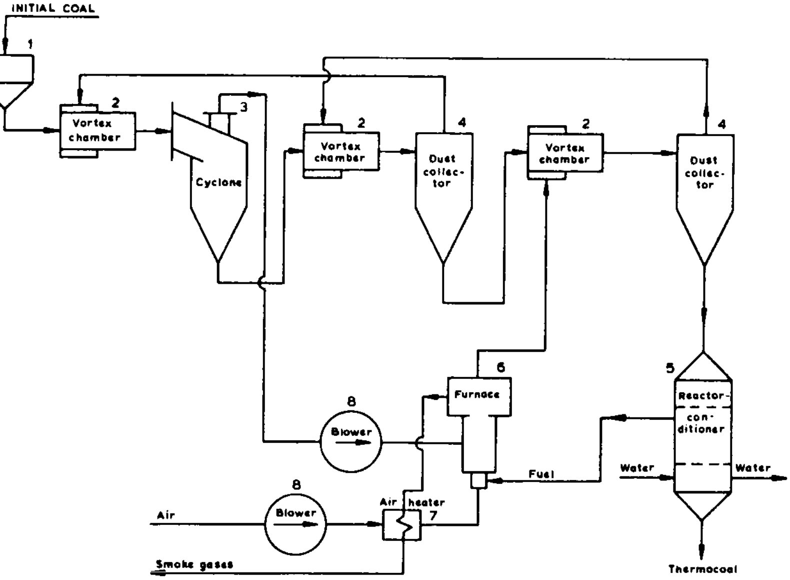


FIG. 6.18. Thermal treatment of coal (predrying in vortex chambers).

TABLE 6.6. *Characteristics of brown coal from the Kansk-Achinsk basin (Irsha-Borodinski deposit) before and after four drying stages*

Index	Initial coal	The product at stages of the thermal drying system (°C)			
		290	400	430	460
% yield	–	66.6	58.0	54.6	51.4
% moisture content (W^a)	30.5	0.5	0.2	–	–
% ash content (A^d)	6.7	7.0	7.4	8.3	8.6
% volatile matter (V^{daf})	48.8	45.4	36.6	32.7	27.1
% elemental content					
C^{daf}	72.91	73.37	76.45	72.22	81.60
H^{daf}	4.89	4.80	4.60	4.60	3.73
N^{daf}	0.86	0.91	1.04	1.14	1.13
S_{com}	0.24	0.19	0.25	0.25	0.23
Heat of combustion (kcal kg^{-1})	3900	6050	6160	6460	6430
Size analysis:					
% (diameter in mm)					
>3.2	1.4	–	–	–	–
3.2–2.5	2.5	1.8	1.2	0.9	1.1
2.5–1.0	27.1	22.6	7.7	7.1	6.1
1.0–0.4	34.8	41.5	48.5	44.4	36.8
0.4–0.2	13.7	14.9	22.3	22.8	30.2
0.2–0.16	4.3	3.5	5.3	3.0	6.0
0.16–0.10	4.9	3.8	6.6	7.2	8.5
0.10–0.063	4.2	3.9	4.6	5.8	5.6
<0.063	7.1	8.0	3.8	8.8	5.7

The comparative effectiveness of various catalysts, added in different ways, is shown in Table 6.7.

The addition of organic compounds, to make a paste, such as anthracene, quinoline, γ-picoline and others mediates transfer of hydrogen to the coal at low pressures in the presence of metallic catalysts. Some experimental work with various solvents acting as hydrogen donors has shown that recirculation of the paste results in deterioration. For this reason a scheme has been developed (Fig. 6.19) using a coal:petroleum ratio of 70:30.

TABLE 6.7. *The effect of difference catalysts and the method of addition on coal hydrogenation at 10 MPa and 425°C*

Catalyst	Method of addition	% organic matter conversion	Hydrogen consumption (%)
Bituminous coal (Kuzbass):			
Without catalyst	–	17.7	0.6
Mo, 0.2%	impregnation	23.6	0.8
Mo, 1.0%	impregnation	66.6	1.6
Fe^{+3}, 1%	impregnation	60.7	2.7
Mo, 0.2% + Fe^{+3}, 1%:	impregnation	88.8	2.7
	powder	54.0	2.8
	powder + vibro-pulverization	86.7	3.9
Brown coal (Kansk-Achinsk):			
Mo, 0.005% + sulphur	emulsion	77.3	1.5
Mo, 0.01% + sulphur	emulsion	92.3	1.7
Mo, 0.05% + sulphur	emulsion	92.5	1.9

As there is a petroleum deficiency, distillate fractions (BP 300–400 °C) from the coal liquefaction process are extracted to be cycled as a hydrogen donor (1.0–1.5% H). Coal-oil paste stability differs according to the coal used.

Development work on the low-pressure system results in a coal-oil paste with hydrogen (after heating within a heat exchanger and tube furnace) being delivered, cold, to a reactor where liquid products and gases such as HN_3, H_2S, CH_4 are formed. Because of the pressure reduction in the process large capacity reactors can be used and a throughput of 1000 t h^{-1} of coal-oil paste obtained. Table 6.8 gives some results of coal hydrogenation using different coals from the USSR.

As the separation of the solid residue from the liquid product presents a number of difficulties (the filtrability of the mixture being low), a two-stage alternative has been developed in the USSR. Centrifuging to remove up to 30% of the solid residue is followed by vacuum distillation (Fig. 6.20). By burning the solids, reclaimed molybdenum is recovered (Pisarev, Krichko and Ivanov, 1975). The heat generated is not wasted but used in the process. The

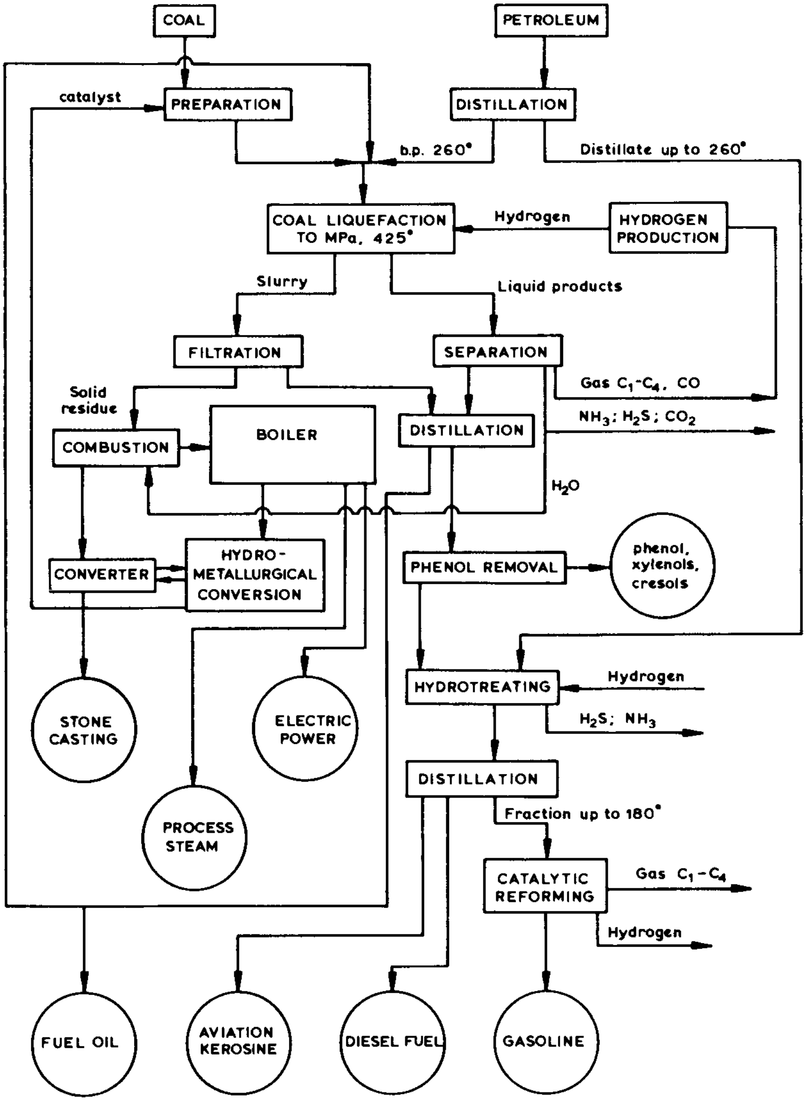


FIG. 6.19. Synthetic liquid fuel production from coal and petroleum developed by the Fossil Fuels Institute, USSR Academy of Sciences, Moscow.

molybdenum is oxidized to MoO_3 and sublimes as it is removed from the furnace with other gas streams. Calcium and magnesium minerals complicate this by reacting with MoO_3 at temperatures around 500 °C to form $CaMoO_4$ and $MgMoO_4$. A cyclone furnace allows high temperatures and the efficient system of fly ash removal and tests for optimizing molybdenum recovery have

TABLE 6.8. *Coal hydrogenation at 10 MPa from different coalfields in the USSR*

Source of coal	% ash	Degree of coal organic matter conversion (%)	H_2 % consumption	Liquid production yield (%)
Brown coal				
Kansk-Achinsk:				
Nasarovskoe deposit	7.59	92.4	3.3	87.2
Irsha-Borodinskoe deposit	7.3	90.2	2.8	85.9
Abanskoe deposit	8.42	92.5	3.2	82.9
Bituminous coal				
Kuzbass:				
The 7th Nojabrja mine	5.65	92.1	2.0	86.6
Kolchuginskaja mine Zurinski seam	3.58	98.2	1.6	86.0
Polysaevskaja mine	10.84	88.8	2.7	88.0
Gramoteinski opencast mine, Krasnoorlovski seam	4.27	89.3	2.0	84.8
Donbass:				
XXVIthe Congress of CPSS mine	2.6	87.1	2.1	71.1
Industrial Association "Sakhalinugol":				
Dolinskaja mine	14.35	93.0	2.0	78.2

been carried out (Ivanov, Smetannikov and Pisarev, 1978; Avgushevich, Lukin and Kaslina, 1975).

The liquid products of brown coal hydrogenation contain considerable quantities of sulphur, nitrogen and unsaturated compounds which together make it unsuitable as a saleable, stable motor fuel oil. The product must, therefore, undergo hydrorefining, catalytic reforming and hydrocracking (Pisarev, Krichko and Ivanov, 1975). The treatment process given to distillate with a boiling point of 400°C produced from brown coal from Irsha-Borodinski is as follows. Hydrogenation of the distillate is conducted using a $Co-Mo-Al_2O_3$ catalyst at 8 MPa. In order to prevent polymerization at high temperatures, a temperature of 230–250°C is first used so that the non-stable unsaturated hydrocarbons inclined to polymerization are hydrogenated. This is followed by temperature treatment of 400°C which hydrogenates the unsaturated and partially saturated aromatic hydrocarbons and decomposes sulphur and nitrogen compounds (Tables 6.9 and 6.10).

The hydrorefined fraction (BP up to 180°C) can be used as a component of motor gasoline. If this fraction is subjected to catalytic reforming, gasoline with a higher octane number is obtained (Table 6.11). It can also be used as a feedstock for benzene, toluene and xylene extraction. Finally, using a paladium catalyst it can be used as a feedstock for hydrocracking to obtain octane numbers high enough for jet fuel.

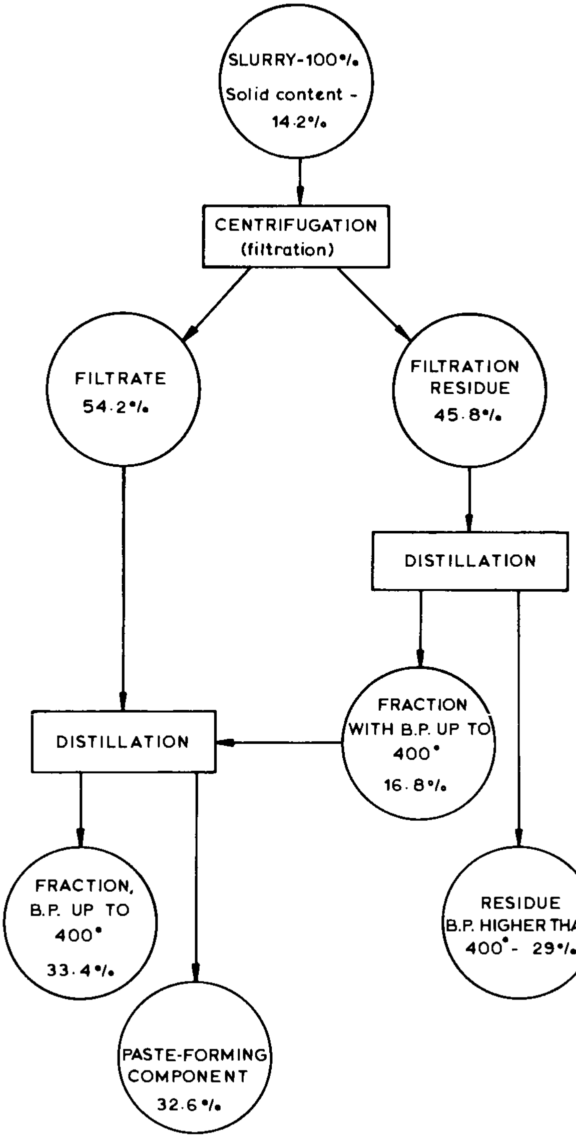


FIG. 6.20. Coal slurry processing.

4. Gasification

Gasification is a well-known technique of coal conversion. The first commercial application of modern gasification techniques was in 1926 with the commissioning of a plant using the Winkler process at Leuna. Since the

TABLE 6.9. *Coal distillate hydrogenation (BP 400°C at 8 MPa) using a Co–Mo–Al_2O_2 catalyst*

Indices	Initial material	Hydrogenate
Density (kg m^{-3})	893	846
Index of refraction (n^{20})	1.4920	1.4755
Content (% V):		
Phenols	11.9	nil
Nitrogen basis	4.9	nil
Iodine number	33.7	3.2
Elemental composition (%)		
Carbon	86.16	87.10
Hydrogen	11.50	12.81
Sulphur	0.62	0.04
Nitrogen	0.38	0.05
Oxygen (by difference)	1.34	–
Hydrogenate yield (%)	–	–
Boiling point to 180°	11.5	15.0
BP 180–300°	18.0	34.1
Hydrogen consumption (%)	–	1.7

TABLE 6.10. *Characteristics of distillate of the hydrofining fractions with a boiling point up to 400° from coal hydrogenate*

	Hydrofining fraction with boiling point, °C		
Indices	to 180	180–300	300–400
Density (kg m^{-3})	768.5	861.0	894.0
Indices of refraction (n^{20})	1.4325	1.4820	1.5003
Iodine number	1.3	1.7	2.4
Sulphur content (%)	0.01	0.03	0.04
Fractional content (°C initial boiling point)	45	175	295
10% evaporates at (°C)	102	198	320
50%	128	260	375
90%	180	297	395
Final boiling point (°C)	184	305	400
Group hydrocarbon content (%)			
paraffins + naphthenes	74.0	55.0	68.5
aromatics[a]	26.0[a]	45.0	31.5
Kinematic viscosity at 20° est.	1.2	2.8	21.9
Solidification temperature (°C)	–	–27	+14
Octane number:			
motor method	62	–	–
experimental method	72	–	–
Cetane number	–	48	–

[a] Including benzene –4.2%, toluene –10.2%, xylene –4.1%.

1930s coal gasification has been applied worldwide for the production of ammonia and hydrocarbons and also for the generation of medium-Btu gas. During the 1960s and 1970s intensive research work has been devoted to the

TABLE 6.11. *The chemical content of the aromatic components of reformed gasoline*

Compounds	Yield (%)	
	Aromatic hydrocarbons	Reformed gasoline
Benzene	16.8	13.3
Toluene	41.3	32.7
Ethyl benzene	20.2	16.0
o-Xylene	1.5	1.2
m-Xylene	4.4	3.5
p-Xylene	2.3	1.8
Isopropylbenzene	4.7	3.7
n-Propylbenzene	1.8	1.4
1,3,5-Trimethylbenzene	2.5	2.0
1,2,4-Trimethylbenzene	3.5	2.8
1,2,3-Trimethylbenzene	0.3	0.2
Diethylbenzenes	0.7	0.6
Total	100.0	79.2

further development of gasification techniques. In the first place, this means improving conventional methods with a view to greater economy. To cope with the eventuality of a shortage of natural gas, there are plans to convert coal directly into substitute natural gas (SNG) to be fed into existing natural gas pipeline networks.

An area which appears highly promising, but as yet is in its infancy, is the application of catalysts in coal gasification. Another area of development is the design and operation of combined processes of coal gasification, with subsequent combustion of the coal gas under pressure, for the environmentally acceptable generation of electricity with high thermal efficiencies. Lastly, efforts are being made to reduce the quantities of coal needed for conversion into gas by using process heat from high-temperature nuclear reactors. (A great number of survey articles have been published; von Fredersdorff, Elliott and Lowry, 1963; Jüntgen *et al.*, 1980; von Gratkowski, 1958; Falbe, 1977; Schmidt, 1966; Dolch, 1936; Schilling, Bonn and Krauss, 1979; VGB Konferenz, 1979; Elgin, 1976; Robson, 1977; Jüntgen, 1977; 1979b; van Heek, 1978; Peters and Schilling, 1977; Schulze, 1974, 1978; Jüntgen and van Heek, 1981.)

4.1. Basic Principles

Coal gasification means the complete conversion of coal into gas using heterogeneous gas solid reactions. The main process is the reaction of the carbon of coal with steam, at pressures below 10 MPa and temperatures above 750°C, to form a "synthesis gas" containing mainly CO and H_2 with smaller amounts of CO_2 and CH_4, depending on process conditions. To perform this strongly endothermal reaction in technically feasible systems, the introduction of process heat is necessary, which is possible by transfer of heat from outside (allothermal processes) or by burning part of the coal inside the reactor using oxygen (autothermal processes). This latter process is the basis for the production of hydrogen, ammonia, methanol, hydrocarbons using the Fischer-Tropsch synthesis and methane using the so-called catalytic methanation reaction. Another method for converting coal into a gas is the direct exothermal conversion of coal with hydrogen into methane at a similar range of pressure and temperatures as mentioned above. This reaction can also be combined with the steam carbon reaction and in this way leads to a direct conversion of coal into SNG.

Coal contains not only carbon but also volatile matter, which is released during the heating up of coal in the gasification reactor by the so-called pyrolysis reactions. The interaction of pyrolysis and gasification of coal is mainly dependent on grain size and rate of heating of the feed coal. At low rates of heating, the pyrolysis products are released first and the remaining char reacts with the gasification agent later on at high temperatures, yielding gaseous products from the carbon. In this case, if a countercurrent of gas and solids in the gasification reactor is induced, the product gas contains pyrolysis products such as tar, phenols and higher hydrocarbons. At high rates of heating, the pyrolysis reaction is shifted to higher temperatures and in this way both—pyrolysis products at the outer grain shell and carbon—react with the gasifying agent to form the gasification products H_2, CO, CO_2, CH_4, and less or no pyrolysis products are found in the product gas.

The nature of the reaction products of coal gasification is primarily determined by the chemical equilibria which are established, especially at high temperatures, and sufficient residence times of coal and products in the gasification reactor. Thus a high pressure and a low temperature increase the methane yield and decrease the hydrogen and the carbon monoxide yield. Much information exists on this topic (Hedden, 1962; Hedden and Mienkina, 1965; Peters, 1976; van Heek, Jüntgen and Peters, 1973; Feistel, van Heek and Jüntgen, 1978; Yoon, Wei and Denn, 1978; Jüntgen and van Heek, 1979a; Jüntgen, 1979a).

4.2. *Commercial Processes*

Table 6.12 outlines the gasification processes most commonly used, and Tables 6.13 and 6.14 list some of the full-scale gasification plants that have been built in the period 1940–1978. The moving-bed type reactor is used in the Lurgi pressure gasification process—the only process which has so far been

TABLE 6.12. *Typical commercial gasification processes*

	Operating principle		
	Moving bed	Fluidized bed	Entrained bed
Typical commercial process	Lurgi pressure gasification	Winkler process	Koppers-Totzek process
Reaction conditions	P up to 35 bar T Dry pyrolysis 400°C T Gasification 800–1000°C T Combustion 1000°C	$P = 1$ bar $T = 1000°\text{C}$	$P = 1$ bar $T = 1800°\text{C}$
Coal type	Lignite Medium-caking bituminous	Lignite Low-caking bituminous coal	Lignite Bituminous coal
Particle size of coal	Relatively large; optimum size range 6–40 mm	Large to medium; optimum size size range 0–8 mm	Fine crushing required; 80–90% < 0.1 mm
Ash removal	Dry	Dry	Slag
Steam consumption	High	Medium	Low
Oxygen consumption	Low	Medium	High
Gas composition			
CH_4	High	Medium	Low
CO	Low	Medium	High
Tars, phenols	+	–	–
Gas production pergasifier	46,000 $Nm^3 h^{-1}$ raw gas 30,000 $Nm^3 h^{-1}$ purified gas (SASOL II)	17,000 $Nm^3 h^{-1}$ 20,000 $Nm^3 h^{-1}$	20,000 $Nm^3 h^{-1}$ (2 burners) 50,000 $Nm^3 h^{-1}$ (4 burners)
Application	Town gas production Synthesis gas for F-T synthesis + CH_3OH synthesis	Synthesis gas for chemical industry	Synthesis gas for NH_3 production

carried out using high pressure—the fluidized-bed principle is used in the Winkler process and the entrained-bed type reactor in the Koppers-Totzek process. These processes are classified according to the conditions of the reaction, which leads to very different temperatures in the gasifiers. In the moving bed, changes in temperature and pressure have a considerable influence on the development of the reaction. In the Winkler gasifier, the influence of pressure and temperature is less marked. In the Koppers-Totzek gasifier, increases in temperature have hardly any influence.

TABLE 6.13. *Dry ash Lurgi commercial installations using the West German Lurgi technology*[a] *(Ghassemi, Crawford and Quinlivan, 1979)*

Plant no.	Location	Year	Type of coal	Gasifier ID (m)	Capacity (Nm^3 day^{-1} × 10^6)	No. of gasifiers
1	Bohlen, Central Germany	1940	Lignite	2.6	0.25	5
2	Bohlen, Central Germany	1943	Lignite	2.6	0.28	5
3	Most, CSSR	1944	Lignite	2.6	0.21	3
4	Zaluzi-Most, CSSR	1949	Lignite	2.6	0.25	3
5	Sasolburg, South Africa	1954	Sub-bituminous with 30% ash and more	3.7	4.20	9
6	Dorsten, West Germany	1955	Caking sub-bituminous with high chlorine content	2.7	1.54	6
7	Morwell, Australia	1955	Lignite	2.7	0.62	6
8	Daud Khel, Pakistan	1957	High volatile coal with high sulphur content	2.7	0.14	2
9	Sasolburg, South Africa	1958	Sub-bituminous with 30% ash and more	3.7	0.53	1
10	Westfield, Great Britain	1960	Weakly caking sub-bituminous	2.7	0.78	3
11	Jealgora, India		Different grades	not available	0.03	1
12	Westfield, Great Britain	1962	Weakly caking sub-bituminous	2.7	0.25	1
13	Coleshill, Great Britain	1963	Caking sub-bituminous with high Cl content	2.7	1.3	5
14	Naju, Korea	1966	Sub-bituminous with 30% ash and more	3.7	2.10	3
15	Sasolburg, South Africa	1966	Sub-bituminous with 30% ash and more	3.7	2.10	3
16	Luenen, FRG	1970	Sub-bituminous	3.4	1400 × 10^6 Btu hr^{-1}	5
17	Sasolburg, South Africa	1973	Sub-bituminous with 30% ash and more	3.7	5.32	3
18	Sasolburg, South Africa	1978	Sub-bituminous with 30% ash and more	4.0	42	36

[a] A parallel development of the Lurgi process has been carried out by the German Democratic Republic since the Second World War and several commercial plants using the East Germany technology currently exist in Eastern European countries. Examples include the Gaskombinant Schwarze Pumpe, East Germany with 24 gasifiers (3.6 m ID).

TABLE 6.14. *Gasification plants using the Koppers-Totzek process (Ghassemi, Crawford and Quinlivan, 1978)*

Location	Fuel	Number of gasifier units	Capacity ($CO+H_2$ in 24 hr)	Use of synthesis gas	Year of order
Charbonnages de France, Paris, Mazingarbe Works (PdC) France	Coal dust, coke oven gas, tail gas	1	75,000–150,000 Nm^3 2,790,000–5,580,000 SCF	Methanol and ammonia synthesis	1949
Typpi Oy, Oulu, Finland	Coal dust, oil peat	3	140,000 Nm^3 5,210,000 SCF	Ammonia synthesis	1950
Nihon Suiso Kaisha, Ltd., Tokyo, Japan	Coal dust	3	210,000 Nm^3 7,820,000 SCF	Ammonia synthesis	1954
Empresa Nacional "Calvo Sotelo" de Combustible Liquidos y Lubricantes, SA, Madrid, nitrogen works in Puentes de Garcia Rodriquez, Corunna, Spain	Lignite dust	3	242,000 Nm^3	Ammonia synthesis	1954
Typpi Oy, Oulu, Finland	Coal dust, oil, peat	2	140,000 Nm^3 5,210,000 SCF	Ammonia synthesis	1955
SA Union Chimique Belge, Brussels, Zandvoorde Works, Belgium	Bunker-C-oil plant convertible for coal dust gasification	2	176,000 Nm^3 6,550,000 SCF	Ammonia synthesis	1955
Amoniaco Portuguese SARL, Lisbon, Estarreja Plant, Portugal	Heavy gasoline, plant extendable to lignite and anthracite dust	2	169,000 Nm^3 6,300,000 SCF	Ammonia synthesis	1956
The Government of Greece; The Ministry of Coordination, Athens; nitrogenous fertilizer plant, Ptolemais, Greece	Lignite dust, bunker-C-oil	4	629,000 Nm^3 23,450,000 SCF	Ammonia synthesis	1959

TABLE 6.14 (*Contd.*)

Location	Fuel	Number of gasifier units	Capacity ($CO+H_2$ in 24 hr)	Use of synthesis gas	Year of order
Empresa Nacional "Calvo Sotelo" de Combustibles Liquidos y Lubricantes, SA, Madrid, nitrogen works in Puentes de Garcia Rodriquez, Corunna, Spain	Lignite dust or naphtha	1	175,000 Nm^3 6,500,000 SCF	Ammonia synthesis	1961
The General Organization for Executing the Five Year Industrial Plan, Cairo; nitrogen works of Societé el Nasr d'Engrais et Chimiques, Attaka, Suez, UAR	Refinery off-gas, LPG, and light naphtha	3	778,000 Nm^3 28,950,000 SCF	Ammonia synthesis	1963
Chemical Fertilizer Company Ltd., Thailand, synthetic fertilizer plant at Mao Moh, Lampang, Thailand	Lignite dust	1	217,000 Nm^3 8,070,000 SCF	Ammonia synthesis	
Azot Sanayii TAS, Ankara, Kutahya Works, Turkey	Lignite dust	4	775,000 Nm^3 28,850,000 SCF	Ammonia synthesis	1966
Chemieanlagen Export-Import GmbH, Berlin für VEB Germania, Chemieanlagen and Apparatebau, Karl-Marx-Stadt, VEB Zietz Works	Vacuum residue and/or fuel oil	2	360,000 Nm^3 13,400,000 SCF	Raw gas to produce hydrogen for hydrogenation	1966
Kobe Steel Ltd., Kobe, Japan, for Industrial Development Corp., Zambia, at Kafue near Lusaka, Zambia, Africa	Coal dust	1	214,320 Nm^3 7,980,000 SCF	Ammonia synthesis	1967
Nitrogenous Fertilizers Industry SA, Athens, nitrogenous fertilizer plant, Ptolemais, Greece	Lignite dust	1	165,000 Nm^3 6,150,000 SCF	Ammonia synthesis	1969

The Fertilizer Corporation of India Ltd., New Delhi, Ramagundam Plant, India	Coal dust	4 (1 of them as standby)	2,000,000 Nm^3 74,450,000 SCF	Ammonia synthesis	1969
The Fertilizer Corporation of India Ltd., New Delhi, Talcher Plant, India	Coal dust	4 (1 of them as standby)	2,000,000 Nm^3 74,450,000 SCF	Ammonia synthesis	1970
Nitrogenous Fertilizers Industry SA, Athens, nitrogenous fertilizer plant, Ptolemais, Greece	Lignite dust	1	242,000 Nm^3 9,009,000 SCF	Ammonia synthesis	1970
The Fertilizer Corporation of India Ltd., New Delhi, Korba Plant, India	Coal dust	4 (1 of them as standby)	2,000,000 Nm^3 74,450,000 SCF	Ammonia synthesis	1972
AE and CI Ltd., Johannesburg, Modderfontein Plant, South Africa	Coal dust	6	2,150,000 Nm^3 80,025,000 SCF	Ammonia synthesis	1972
Indeco Chemicals Ltd., Lusaka, Kafue Works, Zambia	Coal dust	1	220,800 Nm^3 8,220,000 SCF	Ammonia synthesis	1974
Indeco Chemicals Ltd., Lusaka, Kafue Works, Zambia	Coal dust	2	441,660 Nm^3 16,440,000 SCF	Ammonia synthesis	1975

SCF = Standard cubic feet.
Nm^3 = Normal cubic metre (0°C and 1 atm).

The Lurgi pressure gasification method has proved exceptionally versatile in its applications to date. In particular, large quantities of synthesis gas are produced worldwide for Fischer-Tropsch synthesis. A contract has also been concluded for the production of synthesis gas for methanol. There are also plants producing gas of town-gas quality. The Winkler method is generally used for producing synthesis gas in the chemical industries, and is operated in the CMEA countries predominantly on lignite. The Koppers-Totzek process is used all over the world with widely differing types of coal wherever gas has to be produced for the synthesis of ammonia. The modern plants are geared to an ammonia production capacity of 1000 tonnes day^{-1}. A number of articles have been published on special gasification processes and are itemized appropriately here (*Moving bed*: Hiller, 1975; Hoogendorn, 1976; Röbke, 1978; *Fluidized bed*: Flesch and Velling, 1962; Davy Powergas, 1973; Banichek, 1973; Franke, 1978; Anwer and Bögner, 1976; *Entrained bed*: Linke and Vogt, 1979; Volkel *et al.*, 1979; Franzen, 1977; Staege, 1976; Seipenbusch and Ruprecht, 1978; Kraaijveld, 1978; Rossbach *et al.*, 1978; *Use of nuclear reactor*: Arndt *et al.*, 1979; Jüntgen and van Heek, 1977, 1979b).

4.3. Application and Improvement of Conventional Processes

4.3.1. Moving-bed gasification

The Lurgi gasification process is the most widely used. In 1981 the SASOL II industrial complex was brought into operation. SASOL III, of similar design, was recently constructed and began operation in 1984. Both plants are designed to convert coal into motor fuels by gasification of coal to synthesis gas using the Lurgi process and conversion of this by Fischer-Tropsch synthesis operated in an entrained phase system (synthol process). The process was developed by SASOL in co-operation with Kellog. Details of the coal input and composition, the design of the gasification and gas purification processes, the synthol process and gas reforming are listed in Table 6.15. Details of the products and labour requirements are also given. A simplified flow sheet of the whole plant is shown in Fig. 6.21. These three SASOL plants are the largest coal gasification complexes in the world producing motor fuels. The plants will be extended by four additional Lurgi gasifiers. Plans for the construction of a further large coal conversion complex are in an early stage, but it is expected that different techniques will be used.

In the USA the first commercial gasification plant was under construction (Great Plains Gasification Project). It was to consist of fourteen 4-m diameter Lurgi gasifiers (two of which are standby units). The throughput capacity of the plant was to be 22,000 t h^{-1} of lignite and the final product SNG (Kuhn, 1981; Torster, Margolis and Kuhn, 1982) but its future is in doubt due to lack of government funds. In the People's Republic of China a similar plant is under construction.

TABLE 6.15. *SASOL II indirect liquefaction plant: selected design and operating data*

Coal composition		
Sub-bituminous		
Heating value, gross, dry basis 23.9 MJ kg^{-1} (10,300 Btu lb^{-1})		
Surface moisture	6 – 11 %	
Inherent moisture	5 %	
Ash (dry basis)	21.5 %	
Carbon (daf)	79.6 %	
Sulphur (daf)	1.3 %	
Hydrogen (daf)	4.3 %	
Nitrogen (daf)	2.0 %	
Oxygen (daf)	13.6 %	
Gasification		
Coal feed	8,400,000 t yr^{-1}	
Hp steam	1230 t h^{-1}	
Oxygen	8600 t day^{-1}	
Gasifiers	36 installed, diameter 4.0 m	
Raw gas	1,650,000 m^3 h^{-1}	
Rectisol gas purification		
Raw gas feed	1,650,000 m^3 h^{-1}	
Pure gas	1,100,000 m^3 h^{-1}	
Number of streams	4	
Total sulphur in pure gas	0.07 ppm	
Gas composition	*Vol %*	
	Raw gas	Pure gas
CO_2	32.0	1.5
$H_2 + CO$	57.1	84.1

Synthol plant	
Number of reactors	7
Total gas feed	1,900,000 m^3 h^{-1}
Gas reforming	
Feed gas	225,000 m^3 h^{-1}
Product gas	550,000 m^3 h^{-1} (90 % CH_4)
Oxygen consumption	3,400 t day^{-1}
Number of reformers	8
SASOL II production	
Motor fuels	1,500,000 t yr^{-1}
Ethylene	185,000 t yr^{-1}
Chemicals	85,000 t yr^{-1}
Tar products	180,000 t yr^{-1}
Ammonia (as N)	100,000 t yr^{-1}
Sulphur	90,000 t yr^{-1}
Total saleable products	2,140,000 t yr^{-1}
Plant labour requirements skilled and semi-skilled	
Production	1000
Maintenance	1800
Technical staff	160
Administrative and general staff	400
Mine	700
Unskilled	
Plant	1000
Mine	2000

CH_4	9.4	13.5
H_2S	0.7	—
$N_2 + A$	0.3	0.5
C_nH_m	0.5	0.4
Oxygen plant		
Capacity	6 units of 2300 t d^{-1} each	
Oxygen pressure	3400 kPa	
Steam plant		
Capacity	6 boilers of 540 t h^{-1} each 430°C, 4000 kPa	
Coal feed	4,400,000 t yr^{-1}	
Cooling water system		
Circulating rate	165,000 m^3 h^{-1}	
Temperature range	27 – 40°C	
Number of cooling towers	4	
Dimensions	bottom diameter 105 m, height 151 m	
Type	natural draught	

Area	
Process plant	196 ha
Tank farms	93 ha
Administration and despatch	135 ha
Central area (workshops, etc.)	33 ha
Effluent treatment	165 ha
Ash disposal	180 ha

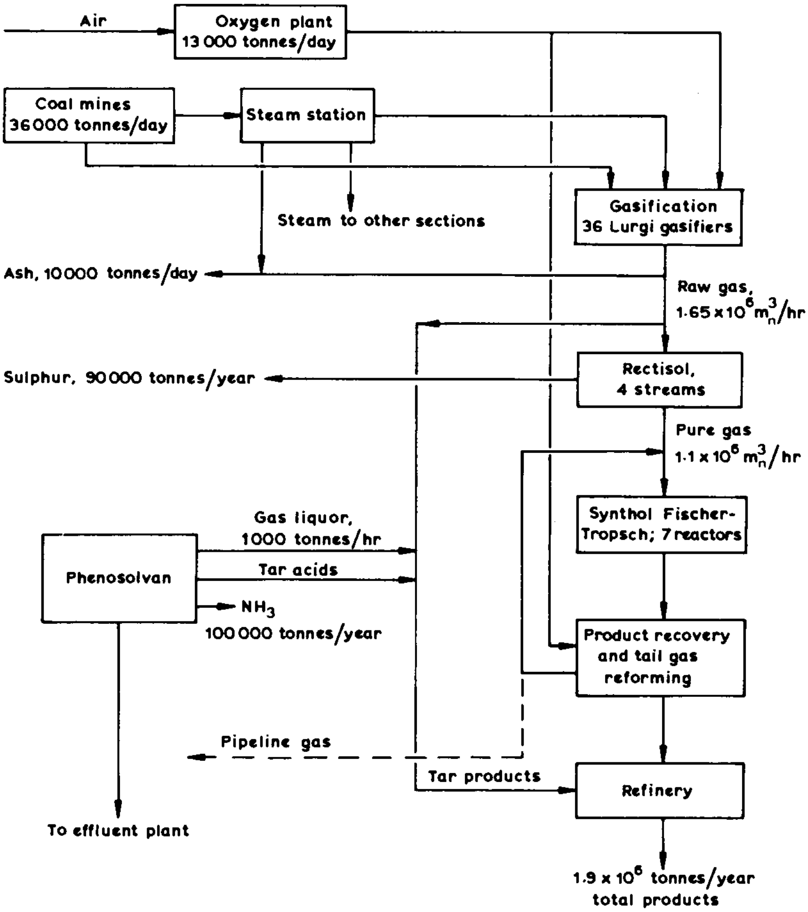


FIG. 6.21. Simplified flow sheet of Sasol II indirect liquefaction plant.

Development objectives for the Lurgi moving-bed gasification process are the increase of pressure to 100 bar and temperature above ash melting point. These are being pursued in different projects.

The pilot plant "Ruhr 100", with its gasifier of 1.5 m diameter and rated operation pressure of 100 bar, was commissioned in 1979 and has since operated for over 4000 hours. Methane concentration could be boosted to 24 % by volume and a CO_2-free, raw gas could be delivered at 90 bar pressure. Under these conditions the tar and oil content of the product gas is decreased. This process is suited for SNG production with minimized energy consumption. A drawback, however, is that only weakly caking coal can be used as feedstock (Schafer *et al.*, 1982).

The British Gas Lurgi slagging gasifier, at present under development, is run at a high temperature. The slagging gasifier's coal throughput is >300 t day^{-1}. The gasifier was run on nine different coal qualities from England, Scotland and the United States (volatile matter content ranging between 31 and 40 % on daf basis and the swelling index ranging between 1 and 7). Compared to low-temperature processes with dry ash removal, the new process exhibits the following advantages: higher throughput, lower steam consumption, less production of liquids and higher thermal efficiency. The slagging gasifier is in an advanced stage of development for commercial operation (King, 1981).

Also, smaller non-pressurized moving-bed gasifiers are under development or being constructed (Dorstewitz and Kaimann, 1981). The so-called FW Stoic two-stage gasifier is under demonstration in Duluth, Minnesota, USA. A 3-m diameter gasifier supplies two boilers with hot fuel gas. The gasifier is run on lignite from Wyoming and Colorado, and uses atmospheric air as the gasifying agent (Shires, 1981).

4.3.2. *Fluidized bed gasifier*

Two kinds of processes are being developed: pressurized fluidized-bed reactors for the production of synthesis gas and high-pressure fluidized-bed gasifiers for hydrogasification.

The development of pressurized fluidized-bed gasification for lignites run at 10 bars has reached an advanced stage of development in the Federal Republic of Germany. A 1.3 t h^{-1} pilot plant has shown that throughput can be increased by raising operational pressures and temperatures. A commercial plant for the production of synthesis gas from lignite is under construction at Wesseling, Federal Republic of Germany (Teggers, Lenz and Schroder, 1981). This plant runs four gasification lines each having a capacity of 55 t h^{-1}.

A fluidized bed gasifier, run on bituminous coal at approximately 5 bars, is being developed by IGT (IGT U-gas process). By using a novel technique for ash agglomeration in the fluidized bed, high carbon utilization is achieved. At Memphis, USA, a demonstration plant for production of 50,000 GJ day^{-1} of industrial fuel gas has reached the design phase. The medium-calorific gas is distributed over a 24-km pipeline network. The plant is comprised of four gasification lines, three in regular operation and one as standby. The internal diameter of the gasifier beds is 3.75 m, whilst above the fluidized bed it is 5 m (Shires, 1981). Finally, the EXXON catalytic gasification process, run in a pressurized fluidized bed gasifier, should be mentioned. This process has now reached the demonstration phase.

The hydrogasification of lignites at approximately 990 °C and 100 bars has been demonstrated in a pilot plant at Wesseling (throughout 0.4 t h^{-1} of lignite). The process yielded a product gas with a maximum CH_4 content of 48 % by volume. The conversion rate was approximately 60 % for a residence time of 30 min. The residual char can be used for the production of synthesis gas by steam gasification in a pressurized fluidized-bed. In this way the hydrogen necessary for hydrogasification can be produced. A 24 t h^{-1} demonstration plant, run on lignite, was under construction and began operation early in 1983 (Teggers, Lenz and Schroder, 1981).

4.3.3. Entrained phase gasifier

A pressurized version of an entrained phase gasifier is under development. The feedstock is pressurized in a lockhopper system and is subsequently injected pneumatically. Shell runs two pilot plants, one of 6 t day^{-1} capacity in Amsterdam and another of 150 t day^{-1} capacity in Hamburg. The latter has operated for over 3000 hours on German and American coals. A coal conversion rate of 99 % was obtained, the CO_2 concentration in the product gas ranging between 1 and 2 % by volume. A high degree of temperature control was achieved. The construction of two 1000 t day^{-1} demonstration plants in the Federal Republic of Germany and in the Netherlands was cancelled. However, research and development will be pursued (Vogt and Eckstein, 1982).

An alternative way of feeding finely ground coal into a gasifier under pressure is to employ a coal/water-slurry feed system. A gasification technique similar to the well-known oil gasification can be run on such a feedstock. This is taken in the Texaco coal gasification process, at present under development in the United States and the Federal Republic of Germany. The 180 t day^{-1} pilot plant of Ruhrchemie AG/Ruhrkohle AG, functional since 1978, has now recorded 11,000 hours of operation. A total of 6000 t of various coal qualities have been converted in this plant. Twelve of these coals had ash contents between 6 and 28 % by weight, volatile matter contents between 16 and 42 % by weight, sulphur contents between 0.8 and 3.9 % by weight, ash-melting temperatures between 1280 and 11,500 °C and Hardgrove indices (for grindability) ranging between 47 and 100. The SAR ("Synthesegasanlage Ruhr") demonstration plant has also been designed with a throughput of 700 t h^{-1} producing 50,000 Nm^3 h^{-1} synthesis gas. The plant will be constructed on the premises of Ruhrchemie AG in Oberhausen and the synthesis gas produced will mainly be used for the manufacture of aldehydes from olefines by the so-called oxosynthesis process (Cornils *et al.*, 1982). Two other similar demonstration plants are operating in the United States. The Tennessee Valley Authority plant was recommissioned in April 1982 after modifications. For the first 3 months the modified plant recorded a conversion of 5000 t of coal in 590 hours of operation.

Another plant in which coal is gasified using air is run by the DOW Chemical Company. Two commercial plants are under construction: the Tennessee Eastman Project in Kingsport (feed rate 900 t day^{-1}), that started up in 1983, and the Cool Water Coal Gasification Programme in Southern

California (coal feed 1000 t day^{-1}), opened in 1984. The syngas from the Tennessee Eastman plant will be used for the production of a wide variety of chemical products. The Cool Water Project is designed to demonstrate the gasification of Utah bituminous coal in a 100 MW combined cycle electric power plant (Schlinger *et al.*, 1982).

4.4. *New Processes Under Development*

4.4.1. *Gasification by nuclear process heat*

This process uses heat supplied by a high-temperature nuclear reactor for endothermal gasification in an allothermal fluidized-bed reactor. Intensified development work on this configuration is in progress. The following advantages are quoted: 30–40% saving of coal, reduced coal-specific emissions and lower gas production costs. A pilot plant for a coal rated input of 100–400 kg h^{-1} has been run to systematically study the interaction between the reaction kinetics of gasification and the heat transfer from a helium stream (supplied from the nuclear reactor) to the heat exchanger which is immersed in the fluidized bed. The results obtained will be used in the design of a fluidized-bed gasifier scaled up to industrial size. A special development of this configuration is a new jet feeder for feeding strongly caking coals into the process. Technical problems with the heat exchanger are being solved. It can be demonstrated that under the gasification conditions oxidized layers form on the outside wall of the heat exchanger tubes, thus inhibiting further corrosion. The state of development of the process, as well as the safety-related and economic features of further plants, are subject to investigation (van Heek, Jüntgen and Peters, 1982).

4.4.2. *Catalytic gasification*

The use of catalysts in coal gasification has been known from patent applications (Tessie du Motay and Marechal, 1867) for more than 100 years. Systematic investigation and research work on a small scale date back to the 1920s (Taylor and Neville, 1921; Kroger, 1939; Veraa and Bell, 1978; McKee and Chatterji, 1978; Harker, 1960; Otto, Bartosiewicz and Shelev, 1979; Wigmans *et al.*, 1981). In commercial gasification processes, however, catalysts have not so far been used.

Thorough discussions on catalytic coal and carbon gasification took place at the International FUNCAT cogas symposium in Amsterdam in 1982 (Jüntgen, 1982c). It was agreed that alkali metals in the order given are the most active catalysts in enhancing the important carbon-steam reactions: Cs, K, Na, Rb, Li (McKee, 1983; Spiro *et al.*, 1983; McKee *et al.*, 1983). In experiments, potassium can be added to the gasification system as K_2CO_3 or KOH by dry mixing, impregnation, or dissolved in high-pressure steam. The effect of potassium compounds on the reaction rate in the systems varies with coal quality and gasification conditions. It is not yet well understood. Side reactions between catalysts and mineral components of the coal which inhibit catalytic activity are important (Sulimma, van Heek and Jüntgen, 1981; Kuhn and Plogmann, 1982). Homogeneous distribution of the catalyst in the narrow pore structure of coal and char is also significant. Evidently, potassium compounds are reduced to potassium metal by coal char carbon at temperatures below 800°C. This metal is highly mobile over the internal carbon surface, but is bound to the carbon of the char if the K/C molar ratio is lower than 0.18. This has been shown for potassium and demonstrates that the rate of vaporization of potassium in presence of this carbon–potassium complex is definitely lower than that of pure potassium (Huhn, Klein and Jüntgen, 1983). The linkages between carbon and potassium are not yet defined. Intercalation compounds, salt formation with carbon–oxygen complexes (Mims and Pabst, 1983), or chemisorption may be an explanation. This is under investigation.

The significance of reaction kinetics for plant design and the alteration of reaction kinetics by the addition of catalysts are key factors for considerations in coal gasification. From a detailed consideration of reaction kinetics it may be concluded that catalysts are only effective on gasification rates in the lower temperature range (below 1000 °C) where the rate of chemical reaction and pore diffusion control the overall gasification rate. The increase in the reaction rate achieved by catalysts can also be obtained by increasing the gasification temperature, but for conventional gasification processes this method is not attractive. The advantages obtained—less coal consumption, reduced heat loss and zero oxygen consumption—are counteracted by additional costs and unwanted side effects. It can be concluded that the use of catalysts will only result in significant advantages with processes where, for any reason, temperature increase is limited, as in allothermal gasification with limited heat carrier temperature or for SNG production processes subject to thermodynamic temperature limitations (Jüntgen, 1982b).

The overall conversion chemistry of the EXXON catalytic coal gasification process may be described by the reaction:

$$CH_{0.9}O_{0.1} + 1.2H_2O \longrightarrow CH_4 + 0.7CO_2$$

for a typical bituminous coal of the molar composition $CH_{0.9}O_{0.1}$. This reaction requires low temperature and high pressures. This can be concluded from thermodynamic considerations. At 700°C and 35 bars the reaction rate is down to such a value that 10–20% (by weight) potassium addition, in the form of K_2CO_3 or KOH, becomes necessary in order to speed up and to catalyse CH_4 formation. As shown in Fig. 6.22, the reaction is run in an extended fluidized bed with recycling of a proportion of the product gas. The composition of the product reaches 31.9% H_2O, 15.7% CO_2, 22.6% CH_4, 22.4% H_2 and 7.4% CO, assuming thermodynamic equilibrium at

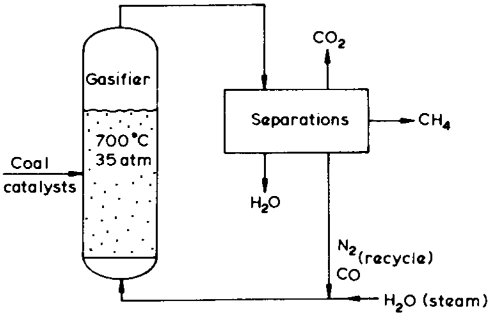


FIG. 6.22. Schematic diagram of the Exxon catalytic coal gasification process showing recycle of hydrogen and carbon monoxide (Nahas, 1983).

700 °C and 35 bars. The overall reaction is a slightly endothermic and the preheating of the fluidizing gas mixture to 800 °C and of coal to 100–300 °C is sufficient to meet the heat demand. The coal char residence time required for complete carbon conversion was found to be 6–10 hours. A 50 kg h^{-1} pilot plant has been run at Baytown, USA, and there are intentions to test a scaled-up design in a 500 t h^{-1} pilot plant at Europort, Rotterdam. The commercial plant will consist of four reactors, each with a diameter of approximately 7 m and a height of 30 m, producing methane at approximately 8×10^6 m^3 day^{-1}. The advantage of this process is that methane can be produced directly from coal in one process phase with very low heat demand (Nahas, 1983).

With gasification by means of heat supplied from a high-temperature nuclear reactor, the temperature of the gaseous heat carrier is limited. The possibility of using a catalyst for steam gasification in a fluidized bed has been examined. Calculations, based on experimental results obtained in the laboratory and at pilot pant scale, show that the throughput of a large-scale nuclear gasification plant can be increased by about 65 % when using a K_2CO_3 catalyst added in quantities of 3–4 % by weight of the coal feed. In this way gas production costs are reduced by approximately 15 %. Corrosion does not create serious problems if the initial concentration of catalyst is kept below 5 % and the temperature remains below 900 °C (Kubiak *et al.*, 1982).

Now, after a long period of fundamental research and development, these two catalytic processes are definitely promising. Investigations with respect to the recovery of the catalyst from the coal ash have given promising results. Some of the potassium components are soluble in water and therefore can be easily recovered. Al_2O_3 and SiO_2 are not soluble and create no disposal problems.

4.4.3. Other processes

There are many other developments which cannot be discussed in detail here. Gasification in the presence of liquid iron is a new process which uses technology well known in iron and steel industries. A low-pressure version is scheduled to be run at Klöckner-Werke AG, Bremen. A high-temperature version is being developed by Klöckner Humboldt Deutz, Köln (Pfeiffer and Paschen, 1981). Sumitomo Metal in Osaka uses this technology in a large pilot plant.

5. Pyrolysis-Based Conversion Processes

The fundamentals of coal pyrolysis and hydropyrolysis are discussed by Howard (1981). The kinetics of pyrolysis in pressurized and non-pressurized plant configurations are described by Jüntgen and van Heek (1979) and van Heek (1982). The pyrolysis mechanisms depend on the decomposition of the basic chemical structure of coal (Jüntgen, 1982b), as described in Section 3. Reactions between single hydro-aromatic nuclei systems take place, thus forming a highly cross-linked network, splitting off hydrogen and resulting in the formation of an aromatic char. Some of the basic pyrolysis reactions are shown in some detail in Table 6.16. The rupture of weaker and stronger chemical bonds takes place within different temperature ranges. At lower temperatures the main reactions produce tar and methane as a result of the rupture of weak bonds, whilst H_2O, CO and CO_2 are released from the decomposition of oxygen-containing structures. The main reaction in the higher temperature range is the formation of char and hydrogen by the cleavage of stronger bonds and linkage to the larger aromatic nuclei systems. Additional methane is released by the cleavage of hydro-aromatic structures or carbon-created oxygen reactions. An additional CO release then occurs as a result of the rupture of heterocyclic C–O–C structures.

This series of reactions can be viewed from the overall kinetics of pyrolysis reactions on the basis of a thermobalance at constant heating rate, as shown in Fig. 6.23. The first peak recorded at temperatures below 550 °C represents the high rate reaction of tar and CH_4 formation. The second slower reaction

TABLE 6.16. *Basic pyrolysis reactions*

Temperature range 300–500°C	 >550 °C
1. Cleavage of weaker chemical bonds of coal structure	1. Cleavage of stronger chemical bonds of coal structure
2. Distillation of volatiles (so-called tar)	2. Formation of methane from aliphatic side chains or hydroaromatic rings
3. Formation of aliphatic hydrocarbons, mainly CH_4 from aliphatic bridges	3. Formation of hydrogen and char or coke by linkage of aromatic rings to graphitic clusters
4. Formation of $H_2O/CO/CO_2$ from oxygen containing groups	4. Formation of CO from heterocyclic O

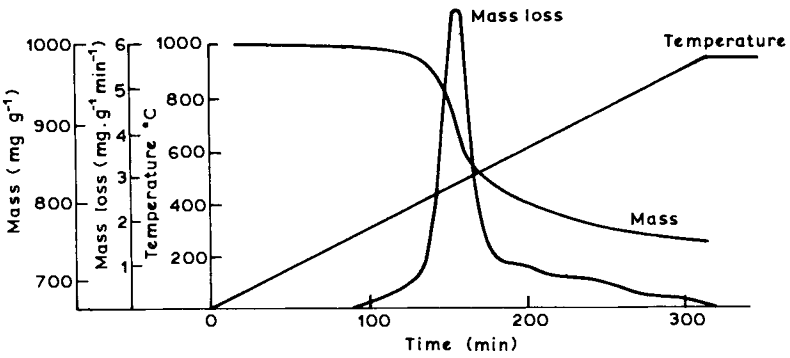


FIG. 6.23. Overall kinetics of coal pyrolysis and rate of char formation.

occurs at temperatures above 550 °C and comprises several reaction steps, with hydrogen as the main gaseous product. During hydropyrolysis, this mechanism involves hydrogen reacting with coal fragments and inhibiting char formation, thus resulting in higher yields of volatile products than obtained by pyrolysis. The main products are methane, phenols, BTX-aromatics and higher nuclei aromatics. According to Cypres *et al.* (1982), hydropyrolysis combined with char combustion on gasification is a third possible method of coal vaporization (an alternative to gasification and liquefaction). Product yields at low heating rates are given by Cypres and Furfari (1981). The influence of the heating rate on kinetics was investigated by Arendt and van Heek (1981). The formation of BTX-aromatics, as a function of temperature and hydrogen pressure, has been studied by Bunthoff *et al.* (1982). A survey of the IEA Pyrolysis Project investigations, carried out at Bergbau-Forschung and resulting in the establishment of the basic kinetic data needed for reactor design, is given by Wanzl, van Heek and Jüntgen (1982).

The state of plasma pyrolysis with regard to the production of acetylene is reviewed by Jüntgen *et al.* (1981). This process is under development and will have to compete with the old, well-known carbide route which consists of CaC_2 production in an electric arc reduction furnace at approximately 2100 °C and subsequent gasification of the CaC_2 with water, producing C_2H_2. In the new AVCO Arc Coal Process, dry pulverized coal is injected in an H_2 stream directly into an electric arc (which is stabilized by rotation in a magnetic field) and is converted directly to acetylene. A part of the product gas is recycled and used for quenching. The acetylene yield is stated to be 30 % by weight of the coal feed, whilst the specific energy consumption is reported to be less than 10 kWh kg^{-1} of acetylene. The acetylene concentration of the product gas reaches 15 %. This process was tried in a pilot plant at a coal throughput of 55 kg h^{-1}. Recent investigations on coal particle sizes between

5 and 100 μm show that a correlation exists between the residence time of coal particles in the hot part of the plasma reactor and the coal particle size. C_2H_2 yields, equal to 40% of the coal feed, can be obtained if this correlation is applied in reactor design (Bittner *et al.*, 1981).

In a joint project, Chemische Werke Hüls and Bergbau-Forschung are developing a process for the conversion of coal to C_2H_2 in a plasma reactor using a design based on laboratory data established by Bergbau-Forschung (Peuckert *et al.*, 1982).

6. Emissions from Gasification and Liquefaction Plants

The best method for estimating emissions from coal conversion plants is to evaluate design data for industrial plants. The Fluor Study (1981) gives emission data, in terms of kg per 1000 kg of coal input for gasification and liquefaction plants run on American coals, as: 0.66–3.09 SO_2; 0.51–2.34 NO_x; 0.02–0.20 CO; 606–1818 CO_2; 0.03–0.14 of particulate matter.

A corresponding study, investigating the conditions prevailing in the Federal Republic of Germany, was undertaken in 1982 (Gesamtverband, 1982). Some data are listed in Table 6.17. Subsequent discussion on plant design and coal feed with respect to emissions concluded that the most important parameters in this respect are coal characteristics, plant efficiency, type and degree of pollution control measures and statutory constraint (Jüntgen, 1981).

The investment cost for pollution control measures account for 5 to 16% of total capital investment. Reliable figures, however, can only be obtained when full design details of a plant for a given location are available.

TABLE 6.17. *Estimated gaseous emissions from projected gasification and liquefaction plants in the Federal Republic of Germany (Jüntgen, 1981)*

Process	Lurgi gasification	Texaco gasification	Direct liquefaction
product	SNG	Methanol	Gasoline
Form of emission ($kg\,TJ^{-1}$ produced)			
CO	85	13	3
CO_2	85,100	119,000	48,100
SO_2	80	107	51
H_2S	<1	<1	<1
NO_x	58–196	78–262	18–61
HCl	2	2	1
HF	<1	<1	<1
Particulates	6	9	3

7. References

ABUSLEME, J. A., BADILLA-OHLBAUM, R. and NEUBURG, H. J. (1983). Kinetic modelling of the catalytic liquefaction of sub-bituminous coals. *International Conference on Coal Science*, Pittsburg.

ANWER, J. and BÖGNER, F. (1976). Kohlevergasung im Fluidatbett unter Druck. *Brennstoff-Wärme-Kraft*, **28**, 57–60.

ARENDT, P. and VAN HEEK, K. H. (1981). Comparative investigations of coal pyrolysis under inert gas and H_2 at low and high heating rates and pressures up to 10^2 MPa. *Fuel*, **60**, 779–87.

ARNDT, E., FISCHER, R., FROHLICH, W., JÜNTGEN, H., TEGGERS, H. and WEISBRODT, I. (1979). Synthetisches Erdgas aus Kohle und Hochtemperatur-Warme. *Erdöl und Kohle, Erdgas, Petrochemie*, **32**, 17–23.

AVGUSHEVICH, I. V., LUKIN, A. M. and KASLINA, I. A. (1975). Molybdenum determination of the products of catalytic fuel conversion and in catalysts. *Khimiya tverdogo topliva*, **3**, 87.

BANICHEK, I. N. (1973). The Winkler process for production of low Btu gas from coal. *Clean Fuels from Coal*, Symposium (IGT), Chicago, Ill.

BITTNER, D., BAUMANN, H., PEUCKERT, C., KLEIN, J. and JÜNTGEN, H. (1981). Direktumwandlung von Kohle zu Acetylen. *Erdol und Kohle, Erdgas, Petrochemie*, **34**, 237–42.

BODLE, W. W. and VYAS, K. C. (1975). Clean fuels from coal-Introduction to modern processing. *Clean Fuels from Coal*, Symposium (IGT), Chicago, Ill.

BUNTHOFF, D., WANZL, W., VAN HEEK, K. H. and JÜNTGEN, H. (1982). Bildungskinetik leichter Aromaten bei der Hydropyrolyse von Steinkohlen. *Erdöl und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 167–9.

C&EN (1981a). International coal liquefaction plant scrubbed. *Chemical and Engineering News, 16 July*, 12.

C&EN (1981b). West Germany pursues active coal conversion development program. *Chemical and Engineering News, 30 November*, 34.

C&EN (1981c). China developing coal gasification industry. *Chemical and Engineering News, 19 October*, 35.

C&EN (1982a). Synfuels Corporation makes some final selections. *Chemical and Engineering News, 28 June*, 19.

C&EN (1982b). Shell halts gas-from-coal plans . . . *Chemical and Engineering News, 14 June*, 10.

C&EN (1982c). H-coal pilot plant test successful. *Chemical and Engineering News, 4 January*, 18.

C&EN (1982d). Ammonia plant based on Texaco coal process. *Chemical and Engineering News, 5 July*, 5.

CEP (1982). Joint use pact reached on prototype coal gas plant. *Chemical Engineering Progress, April*, 34.

CHADWICK, M. J. and LINDMAN, N. (1982). *Environmental Implications of Expanded Coal Utilization*. Pergamon, Oxford.

CONN, A. L. (1981). Conversion of coal or oil to gas. *Chemical Engineering Progress, May*, 11–20.

CONSIDINE, D. M. (1977). *Energy Technology Handbook*. McGraw-Hill, USA.

CORNILS, B., HIBBEL, J., RUPRECHT, P., DURRFELD, R. and LANGHOFF, J. (1981). RCH/RAG's version of Texaco coal gasification. *J. Hydrocarbon Processing, January*, 149–56.

CORNILS, B., KONKOL, K. I., RUPRECHT, P., LANGHOFF, J. and DURRFELD, R. (1982). Entwicklung und Einsatzmöglichkeiten der Texaco-Kohlevergasung in der technischen Version der Ruhrchemie AG Ruhrkohle AG. *Erdol. und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 109–10.

CYPRES, R. and FURFARI, S. (1981). Fixed-bed pyrolysis of coal under hydrogen pressure at low heating rates. *Fuel*, **60**, 768–78.

CYPRES, R., FURFARI, S. L., NEWMANN, C. and GHODSI, M. (1982). Hydropyrolysis, a third way for coal conversion. *Erdol und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 106–6.

DAVY POWERGAS (1973). *Winkler Generator Units*, Sales Brochure 21e/6/73.

DOLCH, P. (1936). *Wassergas, Chemie und Technik*. J. A. Barth Verlag.

DORSTEWITZ, U. and KAIMANN, W. (1981). Stand der drucklosen KNG-Kohle-vergasung, HdT Vortragsveröffentlichungen, *Heft 453*, 67–72.

ELGIN, D. C. (1976). Conventional gasification technologies, *ECE Symposium on Gasification and Liquefaction of Coal*, Dusseldorf.

EPPERLY, W. R., PLUMLEE, K. W. and WADE, D. T. (1981). EXXON Donor Solvent coal liquefaction process: Development Program Status V. *Eighth Energy Technology Conference and Exposition*, Washington, D.C.

EPPERLY, W. R. and TAUNTON, J. W. (1979). EXXON Donor Solvent coal liquefaction process development. *Coal Conversion Technology: Problems and Solutions*. American Chemical Society Symposium Series, **110**, 71–89.

FALBE, J. (1977). *Hrg Chemierohstoffe aus Kohle*. Georg Thieme Verlag, Stuttgart.

FEISTEL, P. P., VAN HEEK, K. H. and JÜNTGEN, H. (1978). *German Chem. Eng.* **5**, 294–8.

FLESCH, W. and VELLING, G. (1962). Die Vergasung Von Kohle im Winkler-Generator. *Erdöl und Kohle, Erdgas, Petrochemie*, **15** (9), 710–13.

FLUOR STUDY (1981). Quantifizierung der Umweltbeeinstrachtigung durch moderne Verfahren der Kohlevergasung und verflussigung. Studie ausfefuhrt von FLUOR GmbH, Dusseldorf.

FRANKE, F. H. (1978). Vergasung von braunkohle zu suynthese und reduktionsgas. *Vortragsveroffentlichungen, Haus der Technik, Heft*, **405**, 60–8.

FRANZEN, J. E. (1977). *Einige Uberlegungen zur Kohlevergasung*. Tech. Mitt, Krupp-Forsch. -Ber., 83–94.

FREDERSDORFF, G. G., VON, ELLIOTT, M. A. and LOWRY, H. H. (1963). *Chemistry of Coal Utilization*. Wiley, London.

GAGARIN, S. G., SMIRNOVA, T. S. and MARKINA, T. I. (1979). Kinetics of measuring residual volatiles yield during heat treatment of Irsha-Borodinski coal in vortex-chambers. *Khimiya tverdogo-topliva*, **1**, 87.

GESAMTVERBAND (1982). *Umweltvertraglichkeit von Anlagen zur Vergasung und Verflussigung von Steinkohle in der BRD*. Studie Gesamtverband Steinkohlenbergbauverein und Bergbau-Forschung GmbH.

GHASSEMI, M., CRAWFORD, K. and QUINLIVAN, S. (1978). *Environmental Assessment Data Base for High Btu Gasification Technology*, vol. II, Appendices A, B and C. US Environmental Protection Agency Report EPA-600/7-78-186b, Washington, D.C.

GHASSEMI, M., CRAWFORD, K. and QUINLIVAN, S. (1979). *Environmental Assessment Report; Lurgi Coal Gasification Systems for SNG*. US Environmental Protection Agency Report EPA-60037-79-120, Washington, D.C.

GORIN, E., LEBOVITZ, H. E., RICE, C. H. and SHRUCK, R. T. (1971). *Proceedings of 8th World Petroleum Congress: Preprints*, Session No. PD 10 (5), 44.

GRATKOWSKI, VON, H. W. (1958). Kohlvergasungsverfahren. *Ullmans Encyklopädie der Techn Chemie*, Urban and Schwarzberg, Munchen, Berlin. **10**, 376–458.

GRASER, U. and JANKOWSKI, A. (1982). Weiterverarbeitung von Klohleol zu marktgangigen Produkten. *Erdol und Kohle, Erdgas, Petrochemie*, **35**, 74–7.

GURRAN, G. P., SHRUCK, R. T. and GORIN, E. (1967). Mechanism of the hydrogen transfer process to coal and coal extract. *Industrial Engineering Chemistry Process Design and Development*, **6**, 166–73.

GOKE, E. (1979). Technische Entwicklung der Kohle-Flugstrom-Vergasung und ihre wirtschaftlichen Perspektiven. Vortrag: *United Nations Symposium on World Coal Prospects*, Katowice.

HANSEN, D. J. (1981). Synfuels progress likely to remain slow. *Chemical and Engineering News, 24 August*, 13–7.

HARKER, H. (1960). Catalysis by alkali-metal salts in carbon gasification reactions. *Proceedings of the 4th Conference on Carbon*. Pergamon, New York.

HEDDEN, K. (1962). Methanbildung aus Wasserstoff und Kohlenstoff bei hohen Temperaturen und Drucken. *Zeitschrift fur Elektrochemie, Berichte der Bundengesellschaft fur Physikalische Chemie*, **66**, 652–61.

HEDDEN, K. and MIENKINA, G. (1965). Zur Reaktionskinetik der Druckvergasung von Kohlenstoff mit H_2O/H_2-Gemischen. *Brennstoff-Chemie*, **46**, 366–71.

HEEK, VAN, K. H. (1978). *Stand der Kohlevergasung*. Vortragsverofflentlichungen Haus der Technik, *Heft 405*, 50–9.

HEEK, VAN, K. H. (1982). Druckpyrolyse von Steinkdohlen. VDI-Forschungsheft 612.

HEEK, VAN, K. H., JÜNTGEN, H. and PETERS, W. (1973). Fundamental studies on coal gasification in the utilization of thermal energy from nuclear high-temperature reactors. *Journal of the Institute of Fuel*, **249**, 249–58.

HEEK, VAN, K. H., JÜNTGEN, J. and PETERS, W. (1982). Wasserdampfvergasung von Kohle mit Hilfe von Prozesswarme aus Hochtemperatur-Kernreaktoren. *Atomenergie/Kerntechnik*, **40**, 225–45.

HILL, G. R., HARIRI, H., REED, R. J. and ANDERSON, L. L. (1966). Kinetics and mechanism of solution of high volatile coal. *Coal Science*. Advances in Chemistry Series, **55**, 427–47.

HILLEBRAND, W. A., LIPHARD, K. G., STROBEL, B., HODEK, W. and HOLLING, G. (1981). Hydrierte Kohleole als Einsatzprodukte fur des Steamcracken. *Erdol und Kohle, Erdgas, Petrochemie*, **34**, 443–6.

HILLER, H. (1975). Kohle-Druckvergasung. *Erdol und Kohle, Erdgas, Petrochemie*, **28**, 74–80.

HOFFMAN, E. J. (1978). *Coal Conversion*. Modern Printing Company, Laramie, Wy.

HOOGENDORN, J. C. (1976). Die neue Anlage Zur Erzeugung flüssiger Kohlenwasserstoffe aus Kohle in Sudafrika. *Gas Wärme Internat*. **25**, 283.

HOWARD, J. B. (1981). Fundamentals of coal pyrolysis and hydropyrolysis. *Chemistry of Coal Utilization* (ed. by M. A. Elliott). Wiley, New York.

HP (1982). Coal gasification unit successfully started up. *Hydrocarbon Processing, June*, 43.

HUHN, F., KLEIN, J. and JÜNTGEN, H. (1983). Investigations on the alkali-catalysed steam gasification of coal: kinetics and interactions of alkali-catalyst with carbon. *Fuel*, **62**, 234–8.

HONING, M. and DONATH, E. E. (1958). Kohle- und Ölhydrierung. *Ullmanns Encyklopädie der Technischen Chemie (3. Auflage)*. ds Verlag Urban und Schwarzenberg.

IVANOV, V. M., SMETANNIKOV, B. N. and PISAREV, YU. K. (1978). Thermal processing of coal liquefaction slurries. *Khimiya tverdogo topliva*, **4**, 68.

JACOBSSEN, T. and GALLEI, E. (1981). Hydrierung von Kohleol aus Saarkohle zu Motorbenzin. *Erdöl und Kohle, Erdgas, Petrochemie*, **34**, 447–50.

JÜNTGEN, H. (1977). Entwicklungstendenzen der Kohlevergasung. *Bergbau*, **28**, 347–55.

JÜNTGEN, H. (1979b). Verwendung von Kohlengas und Kohlenol. *Gluckauf*, **115**, 329–38.

JÜNTGEN, H. (1981). Umweltrelevanz der Kohlevergasung und der Kohleverflussigung. HdT Vortragsveroffentlichungen, *Heft 453*, 25–32.

JÜNTGEN, H. (1982a). Gasification and liquefaction of coal and data needed for plant design and operation. *8th International CODATA Conference*. Jachranka/Zegrzynek, Poland.

JÜNTGEN, H. (1982b). Basic reactions and technical performance of coal gasification and liquefaction. *International Symposium on Carbon: New Processing and New Applications*. Toyohashi, Japan.

JÜNTGEN, H. (1982c). The application of catalysts in coal gasification processes: incentives and perspectives. *Proceedings of International Symposium Fundamentals of Catalytic Coal and Carbon Gasification*. Amsterdam.

JÜNTGEN, H. and VAN HEEK, K. H. (1977). A technical scale gas generator for steam gasification of coal using nuclear heat. *Nuclear Technology*, **35**, 581–90.

JÜNTGEN, H. and VAN HEEK, K. H. (1979). An update of German non-isothermal coal pyrolysis work. *Fuel Processing Technology*, **2**, 261–93.

JÜNTGEN, H. and VAN HEEK, K. H. (1981). Kohlevergasung-Grundlagen und technische Anwendung. Verlag Karl Thiemig, Munchen.

JÜNTGEN, H. and KLEIN, J. (1976). Purification of wastewater from coking and coal gasification plants using activated carbon. *Energy Sources*, **2**, 311–30.

JÜNTGEN, H., KLEIN, J., KNOBLAUCH, K., SCHROTER, H. J. and SCHULTZE, J. (1980). Other uses of coal. *Chemistry of Coal Utilization* (ed. by H. H. Lowry).

JÜNTGEN, H., KLEIN, J., KNOBLAUCH, K., SCHROTER, H. J. and SCHULZE, J. (1981). Conversion of coal and gases produced from coal into fuels, chemicals and other products. *Chemistry of Coal Utilization* (ed. by M. A. Elliott). Wiley, New York.

JÜNTGEN, H. L., NITSCHKE, E. and WIEGAND, D. (1982). Emission control and its effect on investment costs for coal conversion plants. *International Symposium on Economic Aspects of Coal Pollution Abatement Technologies*. Petten, Netherlands.

KING, W. E. H. (1981). Coal gasification. *Fuel*, **60**, 803–8.

KOH, K. K. (1982). What's happening with coal gasification technology? *Hydrocarbon Processing, June*, 85–8.

KONKOL, W., RUPRECHT, P., CORNILS, B., DURRFELD, R. and LANGHOFF, J. (1982). Autothermal coal gasification. *Hydrocarbon Processing, March*, 97–102.

KRAAIJVELD, H. J. (1978). *Technische und wirtschaftliche Aspekte des Shell/Koppers-Kohlevergasungsverfahrens*. Vortragsveroffentlichungen, Haus der Technik, *Heft 405*, 74–7.

KREVELEN, VAN, D. W. (1961). *Coal—Typology, Chemistry, Physics and Constitution*.

KRICHKO, A. A., LEBEDEV, V. V. and FARBEROV, I. L. (1978). *Nonfuel Utilization of Coal*. Nedra, Moscow.

KROGER, C. (1939). Die Vergasung von Kohlenstuff durch Luft, Kohlendioxid und Wasserdampf und ihre Beeinflussung durch anorganische Katalysatoren. *Angewandte Chemie*, **52,** 129–30.

KRONIG, W. (1978). Chemische Rohstoffe durch direkte Kohlehydrierung. *Forschung Aktuell-Kohleverflussigung*. Umschau, Frankfurt am Main.

KUBIAK, H., SCHROTER, H. J., SULIMMA, A. and VAN HEEK, K. H. (1982). Applications of potassium carbonate catalysts in the coal gasification process using nuclear heat. *Proceedings of International Symposium on Fundamentals of Catalytic Coal and Carbon Gasification*, Amsterdam.

KUHN, A. K. (1981). Synfuels processing: The Great Plains Gasification Project, *Chemical Engineering Process, 2 April*, 64–8.

KUHN, L. and PLOGMANN, H. (1982). Reaction of catalyst with mineral matter during coal gasification. *Proceedings of International Symposium on Fundamentals of Catalytic Coal and Carbon Gasification*, Amsterdam.

LANGHOFF, J., DURRFELD, R. and WOLOWSKI, E. (1981). *Aktivitaten der Ruhrkohle AG auf dem Gebiet der Hydrierung und Vergasung von Kohle*. HdT Vortragsveroffentlichungen, *Heft 453*, 9–17.

LINKE, A. and VOGT, E. V. (1979). *Die Druckvergasung von Kohle im Flugstrom nach Shell/Koppers*. Vortrag: Achema, Frankfurt.

LIPHARD, K. G., GALE, M., HANDRICK, K., STROBEL, B., RIEPE, W. and KOLLING, G. (1981). Chromatographie von Kohleol. *Erdöl und Kohle, Erdgas, Petrochemie*, **34,** 488–91.

MCKEE, D. W. (1983). Mechanisms of the alkali metal catalysed gasification of carbon. *Fuel*, **62,** 170–5.

MCKEE, D. W. and CHATTERJI, D. (1978). The catalysed reaction of graphite with water vapour. *Carbon*, **16,** 53–7.

MCKEE, D. W., SPIRO, C. L., KOSKY, P. G. and LAMBY, E. J. (1983). Catalysis of coal char gasification by alkali metal salts. *Fuel*, **62,** 217–20.

MIMS, C. A. and PABST, J. K. (1983). The role of surface salt complexes in alkali catalysed carbon gasification. *Fuel*, **62,** 176–9.

NAHAS, M. C. (1983). The EXXON catalytic coal gasification process: fundamentals to flowsheets. *Fuel*, **62,** 239–41.

OTTO, K., BARTOSIEWICZ, L. and SHELEV, M. (1979). Catalytic steam gasification of graphite: effects of calcium, strontium and barium with and without sulphur. *Carbon*, **17,** 351–7.

OWEN, J. (1982). Coal derived liquids as a source of chemical feedstock. Lecture to "Answer" Conference, Society of Chemistry and Industry, Brussels.

PASSLER, P. (1982). Acetylene: a basic chemical with a future? Lecture to "Answer" Conference, Society of Chemistry and Industry, Brussels.

PETERS, W. (1976). Grundlagen der Kohlevergasung. General Report. Symposium uber Kohlevergasung und verflussigung der UN-Economic Commission for Europe, Dusseldorf.

PETERS, W. and SCHILLING, H. D. (1977). Perspektiven der Kohlevergasung under der Kohlevergasung aus der Sicht der Steinkohle. *Erdöl und Kohle*, **30,** 552–7.

PEUCKERT, C., MULLER, R., WILMING, F. and BITTNER, D. (1982). Neuere Erkenntnisse auf dem Gebiet der Plasmapyrolyse von Steinkohle. *Erdöl und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 158–9.

PFEIFFER, R. and PASCHEN, P. (1981). Das Humboldt-Kohlevergasungsverfahren. HdT Vortragsveroffentlichungen, *Heft 453*, 43–6.

PISAREV, YU. K., KRICHKO, A. A. and IVANOV, V. M. (1975). The energy-technological process of hydrogenation slurry utilization in cyclone furnaces. *Trudy Tsentralnogo Kotloturbinnogo Instituta*, Polzunova.

ROBSON, B. (1977). A review of gasification of power generation. *Energy Research*, **1,** 157.

RÖBKE, G. (1978). *Weiterentwicklung des Lurgi-Druckvergasungsverfahrens*. Vortragsveroffentlichungen, Haus der Technik, *Heft 405*, 85–90.

ROSSBACH, M., KUFFNER, P., FUSSMAN, G., GRAMS, W. and PETER, K. (1978). *Weiterentwicklung des Lurgi-Druckvergasungsverfahrens*. Vortragsveroffentlichungen, Haus der Technik, *Heft 405*, 78–84.

SCHAFER, W., HEINRICH, G., LOHMANN, G. and PEYRER, H. P. (1982). Ruhr 100—Neue Ergebnisse von der Weiterentwicklung der Lurgi Druckvergasung. *Erdöl und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 114–5.

SCHILLING, H. D., BONN, B. and KRAUSS, U. (1979). *Kohlevergasung*. Band 4, Rohstoffindustries International. Verlag Gluckauf GmbH, Essen.

SCHINDLER, H. (1982). Coal liquefaction. *Hydrocarbon Processing, June*, 88.

SCHLINGER, W. G., KOLAIAN, J. H., CORNILS, B. and LANGHOFF, J. (1982). Commercialization status of the Texaco Coal Gasification Process. Paper No. 2588, *Coal Gasification Conference, Europe 82, Amsterdam.*

SCHMIDT, J. (1966). *Technologie der Gaserzeugung.* Band II, Vergasung: VEB Deutscher Verlag fur Grundstoffindustrie, Leipzig.

SCHULZE, J. (1974). Qualitative und quantitative Substitutionschancen fur die Kohlechemie. *Chem-Ing-Techn.* **46**, 976-80.

SCHULZE, J. (1978). *Die Erzeugung von Chemierohstoffen aus Kohle.* Vortragsveroffentlichungen, haus der Technik, *Heft 405*, 16-29.

SCHULZE, J. and SCHMIDT, B. (1981). Kohlebedarf bei der Substitution der Petrochemie durch Kohlechemie. *Chemistry and Industry*, **23**, 623-6, 686-9 and 791-3.

SEIPENBUSCH, J. and RUPRECHT, P. (1978). *Das RAG/RCH-Projekt Kohlestaub-Druckvergasung*, Vortragsveroffentlichungen, Haus der Technik, *Heft 45*, 69-73.

SHIRES, M. J. (1981). Fuel gas from coal. *Fuel*, **60**, 809-16.

SPEICH, P. (1981). Lignite processes planned in Germany. *Hydrocarbon Processing, July*, 171-6.

SPIRO, C. L., MCKEE, D. W., KOSKY, P. G. and LAMBY, E. J. (1983). Catalytic CO_2 gasification of graphite versus coal char. *Fuel*, **62**, 180-4.

STAEGE, H. (1976). Die Kohlevergasung nach Koppers-Totzek. *Erdöl-och Erdgas-Zeitschrift*, **92**, 381-7.

STERBENZ, PL. (1980). Coal conversion today. *Hydrocarbon Processing, July*, 68-72.

STROBEL, B., FRIEDRICH, F., KOLLING, G., LIPHARD, K. G. and ROMEY, I. (1981). Kohlehydrierung bei der Bergbau-Forschung. *Erdol und Kohle, Erdgas, Petrochemie*, **34**, 391-6.

STRUERMER, D. H., NG, D. J. and MORRIS, C. J. (1982). Organic contaminants in ground water near an underground coal gasification site in north-eastern Wyoming. *Environmental Science and Technology*, **16**, 582-7.

SULIMMA, A., VAN HEEK, K. H. and JÜNTGEN, H. (1981). Einfluss und Verbleib der Alkali-Katalysatoren bei der Kohlevergasung. *Proceedings of the International Conference on Coal Science, Dusseldorf.* Verlag Gluckanf GmbH, Essen.

TAUNTON, J. W., TRACHTE, K. L. and WILLIAMS, R. D. (1981). Coal feed flexibility in the EXXON Donor solvent coal liquefaction process. *Fuel*, **61**, 788-94.

TAYLOR, H. S. and NEVILLE, H. A. (1921). Catalysis in the interaction of carbon with steam and with carbon dioxide. *Journal of the American Chemical Society*, **43**, 2055-71.

TEGGERS, H., LENZ, U. and SCHRODER, L. (1981). Aktivitäten der Rheinischen Braunkohlenwerke AG auf dem Gebiet der Vergasung und Verflussigung von Braunkohle. HdT Vortragsveroffentlichungen. *Heft 453*, 33-43.

TESSIE DU MOTAY, C. M. and MARECHAL, C. R. (1867). British Patent No. 2548.

THEIS, K. A. and NITSCHKE, E. (1982). Make syngas from lignite. *Hydrocarbon Processing, September*, 233-7.

THURNAU, R. C. and BATES, E. R. (1980). EPA research: *in situ* gasification results to date. *Sixth Underground Coal Conversion Symposium.* Afton, Oklahoma.

TORSTER, K., MARGOLIS, R. L. and KUHN, A. K. (1982). Implementing America's first commercial synfuels project—The Great Plains Coal Gasification Project. *Chemistry, Economy and Engineering Review, March*, 7-14.

VERAA, M. J. and BELL, A. T. (1978). Effects of alkali metal catalysts on gasification of coal char. *Fuel*, **57**, 194-200.

VGB Konferenz (1979). Kohlevergasung in der Energietechnik, VGB-TB 201, Essen.

VOGT, E. V. and ECKSTEIN, G. (1982). Zur Entwicklung des Shell-Kohlevergasungs-Prozesses (SCGP). *Erdol und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 111-3.

VOLKEL, H. K., ECKSTEIN, G., LINKE, A. and VOGT, E. V. (1979). Operation and economics of the Shell-Koppers coal gasification process. *14th World Gas Conference of the International Gas Union*, Toronto.

WADDEN, R. A. (1976). Coal hydrogenation and environmental health. *Environmental Health Perspectives*, **14**, 201-6.

WANG, F. T., MEAD, S. W. and STRUERMER, D. H. (1982). *Water quality monitoring at the Hoe Creek test site: Review and preliminary conclusions.* UCRL-87650, Lawrence Livermore Laboratory, University of California.

WANZL, W., VAN HEEK, K. H. and JÜNTGEN, H. (1982). Results of the IEA-Pyrolysis Project at

Bergbau-Forschung. *Application of Pyrolysis Technology to Utilization of Eastern Canadian Coals*. Fredericton, Canada.

WENDER, J., HEREDY, L. A., NEUWORTH, M. B. and DRYDEN, I. G. C. (1981). Chemical reactions and the constitution of coal. *Chemistry of Coal Utilization* (ed. by M. A. Elliott). Wiley, New York.

WHITEHURST, D. D., MITCHELL, T. O. and FARCASIU, M. (1980). *Coal Liquefaction—The Chemistry and Technology of Thermal Processes*. Academic Press, New York.

WIGMANS, T., ELFRING, M. L., HOOGLAND, A. and MOULIJN, J. A. (1981). Reaction mechanism of catalytic carbon gasification: differences and similarities between nickel, potassium and vanadium catalysis. *Proceedings of the International Conference on Coal Science, Dusseldorf*. Verlag Gluckauf GmbH, Essen.

WISER, W. H. (1968). A kinetic comparison of coal pyrolysis and coals dissolution. *Fuels*, **47**, 475–86.

WOLOWSKI, E. (1982). *Betriebsergebnisse aus der Kohleolänlage Bottrop*. Vortrag DGMK-Bezirksgruppe-Ruhr, Bottrop.

WOLOWSKI, E. and HOSANG, H. (1982). Erste Betriebserfahrunger mit der Kohleol-Anlage Bottrop. *Erdol und Kohle, Erdgas, Petrochemie*, Compendium 82/83, 93–4.

WÜRFEL, H. (1981). Aktivitaten der Saarbergwerke AG auf dem Gebiet der Hydrierung und Vergasung. HdT Vortragsveroffentlichungen. *Heft 453*, 17–20.

YOON, H., WEI, J. and DENN, M. M. (1978). A model for moving bed coal gasification reactors. *AIChE Journal*, **25**, 885.

ZVEREV, D. P., KRICHKO, A. A., SMIRNOVA, T. S. and MARKINA, T. I. (1981). Synthetic solid fuel production from Kansk-Akchinsk coal. *Khimiya tverdogo topliva*, **7**, 39.

7

Application of the Exposure Commitment Model to Coal Utilization Emission Problems

1. The Exposure Commitment Concept 156
   1.1. Concepts and Definitions 157
   1.2. Evaluation of the Applicability of the Methods 158
2. Application of the Exposure Commitment Model 160
   2.1. Health Effects of Electric Power Generation 160
   2.2. Collective Dose Commitments Due to Radioactive Effluents from Power Stations 162
      2.2.1. Introduction 162
      2.2.2. Effluents and collective dose commitments for model plants 162
      2.2.3. Risk comparisons from radiation exposure from the use of coal and nuclear energy 163
   2.3. Comparative Risks Associated with Total Fuel Cycles 166
3. Comparative Assessments and the Exposure Commitment Method 168
4. References 169

1. The Exposure Commitment Concept

In order to assess the total effects of releases of radioactive material into the environment by such practices as detonating atomic devices, the dose commitment concept was developed by Lindell (1960) and the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Information on the sources and effects of ionizing radiation are published from time to time (UNSCEAR, 1977). It was subsequently suggested by Butler *et al.* (1972) and Lindell (1978) that the concept could be applied to non-radioactive environmental pollutants. The concept has been developed into a method, now called the exposure commitment method (Barry, 1979; O'Brien, 1979; Bennett, 1981; Buchanan and Bennett, 1980), to give a time-independent approach to environmental pollutant assessment. A brief outline of the concepts and definitions of the method are given here, followed by a discussion of its applicability to coal combustion and conversion activities.

1.1. Concepts and Definitions

Two recent publications on the exposure commitment method (Bennett, 1981; Buchanan and Bennett, 1980) emphasize that exposure commitment is a combined measure of the concentration and duration of a pollutant's presence in an environmental medium. In the entire period of a particular exposure:

$$E = \int_0^\infty C(t)\,\mathrm{d}t$$

where $C(t)$ is the concentration at any particular time t. If the probability of occurrence of a response rather than the severity, increases with exposure (as is postulated with radionuclide exposure–response relationships) then the curve relating concentration and time (CT) is one of equal probability effects. This is referred to as a stochastic effect. If a "threshold" is postulated, but stochastic effects are exhibited once this is exceeded, the concentration × time curve will indicate threshold exposure. For either, the exposure commitment is time-independent: various combinations of concentration and time periods can give the same exposure commitment and equal probability of effect or transcendence of the threshold.

Exposure commitment may be applied for the site of impact but it can also be used as a measure of the exposure in the preceding medium. Thus intake may be related to the external conditions and also transfer from one environmental compartment to another quantitatively described.

If linearity is assumed for the transfer process, transfers are additive and intercompartmental movement (from compartment i to compartment j) of the pollutant may be described by its flux:

$$F_{ij}(t) = K_{ij}Q_i(t)$$

where $Q_i(t)$ is the amount of pollutant in the donor compartment and K_{ij} the proportionality constant (the transfer rate constant), depending upon the ratio of concentrations $C_i(t)$ and $C_j(t)$. Then:

$$F_{ij}(t) = K_{ij}M_i\frac{Q_i(t)}{M_i} = K_{ij}M_iC_i(t)$$

where M_i is the mass or volume of compartment i.

The intake to a compartment is the integral of the flux (the product of the average amount passing per unit time and the time period). Hence the total intake can be expressed as:

$$\begin{aligned}\textstyle\sum_i I_{ij} &= \textstyle\sum_i \int_0^\infty F_{ij}(t)\,\mathrm{d}t\\ &= \textstyle\sum_i K_{ij}M_i \int_0^\infty C_i(t)\,\mathrm{d}t\\ &= \textstyle\sum_i K_{ij}M_iE_i\end{aligned}$$

The total intake is directly proportional to the exposure commitment in the donor compartment and is thus a useful measure of environmental transfer. As intake commitment and exposure commitment are integral quantities, time integrals of flux in concentration remove time-dependence and give an expression of the overall transfer results.

Various integral quantities can be related by means of a transfer factor (P_{ij}). For example:

$$P_{ij} = \frac{E_i}{E_j} = \frac{\int_0^\infty C_i(t)\,dt}{\int_0^\infty C_i(t)\,dt}$$

For continuing releases steady-state levels in compartments are commonly attained. With concentrations that remain fairly constant, transfer factors may be evaluated by taking the ratios of equilibrium concentrations on fluxes.

1.2. Evaluation of the Applicability of the Methods

In terms of measuring or assessing likely effects, the exposure commitment method must be applied with caution to coal combustion and conversion impacts. Various combinations of concentration and time period will give an equal exposure commitment, but these may not be associated with equal occurrences of harmful effects. This will be particularly true if threshold levels apply. There is a considerable body of opinion that this is so for most emissions from coal combustion and conversion (Ulmer, 1977) and that no linear dose–response relationship exists. Repair mechanisms following exposure are cited as an added reason for rejecting the "no zero-effect dose" hypothesis.

However, others have argued in favour of the linear no-threshold model (LNTM), as a working hypothesis, on the basis of synergistic effects, the carcinogenic effect of some products of coal combustion and conversion, and the wide confidence limits that need to be set on data (Butler, 1978; Intl. Symp., 1978; Wilson, 1978). Some have concluded that for chemical pollutants of air it would be prudent to assume a linear no-threshold model.

As explained, this assumption permits the application of the exposure commitment method with relative ease. It is also worth noting that the LNTM has another important consequence for pollution control practices. If the quantity of a pollutant to which a population is exposed is constant, the total number of health effects produced is constant (with an uncertainty of the square root of the number of effects). Thus no benefit results from measures that expose larger numbers of people to lower concentrations as results from using higher stacks for air effluents and discharging liquid effluents into larger volumes of water. The only control measures that succeed are those that prevent pollutants from coming in contact with receptors such as retention of the pollutant at the source or, for polluted air, discharging it at the seaboard where it will be carried out to the ocean. But even this may have its limitations.

Another biological assumption basic to the exposure commitment method, already referred to, is that there is no repair of the effects, however small. This means that the effects per unit exposure will be the same, whether this is over a short or a prolonged period.

Following on these two assumptions it is possible to conclude that all exposure commitments to a population, however large, over all time, however long, contribute to the total impact. Thus for every action that contaminates the environment the total CT for the receptor must be assessed together with the total of the resulting effects or harm, both short-term and long-term. The various types of situations to be considered have been classified as follows (Butler, 1978):

Release	*Contaminant*
A. Single	1. Short-lived
	2. Long-lived
B. Continued	1. Short-lived
	2. Long-lived

For case A.1 the annual exposure commitment will encompass the total and will be adequate to assess the total detriment. For the other three cases the estimates must extend beyond 1 year. The assessment of effects beyond 1 year may be necessary because:

(1) the amount of the practice may increase in the future;
(2) the pollutant may persist in the environment long after its release;
(3) the pollutant may be produced in the environment long after the release of its parent substance, e.g. methyl mercury;
(4) the pollutant may remain in the body of the receptor long after its intake;
(5) the biological effects may be expressed long after the initial intake.

The information required for calculating exposure commitment usually consists of data obtained by survey, varying in time and space, on sources, pollutants released, receptors and information from research on the organs and processes of the receptor affected by the pollutant. However, it requires a more limited data base than time-dependent assessments. Exposure commitment, multiplied by the appropriate risk estimate, allows an estimate of the harm commitment to be obtained.

The harm commitment method is most useful for assessment and planning purposes when each estimate is normalized to the amount of practice that generates the exposure commitment. When given in this way, the commitments from alternative practices can be compared and the consequences of future expansion can be calculated.

2. Application of the Exposure Commitment Model

Non-stochastic effects that resulted from the burning of fossil fuels were the increased deaths that arose promptly from the heavy fogs that occurred in London, Oslo and the Meuse valley (Hamilton, 1980). These were probably due to the combined action of soot with oxides of sulphur and nitrogen.

The stochastic effects of most concern in radiation protection are increased malignancies and mutations. The pollutants released to the atmosphere by combustion of fossil fuels, that may affect human health, are: oxides of carbon, nitrogen and sulphur; heavy metals; polynuclear aromatic hydrocarbons such as benzo(α)pyrene; radionuclides. Some of these are probably carcinogenic or mutagenic. Another kind of stochastic effect, probably caused by oxides of sulphur (sulphate) combined with soot and aggravated by cigarette smoking, is the development of chronic bronchitis after long exposure (Nishiwaki *et al.*, 1971).

2.1. Health Effects of Electric Power Generation

Effluents from a coal-fired power plant generating 1000 MW(e) yr^{-1} (8.76×10^9 kWh yr^{-1}) are given in Table 7.1. They are calculated from data given by Wilson and Jones (1974) and assume the power plant burns 3×10^6 t coal with a 2 % sulphur content, an energy content of 2.74×10^7 kJ t^{-1} and the thermal efficiency of the power plant is 38 %; a fly ash removal efficiency of 99 % and no flue gas desulphurization is assumed. Such a plant would have a capacity greater than 1000 MW(e), (a power plant with a generating capacity of 1000 MW(e), operating at 0.75 capacity would produce 6.75×10^9 kWh yr^{-1}).

TABLE 7.1. *Annual effluent production (tonnes) from a 1000 MW(e) coal-fired power plant (UNEP, 1981)*

Airborne effluents:	
Particulates	3×10^3
Sulphur dioxides	11×10^4
Nitrogen oxides	2.7×10^4
Carbon monoxide	2×10^3
Hydrocarbons	400
Liquid effluents:	
Organic material	66.2
Sulphuric acid	82.5
Chloride	26.3
Phosphate	41.7
Boron	331
Suspended solids	497
Solid wastes:	
Bottom ash + recovered fly ash	3.6×10^5

Emissions from power plants in the Federal Republic of Germany, burning bituminous coal and conforming to the prevailing emission standards, show similar stack releases except that, due to some removal of sulphur and particulates in the flue gas, SO_2 and particulate values are considerably lower. Brewer (1976) estimates trace metal and radioactivity emissions from such a coal-fired power station as 0.67 t and about 1 GBq respectively. Table 7.2 gives comparative figures for different forms of fuel for airborne effluents. Emissions such as these contribute to levels of air pollution in the USA shown in Table 7.3.

The examples of power generation by burning coal, and the emissions that result, represent a long-term continuous process of type B (Butler, 1978). The pollutants emitted, and their effects, are both short- and long-lived. A number of epidemiological studies have shown that an increase in airborne sulphates is associated with an increase in death rate; the results of several such studies are summarized by Hamilton (1980). There the risk estimate given is 33 excess deaths per year in a population of a million persons exposed to an increase in airborne sulphates of 1 $\mu g\ m^{-3}$.

To estimate the exposure commitment and harm commitment from these data it must be assumed that the emissions are constant and impinge on some population distributed around the power plant. For a population of 3 million uniformly distributed within a radius of 50 miles, Hamilton (1980) assumed an SO_2 emission rate of 0.12 lb (54 g) of SO_2 per million Btu in the fuel ($< 1\%$ sulphur coal with 90% removal of sulphur in flue gas). The calculated deaths per year from the generation of 1000 MW(e) (plant operating at 65% capacity) were: 0–3, mean = 0.6 in a 50-mile radius, and 0.30, mean = 6 for the whole USA.

TABLE 7.2. *Airborne effluents (tonnes) from 1000 MW(e) yr^{-1} power plants (UNEP, 1981)*

Fuel	Particulates	CO	NO_x	SO_2	Hydrocarbons
Coal	3×10^3	2×10^3	2.7×10^4	11×10^4	400
Oil	1.2×10^3	0.7×10^3	2.5×10^4	3.7×10^4	470
Natural gas	0.5×10^3	negligible	2×10^4	20.4	34

TABLE 7.3. *Concentrations of pollutants in urban and rural air ($\mu g\ m^{-3}$) (Natusch, 1978)*

	Urban air		Rural air
Substance	Range	Average	Range
SO_2	20–1200	70	0.1–5
NO_x	50–400	100	2–6
Particulates	$(60\text{–}220) \times 10^3$	140×10^3	$(5\text{–}60) \times 10^3$

2.2. *Collective Dose Commitments Due to Radioactive Effluents from Power Stations*

2.2.1. *Introduction*

UNSCEAR (1977) have made a comparative radiation exposure assessment for coal and nuclear energy-based electricity generation. Revised calculations have recently been made (Halbritter *et al.*, 1981) for siting and technical conditions in the Federal Republic of Germany (FRG). It is thus possible to use these calculations to compare a model coal-fired power plant and a model nuclear power plus full reprocessing and waste treatment plant. Comparisons of radiation exposure from the total nuclear fuel cycle and coal-fired power plants must include exposure due to accidents from nuclear sources, and exposure values for these are taken from a study undertaken in the FRG (Ministry for Research and Technology, 1979).

2.2.2. *Effluents and collective dose commitments for model plants*

Calculations concerning radioactive effluents are standardized on a GW(e)yr basis for a bituminous coal-fired power plant and a nuclear fuel power plant together with all the relevant facilities. For the coal-fired power plant assumptions similar to those of McBride *et al.* (1977) were made and these lead to emission rates that agree well with those of UNSCEAR (1977). Measurements of fly ash activity (Chatterjee *et al.*, 1980; Kolb, 1979) are also of the same order of magnitude except that Pb-210 and Po-210 are highly enriched in fly ash where there is liquid ash removal, and such a condition was assumed for the model plant. Table 7.4 gives effluent values. Emission data for a typical PWR were based on actual emissions of three plants in the FRG since 1972 (Table 7.5). For the fuel reprocessing and waste treatment plant a global estimation on the basis of release factors was made and is in good agreement with other estimates (USDOE, 1979). Effluent values are given in Tables 7.5 and 7.6.

To make a risk specific comparison collective dose commitments must be calculated. To allow comparison, values from UNSCEAR (1977) were converted to dose equivalents (ICRP, 1976). Effective dose commitments were obtained to take account of specific conditions in the FRG (siting and technical features; population density, 250 km^{-2}) using dose conversion factors (ICRP, 1979). Generally the same model assumptions made by UNSCEAR (1977) were used, although for the coal-fired power plant some additional exposure pathways were taken into account.

It will be seen (Table 7.4) that the new estimates for coal-fired power plants yield about 4 man-Sv per GW(e)yr as compared to about 0.6 man-Sv per GW(e)yr. The higher exposure is due mainly to conservative assumptions in relation to the distribution of radionuclides in the soil that affect the ingestion

TABLE 7.4. *Radionuclide emissions and collective dose equivalent commitment from coal-fired power plants*

	Emissions (GBq per GW(e)yr)		Collective dose equivalent commitment (man-Sv per GW(e)yr)			
			UNSCEAR (1977)		Halbritter *et al.* (1981)	
Nuclide	UNSCEAR (1977)	Halbritter *et al.* (1981)	Ingestion	Inhalation	Ingestion	Inhalation
U-238	1.85	0.48	0.1		0.001	0.085
Th-234		0.27			–	–
U-234		0.48			0.002	0.17
Th-230		0.27			0.023	0.137
Ra-226	0.37	0.35	0.014		0.083	0.005
Pb-210	3.7	2.18	0.4		1.54	0.078
Po-210		5.55		no values given	0.91	0.098
Th-232	0.37	0.18	0.002		0.032	0.315
Ra-228	0.37	0.23	0.03		0.055	–
Th-228	0.37	0.18			0.005	0.105
Ra-224		0.23			0.003	0.003
K-40	5.55	0.78			–	–
Total			0.546		2.65	0.996
					3.73[a]	

[a] Includes 0.08 γ soil radiation.

pathway. It is now estimated that the ingestion pathway results in the highest individual doses, whereas the inhalation pathway makes the dominant contribution to collective dose. Calculations of the radiological impact of atmospheric discharges from a hypothetical 2000 MW(e) power station in Great Britain give a dose of 10 man-Sv per GW(e)yr (Camplin, 1980), a value equal to exposure to the nuclear fuel cycle of reactors other than PWR. In fact, individual doses emanating from coal-fired power stations are likely to be considerably lower than this.

For the nuclear fuel cycle, modifications to suit conditions in the FRG were made to the basic assumptions (UNSCEAR, 1977). Data from Kelly (1975) concerning C-14 were used, resulting in lower exposure values (Tables 7.5 and 7.6). The lower noble gas and tritium emissions expected from the PWR model power plant all contribute to lower exposure values than those given by UNSCEAR (1977).

2.2.3. Risk comparisons from radiation exposure from the use of coal and nuclear energy

Table 7.7 gives estimates of all exposures from different sectors of the nuclear fuel cycle. The 4 man-Sv per GW(e)yr occupational exposure is valid

TABLE 7.5. *Collective dose equivalent commitment due to radioactive emissions from nuclear power plants*

				Collective dose equivalent commitment (man-Sv per GW(e)yr)		
		Radioactive emissions (GBq per GW(e)yr)		UNSCEAR (1977)	Halbritter *et al.* (1981)	
Nuclide	Exposure pathway	UNSCEAR (1977)	Halbritter *et al.* (1981)	Gonads	Gonads	Effective dose
Kr, Xe, Ar	atmosphere (local and regional)	3.7×10^7	3.5×10^5	2	0.09	0.09
H-3		6.7×10^5	1.4×10^3	0.04	0.0007	0.0007
C-14		2.2×10^2	2.2×10^2	0.006	0.015	0.015
Cs, Co, Sr, Ru		2.6×10^0	2.6×10^0	0.033	0.03	0.031
H-3	water (local and regional)	1.5×10^5	1.5×10^4	0.3	0.028	0.028
Cs, Co, Mn, I		2.2×10^2	8.1×10^1	0.06	0.032	0.033
H-3	atmosphere and water (global)	5.9×10^5	1.9×10^4	0.5	0.015	0.015
C-14		2.2×10^2	2.2×10^2	2.7	1.35	1.35
Total				5.64	1.56	1.56

TABLE 7.6. *Collective dose equivalent commitment due to radioactive emissions from reprocessing plants*

				Collective dose equivalent commitment		
		Radioactive emissions (GBq per GW(e)yr)		UNSCEAR (1977)	Halbritter *et al.* (1981)	
Nuclide	Exposure pathway	UNSCEAR (1977)	Halbritter *et al.* (1981)	Gonads	Gonads	Effective dose
Kr-85	atmosphere (local and regional)	13.875×10^6	3.7×10^5	0.007	0.0005	0.0005
H-3		37×10^3	3.7×10^4	0.002	0.005	0.005
C-14		5.18×10^2	3.7×10^2	0.01	0.018	0.18
Cs, Ru, Sr		3.7×10^0	1.48×10^1	0.002	0.56	0.57
H-3	water (local and regional)	2.22×10^5	3.7×10^5	0.4	0.76	0.67
Cs, Ru, Sr, I		3.7×10^3	3.7×10^0	0.4	0.017	0.016
H-3	atmosphere and water (global)	2.59×10^5	4.07×10^5	0.22	0.34	0.34
Kr-85		13.875×10^6	3.7×10^5	0.9	0.025	0.025
C-14		5.13×10^2	3.7×10^2	6.3	2.25	2.25
Total				8.24	3.89	3.90

only for the PWR; other reactor types would yield 10 man-Sv per GW(e)yr, similar to the value given by UNSCEAR (1977). The radiation exposures made use of UNSCEAR (1977) data for mining, milling, fuel fabrication and transportation and extrapolated experience at Karlsruhe for fuel reprocessing. The new estimates show that the radiation exposure due to the emissions from coal-fired units is comparable to the non-occupational radiation exposure from all relevant stages of the nuclear fuel cycle at normal operation. Taking occupational and non-occupational exposures for normal operation, the contribution from the nuclear fuel cycle is about four times that of energy conversion by coal.

Potential accidents are a major nuclear energy risk. The nature of such accidents means that possible exposure values could be so different in relation to impact areas that both stochastic and non-stochastic health impacts must be considered, which is only possible using the detriment level approach. The FRG risk study for nuclear power plants (Ministry for Research and Technology, 1979) gives stochastic risk (which is proportional to the expected dose commitment value) and is about 30 man-Sv per GW(e)yr (Table 7.7). Using dose-effect relationships for stochastic effects (ICRP, 1976) and estimates of non-stochastic accidental risk from the Ministry of Research and Technology (1979) an estimate of the total health impact from radiation exposure from the two fuels can be made. This indicates that the risk from radiation caused by coal combustion is about an order of magnitude smaller than the total radiation exposure risk of nuclear power. There are also,

TABLE 7.7. *Collective dose equivalent commitments and number of fatalities (somatic) for the nuclear fuel cycle and for coal-fired power plants*

	Collective dose equivalent commitment (man-Sv per GW(e)yr)			
	UNSCEAR (1977)	Halbritter *et al.* (1981)		Number of fatalities (Somatic) $(\mathrm{GW(e)yr})^{-1}$
Exposure	Gonads	Gonads	Effective dose	
Nuclear power plant:				
Public	6	1.6[a]	1.6[a]	0.02
Occupational	10	4[a]	4	0.05
Reprocessing:				
Public	8	4	4	0.05
Occupational	12	1	1	0.01
Mining, milling, fuel fabrication and transportation:				
Public	0.2	0.2	5	0.06
Occupational	2	2	2	0.02
Total for nuclear fuel cycle (normal operation)	38.2	12.8	17.6	0.22
Nuclear accidents:				
Non-stochastic effects				4×10^{-5}
Stochastic effects			30	0.4
Coal-fired power plants	0.6	–	3.7	0.05

[a] For PWR plants.

however, non-radiation risks associated with the use of the two fuels. These are dealt with briefly in the following sections.

2.3. *Comparative Risks Associated with Total Fuel Cycles*

The effects of electric power generation that have been considered are only part of the whole health impact of coal-fired electric power plants. It is necessary also to take into consideration the health effects of the complete operation with coal mining, processing, transporting and burning. These have been summarized for the USA by Hamilton (1984), as shown in Table 7.8, where they were compared with the same harm commitments from nuclear power generation (Hamilton, 1984). Tables 7.9 and 7.10 make a similar comparison for the USSR.

In making decisions about the control of a practice the scientific administrator will gain perspective by making other kinds of comparison between sources of power and causes of death. The amounts of electricity generated

TABLE 7.8. *Health impacts in the USA from coal and nuclear fuel cycles as deaths or cases per GW(e) yr (Hamilton, 1984)*

	Coal effects		Nuclear effects	
Exposure	Deaths	Disease/ injury	Deaths	Disease/ injury
Extraction				
Occupational:				
Accidents – surface	0.15–0.17	16	0.11	4.0
– underground	1.2–1.5	170	0.36	13.8
Disease	0.61–1.46	22	0.15	0.03
	(underground)		(radiation induced)	
Public:				
Disease	–	–	0.05	0.01
Processing				
Occupational:				
Accidents	0.034–0.116	3–5	0.006	1.6
Disease	–	–	0.05	0.05
Public:				
Disease	–	–	0.013	0.003
Transport				
Occupational:				
Rail	0.005–0.022	1.1	–	–
Truck	0.6–2.4	20.0	–	–
Barge	0.25–1.0	1.6	–	–
Pipeline	0.16	0.26	–	–
Routine operation	–	–	4.5×10^{-4}	5.5×10^{-4}
Accident	–	–	0.001	$< 10^{-4}$
			(radiological)	
			0.01	0.1
			(non-radiological)	
Public:				
Rail	0.37–0.54	0.87	–	–
Truck	0.6–2.4	20.0	–	–
Barge	0.07–0.28	0.15	–	–
Pipeline	0	0	–	–
Routine operation	–	–	4.5×10^{-4}	4.6×10^{-4}
Generation				
Occupational:				
Accidents	0.09–0.2	5	0.013	1.31
Rad-induced cancer	–	–	0.13	0.16
Public:				
Air pollution	15 (0–77)	77 (0–385)	–	–
Rad-induced cancer	–	–	0.02	0.02
Catastrophic accident	–	–	0.1	–
Waste Management				
Occupational:				
Rad-induced cancer	–	–	4.6×10^{-4}	5.4×10^{-4}
Decommissioning radiological	–	–	5×10^{-3}	5×10^{-3}
Decommissioning non-radiological	–	–	8×10^{-4}	0.07
Public:				
Rad-induced cancer	–	–	8×10^{-6}	7×10^{-6}

TABLE 7.9. *Possible health damage resulting from a complete annual coal fuel cycle of a coal-fired power plant (per GW(e))*

	Causes of damage					
	Radiation			Non-radiational etiological diseases		
Type of damage	Personnel in mines	Population due to station waste	Accidents on the production line: extraction and transportation of coal	Personnel during extraction and treatment of coal	Population due to station waste	All causes
No. of cases of premature death	0.09	0.044	3.42	5.51	290	299
No. of cases of disablement	0.09	0.044	130.2	10.76	290	431
Reduction of life expectancy (years)	0.9	0.44	492.5	55.1	8700	9249
Losses in work capacity (years)	0.45	0.22	2021	107.6	5800	7929

TABLE 7.10. *Cancer-induced mortality attributable to radiation from all stages of the nuclear fuel cycle (deaths per GW(e) produced)*

Stage in the fuel cycle	Personnel	Population	Total
Extraction of uranium ore	0.015	0.0	0.015
Treatment of ore and manufacture of fuel	0.01	0.0	0.01
Receipt of energy at plant	0.1	0.015	0.115
Regeneration of fuel	0.12	0.044	0.164
Dismantling of reactor	0.003	0.0	0.003
Research investigations	0.05	0.001	0.051
All stages	0.3	0.06	0.36

from the four chief sources of energy are shown in Table 7.11 (Hamilton and Manne, 1977).

3. Comparative Assessments and the Exposure Commitment Method

It was stated at the outset that the dose commitment concept was developed to assess total effects of released radioactive material in the environment. This

TABLE 7.11. *Estimated health effects in 1975 associated with production of electric power in the USA*

Fuel	KWh(e) × 10^{9a}	Equivalent no. 100-MW(e) plants	Estimated deaths	Estimated disabilities
Coal	844	128	1900–15,000	25,000–39,000
Oil	292	44	88– 4000	4000–7000
Gas	297	45	6	600
Nuclear	168	26	18–42	130–470
TOTALS	1604	243	2000–19,000	29,000–48,000

[a] Preliminary.

has been developed as the exposure commitment method for assessing the effects of other environmental pollutants. This methodology allows comparative environmental impacts to be displayed. As an example of this, the results presented by UNSCEAR that compare the collective dose commitments from nuclear power with those of other sources of radiation (Butler and Hyslop, 1980) are of interest.

It is necessary to develop and particularly to apply the method on the basis of a unit measure of a given practice in order to provide a rational approach to the comparative assessment of environmental impacts of coal combustion and conversion, and other fuel cycles.

4. References

BARRY, P. J. (1979). *An Introduction to the Exposure Commitment Concept with Reference to Environmental Mercury.* Report No. 12. Monitoring and Assessment Research Centre, London.

BENNETT, B. G. (1981). The exposure commitment method in environmental pollutant assessment. *Environmental Monitoring and Assessment,* **1,** 21–36.

BREWER, S. T. (1976). Quantification and comparison of external costs of nuclear and fossil electrical power systems. *Energy and the Environment-Cost-Benefit Analysis* (ed. by R. A. Karam and K. Z. Morgan). *Energy, Suppl.* **1,** 506–30. Pergamon Press, New York.

BUCHANAN, B. G. and BENNETT, B. G. (1980). *Relevance of the Commitment Method in Environmental Monitoring and Pollutant Assessment.* Report of the Monitoring and Assessment Research Centre, London.

BUTLER, G. C. (1978). Estimation of doses and integrated doses. *Principles of Ecotoxicology,* SCOPE Report 12 (ed. by G. C. Butler), pp. 98–112. John Wiley, Chichester.

BUTLER, G. C. and HYSLOP, C. (1980). Radioactive contamination of the environment. *Handbook of Environmental Chemistry* (ed. by O. Hutzinger). Springer-Verlag, Heidelberg.

BUTLER, G. C., KAROL, I. L., LINDELL, B. G., STEVENS, D. J. and ZELENY, V. (1972). Assessment and control of environmental contamination: experience with artificial radioactivity. *Biological Conservation,* **4,** 177–84.

CAMPLIN, W. C. (1980). *Coal-fired Power Stations—the Radiological Impact of Effluent Discharges to the Atmosphere.* National Radiological Protection Board, R107. HMSO, London.

CHATTERJEE, B., HOTZL, H., ROSNER, G. and WINKLER, R. (1980). Untersuchungen über die Emissionen von Radionukliden aus Kohlenkraftwerken. GSF-Bericht. S-617.

HALBRITTER, G., BRAÜTIGAM, F. W., FLUCK, F. W. and LESSMANN, E. (1981). Contribution to a comparative environmental impact assessment of the use of coal and nuclear energy for electricity generation for selected German site conditions. *International Symposium on Health Impacts of Different Sources of Energy.* Nashville, Tennessee.

HAMILTON, L. D. (1980). Comparative risk assessment in energy industry. *Proc. Symp. on Health Risk Analysis.* Gatlinburg, Tennessee.

HAMILTON, L. D. (1984). Health and environmental risks of energy systems. *Risks and Benefits of Energy Systems,* pp. 21–57. International Atomic Energy Agency, Vienna.

HAMILTON, L. D. and MANNE, A. S. (1977). Health and economic costs of alternative energy sources. *International Conference on Nuclear Power and Its Fuel Cycle.* IAEA-CN-36/448. IAEA, Vienna.

ICRP (1976). *Recommendations of the International Commission on Radiological Protection.* ICRP Publication 26. Pergamon, Oxford.

ICRP (1979). *Limits for Intakes of Radionuclides by Workers.* ICRP Publication 30 (supplement to Part 1), Annals of ICRP 3.

INTL. SYMP. (1978). International Symposium on General Air Pollution and Human Health with Special Reference to Long-term Effects. *Environmental Health Perspectives,* **22,** 1–123.

KELLY, G. N. (1975). *The Predicted Radiation Exposure of the Population of the European Community Resulting from Discharges of Krypton-85, Tritium, Carbon-14 and Iodine-129 from the Nuclear Power Industry to the Year 2000.* Commission of the European Communities, Luxembourg.

KOLB, W. (1979). Radioaktive Stoffe in Flugaschen aus Kohlenkraftwerken. *PTB-Mitteilungen* **89,** 77–82.

LINDELL, B. (1960). An approach to the question of computing doses and effects from fall-out. *Health Physics,* **2,** 341–65.

LINDELL, B. (1978). Source-related detriment and the commitment concept: applying the principles of radiation protection to non-radioactive pollutants. *Ambio,* **7,** 250–9.

MCBRIDE, J. P., MOORE, R. E., WITHERSPOON, J. P. & BLANCO, R. E. (1977). *Radiological Impact of Airborne Effluents of Coal-fired and Nuclear Power Plants.* ORNL-5315, Oak Ridge National Laboratory, Tennessee.

MINISTRY FOR RESEARCH AND TECHNOLOGY (1979). *German Risk Study for Nuclear Power Plants.* TUV Rheinland, Cologne.

NATUSCH, D. F. S. (1978). Potentially carcinogenic species emitted to the atmosphere by fossil-fueled power plants. *Environmental Health Perspectives,* **22,** 79–90.

NISHIWAKI, Y., TSUNETOSHI, Y., SHIMIZU, T., UEDA, M., NAKAYAMA, N., TAKAHASHI, H., ICHINOSAWA, A., KAJIHARA, S., OHSHINO, A., OGINO, M. and SAKAKI, K. (1971). Atmospheric contamination of industrial areas including fossil-fuel power stations, and a method of evaluating possible effects on inhabitants. *Environmental Aspects of Nuclear Power Stations,* pp. 247–78. STI/PUB/261. International Atomic Energy Agency, Vienna.

O'BRIEN, B. J. (1979). *The Exposure Commitment Method with Application to Exposure of Man to Lead Pollution.* Report No. 13. Monitoring and Assessment Research Centre, London.

ULMER, W. T. (1977). Wirkung von SO_2 in der Atemluft. *VGB-Kraft-werkstechnik,* **57,** Heft 8, August.

UNEP (1981). *Environmental Impacts of Production and Use of Energy,* Vol. 1. Study Director E. El-Hinnawi. Tycooly International, Dublin.

UNSCEAR (1977). *Sources and Effects of Ionizing Radiation.* Official Records of the General Assembly, Thirty-second Session Suppl. No. 40 (A/32/40). United Nations, New York.

USDOE (1979). *Technology for Commercial Radioactive Waste Management.* DOE/ET-028, US Department of Energy, Washington.

WILSON, R. (1978). Risks caused by low levels of pollution..*The Yale Journal of Biology and Medicine,* **51,** 37–51.

WILSON, R. and JONES, W. J. (1974). *Energy, Ecology and the Environment.* Academic Press, New York.

8

The Environmental Significance of Trace Elements from Coal Combustion and Conversion Processes

1. Occurrence and Distribution of Trace Elements in Coal 171
  1.1. The Origin of Trace Elements in Coal 171
    1.1.1. The chemical association of trace elements with coal 172
  1.2. Concentrations of Trace Elements in Coal 172
    1.2.1. Evaluation of reported values 172
    1.2.2. Variation in trace element content 173
    1.2.3. Estimation of average trace element content and enrichment with respect to crustal abundance 177
2. Mobilization of Trace Elements by Coal Utilization 178
  2.1. Conventional Coal-fired Power Plants 178
    2.1.1. Behaviour of trace elements within power plants 179
  2.2. Other Combustion Facilities 184
    2.2.1. Domestic coal combustion 184
  2.3. Coal Conversion Processes 185
    2.3.1. Gasification 186
    2.3.2. Liquefaction 190
3. Environmental Effects of Coal Utilization 192
  3.1. Atmospheric Emissions of Trace Elements 192
    3.1.1. Fallout in the vicinity of power plants 192
  3.2. Solid Wastes from Coal Utilization 202
    3.2.1. Coal combustion 202
    3.2.2. Coal conversion processes 206
4. Conclusions 207
  4.1. Summary of Evidence from Existing Combustion Processes 207
  4.2. Predicting Likely Effects of New Conversion Technologies 209
5. References 211

1. Occurrence and Distribution of Trace Elements in Coal

1.1. The Origin of Trace Elements in Coal

The large numbers of elements present in coal are derived from several processes which took place before, during and after its formation, Coalification itself consists of two distinct stages (Firth, 1971). The first involves formation of peat from the carbonaceous flora by bacterial and fungal action, resulting in the production of large amounts of humic acids and chelating agents, which can efficiently take up cations. The covering of the peat layer by sediments marks the beginning of the second stage, in which biological processes are replaced by the action of pressure, temperature and time.

Some of the original trace elements, taken up by the plant species of the carbonaceous swamps, will have been retained after their breakdown and incorporated into the developing coal matter. Contemporaneous sedimentation may also have introduced high concentrations of minerals, with humic acids from the decaying flora solubilizing and binding these elements. Trace elements will also have been gained from and lost to inflowing ground-water during and after the coalification process. Finally, migration of the metal-rich magmatic fluid to areas adjacent to coal deposits can result in epigenic mineralization, as has occurred in the South Wales Coalfield (Firth, 1971).

1.1.1. The chemical association of trace elements with coal

Trace elements in coal may be classified according to whether they are primarily associated with the organic fraction of coal, with the mineral fraction or with both. Ruch, Gluskoter and Shimp (1974) examined the "organic affinity" of trace elements in various coals by analysis of specific gravity fractions. Most of the elements of environmental concern, including As, Cd, Hg, Pb and Zn, were concentrated in the inorganic fraction of coal. Some, such as Cr, Cu and Se, were present in both the mineral and organic matter, but those also tended to be inorganically associated; B and Be were found to be largely combined with the organic fraction of coal. In view of these findings, Ruch, Gluskoter and Shimp (1974) suggested that those elements with organic affinities were probably inherited from an early period of coal swamp formation. On the other hand, those in inorganic association originated from emplacement by epigenetic mineralization, as they were often present in discrete minerals such as sulphides and carbonates.

1.2. Concentrations of Trace Elements in Coal

1.2.1. Evaluation of reported values

Accurate values for the trace element content in coal are required before any assessment of their significance can be made. Prior to 1970, however, elemental measurements in coal employed high temperatures to remove the organic fraction before analysis (Clarke and Swaine, 1962; Zubovic, Sheffey and Stadnichenko, 1967; Abernathy, Peterson and Gibson, 1969). This method resulted in losses of volatile elements, an underestimate of their concentration and the production of unreliable data. More sophisticated analytical techniques have been developed in recent years and standard coal samples from the US Bureau of Standards are available for inter-laboratory comparison (Ondov *et al.*, 1975).

TABLE 8.1. *Concentrations ($\mu g\ g^{-1}$ dry weight) of trace elements for 82 coals from the Illinois Basin (Ruch, Gluskoter and Shimp, 1974)*

Element	Mean	Standard deviation	Range
Antimony (Sb)	1.35	1.42	0.2 – 8.9
Arsenic (As)	14.9	18.9	1.7 – 93.0
Beryllium (Be)	1.72	0.83	0.5 – 4.0
Boron (B)	113.8	51.7	12.0 –224.0
Cadmium (Cd)	2.89	8.32	0.1 – 65.0
Chromium (Cr)	14.1	7.48	4.0 – 54.0
Fluorine (F)	59.3	19.8	30.0 –143.0
Lead (Pb)	39.8	45.9	4.0 –128.0
Mercury (Hg)	0.21	0.22	0.03– 1.6
Nickel (Ni)	22.4	10.8	8.0 – 68.0
Selenium (Se)	1.99	0.93	0.45– 7.7

Unrepresentative trace element levels in coal may also be found in those geological studies concerned with the exploitation of coal as a commercial source of certain elements. In such reports, only unusually high values were selected for publication (see, for example, Bethell, 1962).

In an attempt to obtain a true picture of the variation in trace element content of coal for a particular region, studies such as the Illinois Geological Survey (Ruch, Gluskoter and Shimp, 1974) have analysed large numbers of samples collected from the same seam. The results of this investigation, summarized in Table 8.1, revealed that the elements As, Cd, Hg, Pb and Zn each displayed a highly skewed distribution, with very large ranges and standard deviations. A second group, comprising B, Cr, F, Ni, Se and V, were approximately normally distributed with smaller ranges and standard deviations.

1.2.2. Variation in trace element content

Reported concentrations of trace elements from different coals often display considerable variability. Table 8.2 gives the concentration of some elements in coals from the UK and the FRG. Many compilations now give extensive data on the range of trace elements in coal (Lim, 1979; Torrey, 1978; Valkovic, 1983). It would be desirable to ascertain whether this variability could be explained by or related to the rank of coal or its geographical location.

Coal rank. In view of the literature available at the time, Bethell (1962) concluded that trace element content generally decreased with increasing rank of coal. Although there have apparently been no recent assessments concerned specifically with this issue, there is little evidence available to support the above suggestion. Indeed, in the South Wales coalfield it is the high-rank,

TABLE 8.2. *Concentrations ($\mu g\ g^{-1}$ dry weight) of trace elements for 15 hard coals from British coalfields and six hard coal samples from the Federal Republic of Germany (Sabbioni et al., 1983)*

Element	UK Mean	UK standard deviation	UK Range	FRG Mean	FRG standard deviation	FRG Range
Antimony (Sb)	3.3	2.3	1 -10	3.5	1.4	1.8-5.0
Arsenic (As)	16.8	15.9	1.9-63.4	6.8	4.3	3.0-13.5
Cadmium (Cd)	0.3	0.2	0.07-0.56	–	–	–
Chromium (Cr)	31.9	7.9	15.8-44.5	17.5	6.7	10.1-28.9
Lead (Pb)	21.7	10.2	9.0-50.7	–	–	–
Mercury (Hg)	0.28	0.3	0.025-1.0	0.42	0.4	0.1-1.0
Selenium (Se)	2.9	1.2	0.3 -5.1	1.5	0.5	0.9-2.3

anthracite coals which contain the highest concentrations of trace elements (Chatterjee and Pooley, 1977).

Table 8.3 presents trace element data, taken from Schwitzgebel *et al.* (1975), for three coals from the USA; a lignite from North Dakota and two sub-bituminous samples, both from Wyoming. With the exception of As and B, concentrations of trace elements in the low-grade lignite are similar or actually lower than in the two sub-bituminous coals from the same state. Table 8.3 also gives the elemental concentrations for another lignite from the FRG (Heinrichs, 1977). Although only a limited number of elements were determined, it is apparent that the values obtained are much lower than in either of the two coal ranks in the American study.

TABLE 8.3. *Trace element content ($\mu g\ g^{-1}$ dry weight) in coals of different rank from the USA and FRG*

	Coal rank			
Element	Lignite (N. Dakota)[a]	Sub-bituminous (Wyoming)[a] 1	Sub-bituminous (Wyoming)[a] 2	Lignite (FRG)[b]
As	7.8	0.8	2.7	1.4
B	146.0	49.0	32.5	ND
Cd	0.2	0.2	0.1	0.02
Cr	12.7	21.0	9.2	3.2
F	54.0	138.0	68.0	94
Pb	0.8	4.2	2.4	1.1
Hg	0.07	0.13	0.14	0.05
Ni	5.5	8.5	2.2	1.3
Se	1.3	2.2	1.7	0.24
U	1.5	1.0	1.3	ND
V	15.0	49.0	20.8	4.2
Zn	7.5	24.0	4.2	2.0

ND = Not determined.
[a] Schwitzgebel *et al.* (1975).
[b] Heinrichs (1977, 1983).

TABLE 8.4. *Average trace element concentration* ($\mu g\ g^{-1}$) *in coal by state in the USA (Dvorak, 1977)*

	N. Appalachian		S. Appalachian				Eastern interior			Powder River region		Four Corners	
Element	W. Va.	Penn.	Eastern Ky.	Ala.	Va.	Tenn.	Ill.	Western Ky.	Ind.	Mont.	Wyo.	New Mexico	Ariz.
As	9	16	6	13	10	9	6	7	7	6	1	2	2
B	20	15	19	30	13	24	81	70	85	60	36	43	49
Cd	–	–	–	–	–	–	2.9	–	–	0.1	0.46	< 2.6	< 0.54
Cr	19	24	19	19	20	19	29	18	19	3	5.8	11	9.7
Cu	11	13	11	14	13	11	8.3	8.8	9.7	3.2	4.4	5.9	4.9
F	70	90	30	90	50	120	59	–	50	70	160	160	70
Pb	4.9	5.2	4	3.7	6.1	4.9	33	6.4	7.2	4.8	0.61	4.7	3.9
Hg	0.12	0.20	–	–	–	–	0.18	–	0.08	0.07	0.05	0.08	0.05
Ni	18	20	16	17	22	16	25	16	33	3.3	4.1	8.1	4.9
Se	3.4	3.7	3.1	5.1	4.4	4.9	2	3.1	4	3	0.8	2	2.1
V	30	33	29	31	33	34	35	32	35	12	15	25	9.7
Zn	17	22	15	22	23	23	140	48	73	42	37	19	9.7

It should be noted that low-grade coal deposits have been associated with elevated concentrations of both As and U in two specific areas of the world. It has been known for some time that certain lignite deposits in the western USA contain high concentrations of U and Th (Denson, 1959). However, the average figure of 25 $\mu g\ g^{-1}$ U for all western lignites given by Smith (1977) requires substantiation. In particular areas, these lignite deposits do certainly contain high concentrations of U; in Willeston Basin, Barber and Giorgio (1977) report a value of 0.18 %. In spite of the environmental implications, it is not known whether these uraniferous coals are being utilized for combustion on any large scale.

In the case of As, particular lignite deposits in Czechoslovakia contain from 900 to 1500 $\mu g\ g^{-1}$; serious As contamination has arisen in the vicinity of the power plant which consumes this coal (Bencko and Symon, 1977).

Geographical location. In the USA, where the most comprehensive studies have taken place, the trace element content of coal is known to vary considerably between regions. Table 8.4 assigns values to the main coal-bearing states; the large range encountered makes it difficult to assess regional patterns or the national average; As, however, would appear to decrease in concentration from eastern to western coalfields. In passing, it is encouraging to note that the analytical values for Illinois in Table 8.4 are similar to those found by Ruch, Gluskoter and Shimp (1974).

National averages have been published for both Australian and British coals (Table 8.5). Compared with coals from the USA and the UK, and the coal burnt in power stations in nine member states of the European

TABLE 8.5. *Trace element content ($\mu g\ g^{-1}$) of British and Australian coals*

	Australian bituminous coals[a]		British coals[b]	
Element	Mean	Range	Mean	Range
As	3	>1–55	18	4–40
B	60	1.5–300	NG	NG
Cd	0.10	0.05–0.2	0.4	0.3–0.8
Cr	6	1.5–30	33.6	12–50
F	80	15–500	114	98–130
Hg	0.10	0.03–0.4	0.5	0.4–0.6
Ni	15	0.8–70	27.9	12–40
Pb	10	1.5–60	38	28–60
Se	0.79	0.21–2.5	2.8	1.8–4.4
U	2	0.4–5	1.3	0.5–2.3
V	20	4–90	76	38–134
Zn	25	12–73	NG	–

NG = Not given.

[a] Swaine (1977, 1979).

[b] Gibson (1979).

No sample size available for either study.

TABLE 8.6. *Weighted most reasonable mean values of trace metal content in hard coals ($\mu g\, g^{-1}$) burnt in power stations in nine member states of the Commission of the European Communities (Sabbioni et al., 1983)*

Element	Member state[a]								
	B	D	DK	F	I	IRL	L	NL	UK
As	6.5	14.5	10	9.7	10	10	10	10	16.8
Cd	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.3
Cr	60	25	44	36	44	44	44	44	32
Cu	42	33	22	8.4	22	22	22	22	18
Hg	0.38	0.4	0.28	0.2	0.28	0.28	0.28	0.28	0.28
Mo	2.2	14	4.6	4.6	4.6	4.6	4.6	4.6	3.0
Ni	55	45	38	38	38	38	38	38	38
Pb	85	68	53	44	53	53	53	53	22
Sb	1.9	1.4	2.2	3.4	2.2	2.2	2.2	2.2	3.3
Se	1.6	1.5	1.7	1.3	1.7	1.7	1.7	1.7	2.8
Tl	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
Th	4.4	2.1	4.0	6	4.0	4.0	4.0	4.0	3.8
U	2.1	1.0	2.2	3.1	2.2	2.2	2.2	2.2	2.0
V	72	75	65	65	65	65	65	65	48
Zn	165	73	107	172	107	107	107	107	57

[a] B – Belgium, D – Federal Republic of Germany, DK – Denmark, F – France, I – Italy, IRL – Republic of Ireland, L – Luxembourg, NL – The Netherlands, UK – United Kingdom.

Greece was not included, as power production by conventional thermal power plants in Greece is at present exclusively based on the burning of oil and lignite.

Community (Table 8.6), Australian coals are generally lower in trace elements. Swaine (1977) suggests this is due to the low levels of pyrite and other sulphide minerals in these coals; certainly elements such as As, Cu, Ni and Pb are known to be found as sulphides in nature, including some coals (Swaine, 1977).

1.2.3. Estimation of average trace element content and enrichment with respect to crustal abundance

Table 8.7 presents an estimated average trace element content of US and Australian coal. The figures given have been assessed after considering the results of studies already discussed and also those of Bertine and Goldberg (1971), Gladney (1974), Swanson *et al.* (1976), Coles, Ragaini and Ondov (1978) and Van Hook (1978). These values have also been compared to the average concentration of each element in the earth's crust, using the figures of Mason (1966). This ratio, known as an enrichment factor, is a simple way of assessing whether the element is enriched or depleted in coal compared to its crustal abundance. It is apparent from Table 8.7 that the trace elements given may be divided into three groups, according to whether they show enrichment, depletion or are actually present at similar concentrations to the earth's crust. Five elements show enrichment factors greater than one; these are As, B, Cd (US coals), Hg and Se; Pb and U show no significant trend of enrichment

TABLE 8.7. *Estimated average trace element content of shale, coal and coal enrichment factor relative to crustal abundance*

Element	Mean value shale[a] ($\mu g\,g^{-1}$)	Mean value coal[b] ($\mu g\,g^{-1}$) USA	Mean value coal[b] ($\mu g\,g^{-1}$) Australia	Enrichment factor[c] USA	Enrichment factor[c] Australia
As	13	15	3	8.3	1.7
B	100	50	60	5.0	6.0
Cd	0.3	1.3	0.1	6.5	0.5
Cr	90	15	6.0	0.2	0.1
Cu	45	19	1.5	0.3	0.03
F	740	74	80	0.1	0.1
Hg	0.4	0.18	0.1	2.3	1.3
Ni	68	15	15	0.2	0.2
Pb	20	16	10	1.3	0.8
Se	0.6	4.1	0.79	82	16
Th	12	4.7	2.7	0.7	0.4
U	3.7	1.8	2.0	1.0	1.1
V	130	20	20	0.1	0.1
Zn	95	39	25	0.6	0.4

[a] Turekin and Wedepohl (1961).

[b] Swanson *et al.* (1976); Swaine (1977).

[c] Calculated as the ratio of the concentration in coal compared to its average crustal value (Mason, 1966).

or depletion, while all others are depleted in coal compared to the crustal value.

2. Mobilization of Trace Elements by Coal Utilization

2.1. Conventional Coal-fired Power Plants

Although potentially toxic elements are generally present at relatively low concentrations in coal, significant mobilization still occurs, because of the very large quantities of coal consumed. The annual world production of coal in 1978 was estimated to be 2.974×10^9 tonnes (Quenton, 1980). A single large power plant (> 2000 MW) will burn as much as 10^7 tonnes of coal a year (Wiersma and Crockett, 1978). In addition, the combustion process may produce physicochemical alterations to trace elements and their compounds which enhance their toxicity.

Coal combustion in modern electric power plants takes place in furnaces operating at about 1600°C. When coal is burnt, a portion of the non-combustible mineral matter will be retained in the furnace as either slag or bottom ash, while the rest will leave as fly ash with the flue gases and volatilized minerals. A further fractionation occurs when the flue stream passes the particulate control devices. These may remove up to 99% of the larger fly ash particulates, but are less efficient in collecting the smaller

(< 1 μm) particles and vapour. The proportion of bottom ash, slag and fly ash produced will vary not only with the ash content of coal, but with the plant design and the particulate collection device employed. For example, ash removed in a wet condition may have considerably higher elemental enrichment. In cyclone-fed units the total ash produced is divided equally between bottom ash and fly ash, while in the more commonly occurring pulverized coal plants over 90 % of the total ash may be fly ash (Bolton *et al.*, 1973).

2.1.1. Behaviour of trace elements within power plants

Several comprehensive investigations into the behaviour of trace elements in power plants have involved a detailed examination of the partitioning of trace elements between the various exit streams, to allow an assessment of their mass-flow rates. Such mass-balance studies have been carried out for a whole range of trace elements (Gladney, 1974; Kaakinen *et al.*, 1975; Klein *et al.*, 1975; Schwitzgebel *et al.*, 1975) and also for single elements (Billings and Matson, 1972; Andren, Klein and Talmi, 1975). The results obtained have been explained in relation to the chemical properties of trace elements and their compounds and to the operating conditions within the power plant.

Partitioning of elements. Mass-balance studies reveal that coal combustion in power plants produced a consistent and selective partitioning of trace elements between the various exit streams, permitting the classification of elements into three groups:

Group I—These elements are partitioned equally between bottom ash and fly ash and are not concentrated in the outlet fly ash. Elements in this group include Al, Ba, Ca, Ce, Cs, Fe, K, Mg, Mn and Th.

Group II—These elements are concentrated in the inlet fly ash compared to the bottom ash and the outlet fly ash compared to the inlet fly ash. This group consists of As, Cd, Cu, Pb, Sb, Se and Zn.

Group III—Elements in this group are mainly present in the gas phase and are depleted in all ashes. This group contains three elements, Br, Hg and I.

Several elements, Cr, Ni, U and V, exhibit intermediate behaviour between Group I and Group II.

This classification may also be extended to the concentration of trace elements as a function of the particle size of fly ash. Natusch, Wallace and Evans (1974) reported that concentrations of elements in Group II increase as the particle size decreases, while Group I elements showed no such relationship, being present in roughly equal concentrations in the various size fractions of fly ash.

An enrichment mechanism. Natusch, Wallace and Evans (1974) explained the enrichment of certain trace elements in smaller particles by a volatilization-condensation mechanism; this also accounts for the observed difference in partitioning between the various exit streams. The model suggests that when coal is burnt, elements from Group II are volatilized and will thus be depleted from the slag and bottom ash. As the flue gases cook, these elements will preferentially condense out on the smaller fly ash particles, because such particles have a larger surface-area-to-volume ratio. Elements in Group I will remain condensed at the temperature of coal combustion and form the bottom ash and the fly ash particles on which the volatile elements condense. The selective retention of courser fly ash particles by the collection devices will further enhance the small particle association of volatile elements. Group III elements, being very volatile, will mostly remain in the gas phase during passage through the power plant, although they may be, in part, associated with fly ash.

Although the manner in which trace elements are partitioned is related to their boiling point, their chemical form in coal also plays an important role. Both Kaakinen *et al.* (1975) and Klein *et al.* (1975) point out that all those elements in coal classified in Group I are lithophiles in crustal rocks, being associated with aluminosilicate minerals. Conversely, elements in Group II are chalcophiles, as they are presented as sulphide minerals in crustal components. Klein *et al.* (1975) suggest that when coal is burnt, the lithophile elements remain condensed, but chalcophiles are decomposed in the reducing conditions of the combustion zone, forming volatile species which are vaporized. However, it has been pointed out (Stinespring and Stewart, 1981) that the adsorption-condensation process is not the only one that can account for the observed surface enrichment. Ash particles produced during coal combustion are formed as a result of thermal treatment of aluminosilicate minerals. Many of the trace elements that enrich the surface are present in the aluminosilicate mineral. At elevated temperatures diffusive transport to the surface of the ash particle occurs (surface segregation), thus contributing to surface enrichment.

The combustion characteristics of uranium illustrates the need to consider the chemical composition of trace elements in coal, in order to understand their behaviour. Although uranium displays little enrichment on fly ash compared to bottom ash, it is associated with the smaller fly ash particles (Coles, Ragaini and Ondov, 1978) and is thus intermediate between Group I and Group II elements. Uranium is known to exist in coal as both the silicate mineral coffinite and uraninite (UO_2). Upon combustion, the refractory coffinite will remain in the slag and bottom ash, but the uraninite will be vaporized, condensing on fly ash particles when the flue gases cool. Furthermore, the silicate content of coal may be an additional factor in the combustion chemistry of uranium, as this coal determines the relative proportions of the two forms of uranium and this affects its combustion behaviour. The partitioning of other trace elements may also be affected by the content of major elements in coal. Kaakinen *et al.* (1975), for example, report that arsenic appears to be retained in ashes of high calcium coals, while in low calcium coals it is more efficiently volatilized.

Enrichment of trace elements in fly ash. The extent of trace element enrichment on fly ash is most conveniently expressed as an enrichment factor (EF). This is the ratio of the concentration of an element (X) to that of aluminium, or another refractory element, in the fly ash, divided by the corresponding abundance ratios in crustal rocks (Mason, 1966).

$$\mathrm{EF} = \frac{([\mathrm{X}]/[\mathrm{Al}])\ \text{fly ash}}{([\mathrm{X}]/[\mathrm{Al}])\ \text{crust}}$$

This normalization technique takes into account the apparent enrichment of fly ash caused by carbon losses in the combustion of coal.

Figure 8.1 shows trace element enrichment factors relative to crustal abundance of suspended fly ash particles collected from three coal-fired power plants in the USA: Chalk Point (Gladney, 1974; Gladney *et al.*, 1976); the Allen Steam Plant (Klein *et al.*, 1975) and an unnamed facility (Natusch, Wallace and Evans, 1974). The values found are generally comparable at the three installations, with significant enrichment factors for As, Cd, I, Pb, Sb, Se and Zn. The Allen Steam Plant did, however, produce greater enrichments than Chalk Point for Co, Cr, Fe, Pb, Sb and V. This difference may be explained by the higher Combustion temperature employed at the Allen Plant, producing a greater vaporization of the above elements in the furnace. Such a suggestion is supported by the finding that As and Se showed greater EF values at Chalk Point; presumably the lower temperatures encountered at this

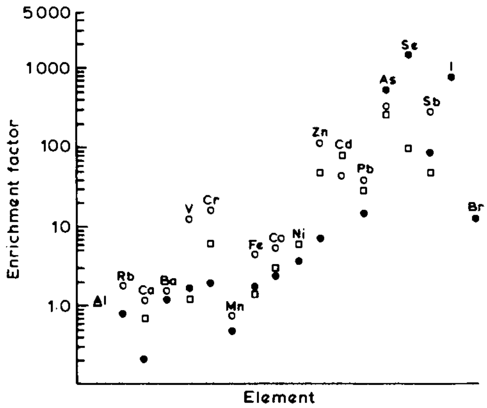


FIG. 8.1. Elemental enrichment factors of suspended fly ash particles relative to crustal abundance from three US power plants: ●, Chalk Point (Gladney, 1974); O, Allen Steam Plant (Klein et al., 1975); □, unnamed facility (Natusch, Wallace and Evans, 1974).

plant allow a greater proportion of these two volatile elements to condense from the gas phase.

The extent of trace element enrichment on the finer fly ash particle is a matter of some importance, because it is these particles which can pass through the collection devices and are deposited efficiently in the pulmonary region of the lung. Natusch, Wallace and Evans (1974) were the first to report dramatic increases in the concentrations of several toxic elements, As, Cr, Ni and Pb, on submicron fly ash particles. Unfortunately, these findings are difficult to evaluate in relation to later studies, as no information was provided on the elemental composition of the original coal. For this reason, no enrichment factors can be calculated for comparisons with other investigations.

In a more comprehensive study, Gladney *et al.* (1976) fractioned suspended fly ash particles with a cascade impactor and examined enrichment factors for a suite of elements. In contrast to Natusch, Wallace and Evans (1974), both Cr and Ni generally showed very little increased enrichment with decreasing particle size, although they do so in some Australian coals (Swaine, 1979). Furthermore, although enrichment factors for Pb and Sb increased on the finer particles, the extent of enrichment was much less than found by Natusch, Wallace and Evans (1974). The volatile elements As and Se also displayed an increased enrichment on smaller particles, but as Fig. 8.2 shows there was also a definite minimum in the mid-sized (1–5 μm) range for Se. This bimodal behaviour cannot be explained by the simple volatilization-condensation model discussed above, and recent work (Smith, Campbell and Nielson, 1979a, 1979b) indicates that little enrichment occurs below particle diameter of 1 μm. These authors suggest that submicron particles may form by larger particles "bursting" during gas expansion, producing many very small

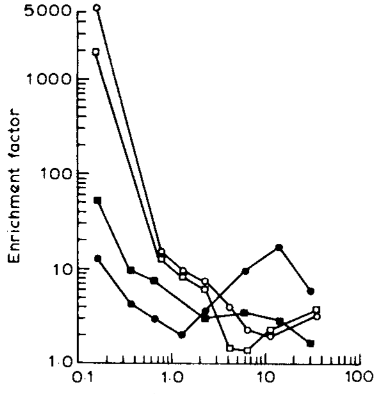


FIG. 8.2. Enrichment factors of As and Se with respect to coal from two US power plants, expressed as a function of particle size: ●, Se; ■, As Gladney *et al.*, 1976). O, Se; □, As (Ondov, Ragaini and Biermann, 1978).

particles which ultimately form particles of 0.1–1.0 μm diameter by coagulation.

The result of Gladney *et al.* (1976) must also be viewed with caution, however, as serious errors in using cascade impactors to estimate enrichment factors as a function of particle size in power plants were reported by Ondov, Ragaini and Biermann (1978). By use of scanning electron microscopy, the latter study found that those particles collected on the upper impactor stages were actually much smaller than indicated by the 50 % cut-off diameters given for the impactor. Furthermore, on the back-up filters, large particles (> 2 μm) constituted more than 90 % of the total mass and 20–30 % of volatile elements such as As and Se. Back-up filters are employed to collect the respirable, submicron particles, too small to be retained by impaction. In view of these findings, Ondov, Ragaini and Biermann (1978) suggested that enrichment factors on small particles may be more than an order of magnitude greater when the large particle component on the back-up filter is taken into account.

In Fig. 8.2 enrichment factors of As and Se vs. particle size have been calculated from the data of Ondov, Ragaini and Biermann (1978), employing the elemental coal consumption given in a different study of the same power plant (Coles *et al.*, 1979). Values on the back-up filter have been corrected for bounce-off effects and are much larger than found by Gladney *et al.* (1976). Additionally, Se shows no evidence of the bimodal behaviour reported by Gladney *et al.* (1976).

Campbell *et al.* (1978) carefully size-separated fly ash collected by an electrostatic precipitator, using a combination of an air centrifugation-elutriation technique; the accuracy of the fractionation was confirmed by scanning electron microscopy. Concentrations of As, Pb and Se showed a dramatic increase with decreasing particle size, with no evidence of bimodal behaviour. Additionally, and in contrast to Gladney *et al.* (1976), Cr, Ni and Zn also displayed small particle association. Unfortunately, it is again not possible to compare these results directly with other studies, as no information on the elemental content of coal was provided.

Clearly, further work is required in order to clarify the particle-size distribution of potentially toxic trace elements in suspended fly ash. Initially, scanning electron microscopy (SEM) should be used to establish the reliability of the fractionation technique employed. Attention can then be directed to the extent of enrichment on submicron particles. Few studies have used SEM to check the actual particle sizes of the fractions being studied. Raask and Goetz (1981) employed the technique when examining the surface morphology of size-graded samples. Although they found that 50 % of the chimney stack solids were in the respirable range (< 5 μm in diameter) compared with only 10 % of ash from the electrostatic precipitators, these were chiefly spherical particles of aluminosilicates and iron oxides and did not show significant trace element enrichment over larger diameter particles. As to whether any variation in enrichment can be linked to specific differences in the operating conditions of coal-fired power plants, a study by Hee *et al.* (1982) showed that stack emissions of Al, Ca, Fe, Na, K and Mg were greatest from a power plant burning high sulphur coal (2.8 % S) than from plants using coal with 0.41–0.52 % S.

The volatilization-condensation model suggests that volatile elements are deposited on the surface of suspended fly ash particles and that such elements may well pose a greater hazard than simple chemical analysis would indicate. To date, no studies have examined the surface predominance of potentially toxic elements on submicron particles, presumably due to technical difficulties. Nevertheless, Linton *et al.* (1976) found that both Pb and Zn showed significant increases in concentration near the surface of 75–100 μm fly ash particles. The layer of surface enrichment appeared to be about 0.1 μm in depth. By extrapolation, this would suggest that a large proportion of the trace element mass of a 1 μm particle would be present within this shell. However, although elements such as Na, C and S decreased with depth in both 1 μm and 2.6 μm particles (Campbell *et al.*, 1979), the relative thickness of the surface layers was similar in the two sized particles. It was to account for both the surface predominance of volatile elements and the smaller surface layer than predicted by the volatilization-condensation model that Campbell *et al.* (1979) proposed the mechanism involving the "bursting" of larger particles to form smaller fragments, with surface deposition of volatile elements occurring before, during and after the process.

As indicated previously, the volatilization-condensation model is not the only possible method of surface trace element enrichment of particles. Raask and Goetz (1981) have proposed that trace elements captured on ash and chimney stack solids may be distributed in four ways: (i) encapsulated or co-crystallized in aluminosilicate and iron oxide particles; (ii) dissolved on the surface layer of aluminosilicate particles when these were in a semi-molten state in the coal flame; (iii) submicronic size particles and co-crystalloids of sulphates, oxides or other oxycompounds captured on aluminosilicate and iron oxide particles; (iv) aerosols of discrete micronic particles and agglomerates of oxycompounds. In attempting to assess hazards from stack emissions not all the emphasis should be focused on enriched respirable particles. Gaseous emissions of highly volatile elements should be considered (particularly As, Se and Hg) and exposure routes through ingestion taken into account, as it is this that may result in the highest individual doses (Camplin, 1980).

2.2. *Other Combustion Facilities*

2.2.1. *Domestic coal combustion*

Coal burning in the domestic sector and away from centralized units has declined in recent years, but 15 % of the total UK consumption was still used for this purpose in 1977–8 (Gibson, 1979).

Not surprisingly, the reported combustion behaviour of trace elements in a domestic unit corresponds to that in power plants, with enhanced concentrations of chalcophiles (Group II elements) on fly ash, which further increase with decreasing particles size (Block and Dams, 1976). What was surprising, however, was that in the absence of any particulate collection control device, over half the suspended fly ash particles from this domestic appliance were smaller than 1 μm in diameter. The significance of this mass distribution is revealed when enrichment factors as a function of particle size are calculated for several elements from the raw data of Block and Dams (1975, 1976). Figure 8.3 reveals that As, Se and Zn each show increasing enrichment as particle size decreases. Compared to power plants, the values found on submicron particles are much greater than reported by Gladney *et al.* (1976) but similar to those computed from the results of Ondov, Ragaini and Biermann (1978). One possible explanation for the high enrichment factors encountered in this domestic appliance is the low temperature (40°C) at the point of particulate sampling in the chimney; this may allow efficient condensation of volatile elements to occur *in situ*. In order to establish whether such enrichments on submicron particles are characteristic of domestic heating units in general, it is necessary to perform further investigations of other appliances.

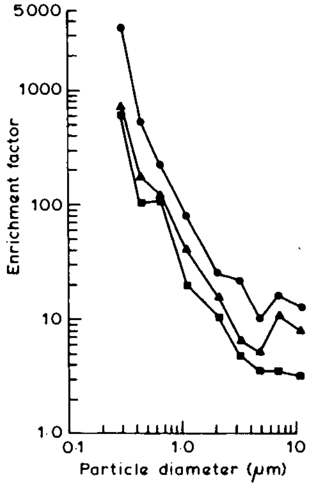


FIG. 8.3. Enrichment factors of selected elements with respect to coal, expressed as a function of particle size, from a domestic heating unit (Block and Dams, 1976): ●, Se; ▲ Zn; ■, As.

2.3. *Coal Conversion Processes*

There are only a few commercial-sized gasification plants in existence and the only liquefaction plant presently operating on a commercial scale is the SASOL plant in South Africa. However, a number of governments are interested in commercial coal conversion processes and the US Government once suggested the production of 2 million barrels of synthetic fuels per day by 1990 (Wade, 1979). It therefore seems desirable to attempt to assess the environmental acceptability of such processes and here the behaviour of trace elements in coal conversion plants is considered. As far as possible, information from commercial-sized conversion plants is evaluated (Bombaugh *et al.*, 1980; Patterson, 1980), but it has proved necessary to rely mainly upon results from bench-scale or pilot plant processes and extrapolation from conventional coal-fired power plants.

One factor which may be of general importance when evaluating the mobilization of trace elements from conversion processes is the choice of coal type to be converted. This is because a higher mineral content in coal, including high FeS_2 and As, has been reported to have a definite catalytic effect upon hydrogenation, the effect depending upon the precise quantity and composition of the mineral matter present (Mukherjee and Chowdhury, 1976). Thus, coals which may be selected for conversion because of this catalytic effect may contain elevated concentrations of those trace elements associated with the ash fraction (Ruch, Gluskoter and Shimp, 1974). It is of interest to note, for example, that the coals of North Assam display unusually high hydrogenation rates compared to other Indian coals and contain elevated concentrations of Cr (200 $\mu g\,g^{-1}$), Mo (200 $\mu g\,g^{-1}$), Ni (225 $\mu g\,g^{-1}$) and V (150 $\mu g\,g^{-1}$) (Mukherjee and Chowdhury, 1976).

2.3.1. Gasification

Gasification processes have been described in Chapter 6. The gas treatment steps following gasification-shift conversion, methanation and dehydration have features common to all processes.

The following assesses trace element behaviour for three gasification processes to illustrate the range of reaction conditions and effluent characteristics encountered.

The Synthane gasifier. An example of this process is the US Bureau of Mines laboratory scale gasifier (Oldham and Wetherold, 1977). In this two gas streams, one of them hydrogenated, are recombined, desulphurized and then methanated using a Ni catalyst.

The trace element content of several of the effluent streams from the laboratory-scale unit have been determined (Forney *et al.*, 1974; Oldham and Wetherold, 1977) and are summarized in Table 8.8. Gasification char is the major sink for most of the elements in the feed coal, and this has been confirmed recently (Koppenhaal *et al.*, 1979) in a study in which subbituminous feed coal was used. This work has also indicated, however, that high enrichment of trace metals takes place on filter fines through a volatilization-condensation mechanism similar to that observed in coal

TABLE 8.8. *Trace element content ($\mu g\ g^{-1}$) of feed coal and effluent streams from a laboratory synthane gasifier*

		Effluent streams			
Element	Feed coal	Filter fumes	Char	Tar	Condensate*
As	0.87	3.7	6.5	0.17	0.054
B	86	64	380	12	35.6
Cd	0.097	0.88	1.6	–	0.001
Co	14	17	95	0.09	–
Cr	170	47	240	7.1	< 0.001
F	490	610	150	0.97	23.5
Hg	0.1	0.2	ND	1.2	0.014
Mo	15	21	14	0.31	ND
Ni	43	12	25	1.2	0.028
Pb	0.55	2.2	21	0.22	0.33
Se	5.3	6.4	17	0.02	0.78
Th	3.0	5.8	4.3	0.06	ND
U	1.4	5.6	5.4	0.01	0.031
V	100	44	190	0.21	0.29
Zn	25	11	100	0.48	3.9

ND = Not determined.
Data from Forney *et al.*, 1974 (*Oldham and Wetherold, 1977).

combustion (Table 8.9). Furthermore, as the gasifier char is intended to be burnt in an auxiliary utility boiler to provide steam and electricity for the gasification unit, particulate control devices will have to be employed in the

TABLE 8.9. *Enrichment ratios (ER) in relation to crustal abundance for various elements of filter fines in a synthetic gasifier (Koppenaal et al., 1979)*

Non-enriched elements		Enriched elements	
Element	ER	Element	ER
Na	2.0	K	3.1
Mg	2.7	Rb	6.5
Al	1.0	Co	4.1
Si	1.2	Ni	3.0
Ca	0.8	Zn	7.0
Ti	1.1	Cu	9.3
Zr	2.9	Ga	4.7
Fe	0.8	Ge	11.0
Cr	1.6	As	6.2
Be	2.4	Br	11.0
Sr	1.2	Sn	3.3
Ba	1.1	Sb	5.7
Hf	1.5	I	14.0
Gd	1.5	Mo	7.3
		Pb	3.8
		Cd	6.5

TABLE 8.10. *Calculated partitioning of Hg, Se, As, B and Pb for various streams in the Koppers–Totzek and Lurgi processes (Anderson, Hill and Fleming, 1979)*

Element	Hg		Se		As		B		Pb	
Concentration in coal (ppm)	0.2		2.08		14		10.2		35	
Process	Koppers–Totzek	Lurgi	Koppers–Totzek	Lurgi	Koppers–Totzek	Lurgi	Koppers–Totzek	Lurgi	Koppers–Totzek	Lurgi
Input streams (kg day^{-1})										
Coal feed	3.672	3.672	38.3	38.3	257.96	257.96	188.3	188.3	641.2	641.2
Output streams (kg day^{-1})										
Discharge ash	neg.	neg.	neg.	19.1	neg.	128.98	neg.	94.1	neg.	641.2
Solids from waste-water treatment	neg.	neg.	neg.	neg.	257.96	100.06	188.3	94.1	641.2	0.005
Product sulphur	0.0007	3.635	38.0	19.1	neg.	28.73	neg.	neg.	neg.	neg.
Sulphur recovery tail gas	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.
CO_2 vent gas (I)	0.0015	0.0092	<0.4	<0.2	neg.	0.275	neg.	neg.	neg.	neg.
Methanation quench	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.
CO_2 vent gas (II)	0.0015	0.0173	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.
Glycol cooler recovery	3.394	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.
Product SNG	0.27	0.0097	neg.	neg.	neg.	neg.	neg.	neg.	neg.	neg.

neg. = negligible.

utility boiler and an investigation into trace metal emissions resulting from char combustion carried out.

Although the condensate, formed after cooling the raw synthesis gas, contains low concentrations of most trace elements, some enrichment of As, B, Hg, Sb, Pb and Se on fine particles is observed (Craun and Massey, 1979). Process waters, however, also contain significant amounts of As, Cd, Hg, Pb, Se and Sb, and particularly high concentrations of Se were found in the condensate process water and tars. Mercury was not determined, but its behaviour would be expected to resemble Se. In addition, relatively high levels of B and F were present in a condensate sample examined by Forney *et al.* (1974).

The Koppers–Totzek gasifier. Information on this process is based on a semi-commercial plant with capacity for 3×10^7 g coal hr^{-1} (Oldham and Wetherold, 1977) and the theoretical calculations by Anderson, Hill and Fleming (1979) which computes volatility-condensation behaviour of volatile elements from thermodynamic properties.

The Koppers–Totzek process is regarded as relatively "clean" compared with other conversion techniques (Oldham and Wetherold, 1977). This is certainly borne out by the comparative data that have been calculated (Anderson, Hill and Fleming, 1979) and presented in Table 8.10. The higher temperature employed in the Koppers–Totzek gasifier will presumably result in a more complete vaporization of moderately volatile trace elements from the coal. For example, Pb shows a much higher volatility in the Koppers–Totzek process than in the Lurgi. The form and distribution of certain elements in raw and quenched gasifier product gas is shown in Table 8.11.

The Lurgi gasifier. No information regarding the trace element content of effluent streams from Lurgi gasifiers operating on a commercial scale was available until data on the Kosovo plant in Yugoslavia were published (Bombaugh *et al.*, 1980). Calculations indicated that the important streams would be the gasifier ash, the process condensate and the ash quench water (Table 8.11). In view of the operating conditions employed, it was suggested (Anderson, Hill and Fleming, 1979) that there would be a less efficient vaporization of trace elements compared to the Koppers–Totzek process, resulting in greater retention of these elements in the gasifier slag or ash (Table 8.11). The recent study (Bombaugh *et al.*, 1980) of the Lurgi Kosovo Kombinant plant in Yugoslavia provides a considerable amount of data on stream characterization of a commercial plant in normal operation; this is presented in Chapter 10 (Table 10.7).

Common auxiliary processes. Synthetic gas is subjected to shifting in all gasification processes, to produce the appropriate ratio of CO to H_2, prior to methanation. This is achieved in a catalytic reactor, employing a Cr-promoted

TABLE 8.11. *Form and distribution of selected trace elements (g-mol g-mol^{-1}) in raw and quenched gasifier product gas in the Koppers–Totzek and Lurgi processes (Anderson, Hill and Fleming, 1979)*

	Lurgi (20 atm operating pressure)	Koppers–Totzek (2 atm operating pressure)
Raw gasifier product gas		
AsH_3	1.71×10^{-7}	2.47×10^{-15}
As_4	1.56×10^{-7}	9.56×10^{-12}
As_2	3.20×10^{-9}	1.20×10^{-6}
H_2Se	1.36×10^{-7}	3.68×10^{-7}
$B(OH)_3$	4.88×10^{-5}	7.29×10^{-5}
PbS	8.13×10^{-14}	7.62×10^{-7}
$PbCl_2$	1.11×10^{-11}	1.30×10^{-12}
PbO	2.81×10^{-25}	2.31×10^{-9}
Pb	6.94×10^{-16}	1.67×10^{-6}
Hg	8.55×10^{-9}	1.28×10^{-8}
Quenched gasifier product gas		
AsH_3	3.19×10^{-7}	2.68×10^{-15}
H_2Se	2.50×10^{-7}	4.00×10^{-7}
Hg	1.58×10^{-8}	1.38×10^{-8}

Fe catalyst. Desulphurization units convert S to its elemental form. This process also employs catalysts, including those used in the Stretford–Beavon technique (Co–Mo and V) and possibly the Giamarco–Vetrocoke process (As). The methanation of synthetic gas is also carried out in all gasification processes, by use of a Ni catalyst. It is not known whether significant amounts of those potentially toxic elements employed in the above catalytic processes become entrained in the gas stream. Nickel carbonyl ($Ni(CO)_4$), a carcinogenic compound, forms in methanizer gas (Ghassemi, Crawford and Quinlivan, 1979). During continuous processing, temperatures well above those at which Ni carbonyl forms ($> 275°C$) are employed, but during start-up and shut-down, when lower temperatures are experienced, some formation of Ni carbonyl may occur. All traces would need to be removed eventually from the SNG delivered to the consumer, as upon decomposition in burners problems would arise resulting in failure of the burners. A detailed analysis of low-Btu product gas from a Wellman–Galusha gasifier (Thomas, Trede and Page, 1979) revealed significant amounts of $Ni(CO)_4$ and iron carbonyl ($Fe(CO)_5$). This is obviously an area of potential environmental concern, as well as technical interest from the point of view of burner efficiency.

2.3.2. *Liquefaction*

As indicated in Chapter 6, the worldwide economic recession has resulted in reduced investment and less research interest in synthetic fuels from coal. The Synthetic Fuels Corporation (SFC) in the USA, set up in 1980 to develop synthetic alternatives to imported oil, has been under adverse pressure from Congress with threats of ending funding altogether. Liquefaction processes that were approaching viability were the H-coal liquefaction project of the Hydrocarbon Research Company, Trenton, New Jersey, USA and the Bergius-Pier process pioneered by Ruhrkohle AG and Veba AG in the Federal Republic of Germany. These, and the donor solvent processes utilize catalysts in the hydrogenation of a slurry (mixed raw product and coal). The solvent-refined coal process (SRC I and SRC II) did not require catalysts, but operated well only with high ash bituminous coals. The synthoil liquefaction process, also discontinued, was the process employing a catalyst for which most data was published on stream composition (Yarovsky and Akhtar, 1974; Schultz *et al.*, 1977, 1978; Lett *et al.*, 1977) together with data for the centrifuged product. For this reason data for the Synthoil process are given here.

The Synthoil liquefaction process. In the Synthoil process a slurry of finely ground coal and oil is supplied to a reactor that contains a fixed-bed catalyst. Turbulent conditions in the reactor ensure contact between the catalyst (cobalt molybdate), coal plus the coal-derived liquid (or carrier) and the hydrogen that is fed in. Hydrogenation and desulphurization reactions occur during relatively short residence times in the reactor (a few minutes only).

The concentration of six trace elements in all the streams of the Synthoil process are given in Table 8.12. There is also some evidence that As may be present at an elevated concentration in the final oil product (Oldham and Wetherold, 1977).

The extent of trace element contamination of the Synthoil product oil will not only be relevant to its commercial usage, but will also be of importance in

TABLE 8.12. *Trace element analysis of all streams of the Synthoil process in* $\mu g\ g^{-1}$ *(Schultz et al., 1977)*

	Element					
Stream	Cu	Cr	Mn	Ni	Cd	Pb
Feed coal	10	21	57	9.3	0.32	4.6
Recycle coal	4.7	7.7	12	7.4	0.083	1.4
Feed paste	6.6	13	29	9.1	0.17	2.4
Scrubber influent	0.037	0.10	0.022	0.012	<0.002	0.025
Gross liquid product	6.7	15	31	10	0.19	3.0
Centrifuged liquid product	2.7	7.6	11	6.6	0.077	1.1
Centrifuge residue	45	84	180	54	1.0	18
Scrubber effluent						
Aqueous	0.15	0.28	0.019	0.029	0.0006	0.18
Organic	0.12	0.17	<0.004	<0.004	<0.014	<0.5
Vapour knock-out traps						
Aqueous	20.1	0.20	0.25	0.45	0.03	1.2
Organic	15.9	0.20	0.30	0.90	0.04	1.2

the liquefaction process itself, as the oil is intended to be used to provide heat for the plant. Presumably, particulate collection devices will be employed to reduce emissions from the combustion of this oil. If estimates of the trace element content are representative then the collected fly ash may contain elevated concentrations of several volatile elements.

In the Synthoil coal conversion process the catalyst is employed in the reactor vessel itself. It is thus possible that the oil products and the effluent streams will be contaminated with Co and Mo. Additionally, it is probable that replacement of this catalyst will have to be carried out frequently, as it will be in direct contact with large amounts of tars and oils in the reactor vessel. This may bring about disposal problems if the spent catalyst is not reclaimed.

3. Environmental Effects of Coal Utilization

3.1. Atmospheric Emissions of Trace Elements

3.1.1. Fallout in the vicinity of power plants

In recent years several field investigations have assessed the magnitude of atmospheric trace element deposition from power plants. Klein and Russell (1973) claimed that soils around a Michigan power plant were enriched in Cd, Co, Cr, Cu, Hg and Ni, with concentration trends reflected in the prevailing wind pattern. This contamination apparently extended 21 km from the plant and covered 113 km^2. However, despite the potential significance of these findings, Klein and Russell (1973) did not subject their data to statistical analysis. However, in a careful statistical analysis of deposition samples before and after a 37 % reduction in power generation by two coal-fired units and replacement by an oil-fired unit (Mastradone *et al.*, 1982), significant reductions were shown to occur in soluble, insoluble and total deposition in Pb. Additionally, a study on the sediments of Lake Michigan (Goldberg *et al.*, 1981) compared the excess of elemental concentration in recent sediments with the concentration in outlet fly ash concentrations and concluded that there was good agreement for Al, Fe, Pb, Cr, Cd, Co, Zn and V, although not all these elements may originate from the coal as corrosion from boiler tubes needs to be taken into account.

Surface soils from industrial areas in Poland reportedly contained increased U and Th concentrations compared to rural regions; this difference was ascribed to the presence of power plants at the industrial sites (Jaworowski and Grzybowska, 1977). Similarly, Martin *et al.* (1971) found elevated air concentrations of Ra, Th and U 6 km downwind of a coal power plant. Another study in the USA (Styron, 1979) could, however, detect no increased radionuclide content in the vicinity of a power plant, possibly because of the use of an efficient particulate collection device in this facility.

An extensive ORNL survey, conducted around the Allen Steam Plant in Memphis, USA (Bolton *et al.*, 1973; Lyon, 1977), detected no impact of plant emissions on trace element levels in the surrounding environment. However, efforts to distinguish atmospheric deposition were hampered by the naturally elevated trace element content of soils in the Memphis area. Background soil concentrations of many trace elements were actually greater than those from the "enriched zone" of the Michigan survey (Klein and Russell, 1973). Additionally, U and Th levels in Memphis soils were four times greater than the values reported from industrial regions in Poland (Jaworowski and Grzybowska, 1977). Two elements of potential significance, As and Se, were unfortunately not examined fully in the Allen Plant study.

Efforts to detect a concentration trend in Hg with distance from the Allen Plant involved the analysis of moss samples but, in contrast to the Michigan study, were unsuccessful. Similarly, Crockett and Kinnison (1977) could find no accumulation of Hg in soils around the Four Corners plant, one of the largest power facilities in the USA. The authors concluded that although emitted Hg may be initially deposited in the vicinity of the plant, its volatile nature leads to rapid vaporization. For this reason, they suggested the significance of Hg emissions should be evaluated on a regional rather than local basis. Aquatic systems adjacent to coal-fired power plants have also been examined for atmospheric Hg deposition (Anderson and Smith, 1977; Roffman, Kary and Hudgins, 1977). Analysis of sediments, water, macrophytes, fish and wildfowl revealed no significant elevation of Hg in these samples. All fish examined by Anderson and Smith (1977), for example, contained less than $0.5\,\mu g\,g^{-1}$ (wet weight), the FDA tolerance limit for consumption.

Serious environmental contamination from power plants is apparently confined to those facilities burning coal with unusually high trace element concentrations. Plants and soils downwind of a power plant in Nevada burning a high As coal ($> 500\,\mu g\,g^{-1}$) were markedly enriched in As (Berry and Wallace, 1974). The authors stressed the hazard of As transfer to humans from the milk of those cows feeding in the area. O'Toole, Wessels and Malaby (1981) devised a method for distinguishing between elemental enrichment of natural vegetation due to physiological processes, soil and power plant plume aerosols. They concluded that stack emissions were the principal source of vegetation enrichment of As in downwind exposed sites. The high As coal burned at a Czechoslovakian plant resulted not only in the contamination of adjacent pasture, but also the poisoning of livestock feeding in the area (Kuhnert, 1973).

The natural variability of trace element levels in soils requires, according to Wangen and Williams (1978), a 50% concentration increase to allow detection of any soil enrichment around power plants. Wangen and Williams (1978) further calculated that such increases would occur only when elemental fly ash concentrations were 100 times greater than local soils. In a subsequent study (Wangen and Turner, 1980) values approaching this were obtained from B, F and Se and it was suggested that these elements were prime candidates for use as indicators of coal-fired power plant contamination.

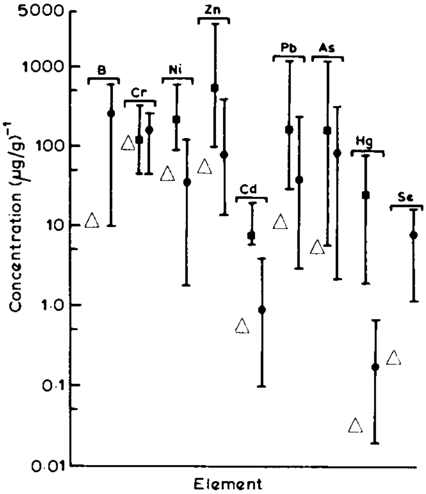


FIG. 8.4. Trace element concentrations (μg g^{-1}) in soil and in fly ashes from different regions of the USA.: △, world average soil (Vinogradov, 1959; Bowen, 1966); ■, mean and range of 11 fly ash samples (Theis and Wirth, 1977); ●, mean and range of 23 fly ash samples (Furr *et al.*, 1977).

In Fig. 8.4, trace element levels in world average soil (Vinogradov, 1959; Bowen, 1966) are shown in relation to the range of values reported for fly ash. The ranges quoted were for fly ash either collected by electrostatic precipitator or ash which has passed through the precipitator, but still sampled within the stack. Fly ash concentrations in the upper range for As, Cd, Pb, Se and Zn are actually greater than soil by a factor of 100 or more. Presumably, the fly ash escaping from such plants would be further enriched in these elements. The degree of enrichment in such ashes would thus appear to be sufficient in theory to produce an observable impact in the vicinity of the power plant involved. However, if efficient precipitators or other means of particle attenuation are employed, then larger-sized particles are removed from the flue gases. If only very fine fly ash particles escape from the stack they will tend to be dispersed widely, although agglomeration of fine particles could increase effective particle size and density and result in increased deposition near the power station.

In conclusion, it is apparent that the impact on soils and biota of trace elements from power plants consuming coal of average trace element content is generally not detectable in relation to the natural elemental levels of these materials. Another factor responsible for the general absence of any detectable deposition in the vicinity of power plants is the dispersion of emissions over a wide area by the tall stacks employed at such facilities. The small particle association of several volatile elements would further enhance long-range dispersal, as would the revaporization behaviour of Hg.

The source of enriched elements in urban aerosols. Several studies (Rahn, 1971, 1976; Gordon, Zoller and Gladney, 1973; Heindryckx, 1976) on the chemical composition of particles in urban aerosols revealed that lithophile elements, such as Fe, Mn, Sc and Th, have EFs close to 1, and are apparently derived from crustal weathering. Other, more volatile, elements, including As, Br, Cd, Cu, Hg, I, Ni, Pb, Sb, Se and Zn, are more enriched in urban particulates compared to crustal material. This finding was viewed with some concern, as many elements which showed the larger enrichments are also potentially toxic. As discussed earlier, many volatile elements have large EFs on suspended fly ash particles. In the case of most elements, however, the EFs for urban aerosols are even higher (Fig. 8.5), suggesting that coal combustion is not responsible for their presence in urban particulates. Both As and Se are notable exceptions as these have similar EF values in suspended fly ash particles and urban aerosols (Gladney *et al.*, 1974; Zoller *et al.*, 1974; Klein *et al.*, 1975). Direct sampling of a power plant plume revealed that the enrichment of As and Se persisted outside the power plant, while in the case of other elements, only the concentrations were enhanced compared to the background aerosol (Zoller *et al.*, 1974). Additionally, results of an attempt to identify air pollution sources by an elemental balance technique (Kowsalczyk, Choquetter and Gordon, 1978) also indicated that coal combustion was the major source of only As and Se in the Washington D.C. aerosol.

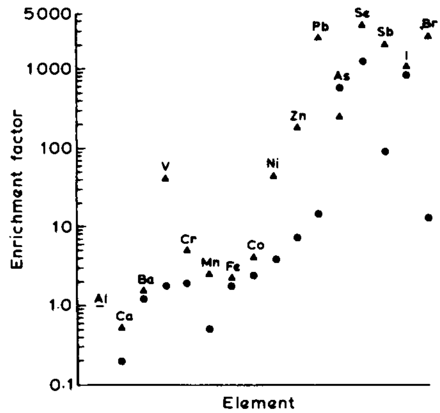


FIG. 8.5. Elemental enrichment factors relative to crustal abundance in-stack particles (●) at the Chalk Point plant and of ambient aerosols (▲) in the vicinity of the plant (Gladney et al., 1974).

Taken together, these findings suggest that beside As and Se the combustion of coal cannot account for the anomalous enrichment of trace elements in urban aerosols. However, examination of the enrichment pattern in coal (Fig. 8.6) reveals that those elements which display significant enrichment in coal are the same elements which are enriched in the urban aerosol. Moreover, the degree of elemental enrichment in coal is generally proportional to that in the aerosol, but is a factor of 10 lower. The additional enrichment caused by coal combustion is shown for Chalk Point power plant in Fig. 8.7 (Gladney *et al.*, 1974). For most elements, the combustion process contributes very little over and above the coal enrichment. Significant enhancement due to

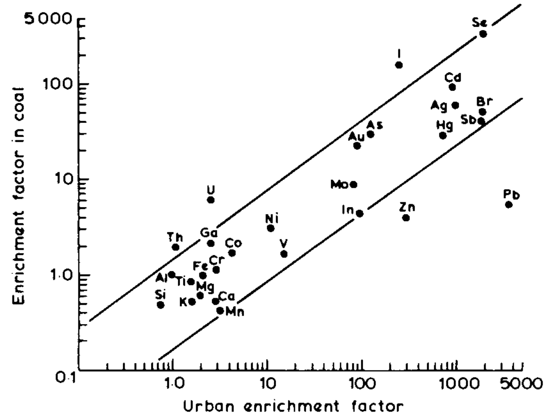


FIG. 8.6. Elemental enrichment factors in coal (with respect to crustal abundance) in relation to the corresponding enrichment factors of the urban aerosol (from Rahn, 1976).

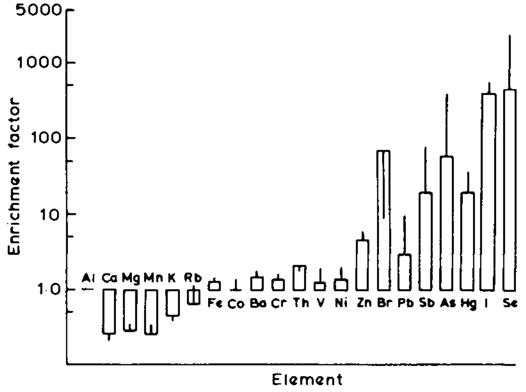


FIG. 8.7. Elemental enrichment factors with respect to crustal abundance at the Chalk Point power plant: □, coal; I, additional enrichment in fly ash attributable to combustion (Gladney *et al.*, 1974, after Rahn, 1971).

combustion itself only occurs for the more volatile elements, As, Hg, Pb, Sb and Se.

A possible reason for the close relationship between enrichment patterns in the urban atmosphere and coal is that coal combustion is actually responsible for the abundance of these elements in the aerosol. Certain elements will, of course, have specific sources; the large enrichment of Pb, for example, is probably associated with the use of leaded petrol. An alternative explanation involves the enrichment of the urban aerosol by a variety of high-temperature process. If this is the case, then the parallel enrichment relationship between coal and urban aerosols (Fig. 8.6) would require further explanation, involving an as yet unknown physicochemical link between the enrichment of trace elements during coal formation and enrichment during high-temperature processes.

If coal combustion is the major source of certain enriched elements in urban aerosols, then account must be taken of the relatively low EF reported for many elements on suspended fly ash particles. This apparent discrepancy may be a reflection of the absence of reliable data on elemental EFs on submicron particles, as discussed earlier. Additionally, the chemical composition of aged power plant plumes may be very different from that of in-stack particulates. The greater settling velocities of larger fly ash particles would be expected to lead to a fractionation with time, resulting in increased EFs for volatile elements, because of their small particle association. More information on the trace element composition and behaviour of power plant plumes with age is required before their contribution to the urban trace element load can be assessed.

Emissions in relation to other sources. The significance of atmospheric trace element emissions from coal combustion may be evaluated on a global scale by comparison with both the total natural and anthropogenic fluxes of the element in question. Relevant data for elements of toxic significance are shown in Table 8.13. The values reported have been obtained either directly from the literature or indirectly where information is lacking or considered

TABLE 8.13. *Estimated global trace element emissions from coal combustion and other anthropogenic sources in relation to natural sources*

	Global annual emission ($\times 10^9$ g)					
Source	As[a]	Cd[b]	Hg[c]	Ni[b]	Pb[b]	Zn[b]
Total natural	7.8	0.83	10	26	24.5	43.5
Coal combustion	5.8	0.06	0.54	0.66	14	15
Total anthropogenic	29	7.3	2.5	47	499	314

[a] Anthropogenic and natural source strengths from Walsh, Duce and Fasching (1979).
[b] All data from Nriagu (1979).
[c] Anthropogenic and natural source strengths from Miller, D. R. (unpublished).

inaccurate. Natural sources of importance include volcanoes, wind-blown dusts and vegetable exudates.

Anthropogenic emissions of Pb, Cd, Ni and Zn are all far in excess of natural rates, but in each case coal combustion accounts for only a small proportion of the anthropogenic total. Nevertheless, the atmospheric emission of Pb from coal combustion occurs at over 50% of the rate of introduction by natural sources, its significance being masked by the consumption of leaded petrol (Nriagu, 1979).

Anthropogenic processes also dominate the introduction of As into the atmosphere, with the largest man-made contribution resulting from Cu production (Walsh, Duce and Fasching, 1979). In this example, the emission from coal combustion is also of importance, being comparable to the total natural rate. The figure estimated for coal combustion is, however, an order to magnitude greater than that calculated by Walsh, Duce and Fasching (1979) in a recent survey of atmospheric As emissions. The disparity arises because Walsh, Duce and Fasching (1979) employ an emission factor taken from the Allen Steam Plant study (Klein *et al.*, 1975) which is felt to be erroneously low. This is because no consideration is made of the large imbalance for As flow through the Allen Plant. In the present study, the atmospheric emission of As from coal combustion was obtained from the mean of two estimates. In the first, the average As content of coal was assumed to be 15 $\mu g\,g^{-1}$ (Table 8.7) and the annual global coal consumption 3×10^9 t (Quenton, 1980). An emission factor of 0.15 $g\,t^{-1}$ was employed, estimated from the wide range of reported values for the element (Natusch, Wallace and Evans, 1974; Schwitzgebhel *et al.*, 1975; Roffman, Kary and Hudgins, 1977) giving an annual As emission of 6.8×10^3 t. The other method utilized the finding that the atmospheric release from nearly 700 power plants averaged 8% of the total ash in the coal burned (McBride *et al.*, 1978). Using an ash content of 13% in coal and an average As fly ash concentration of 150 $\mu g\,g^{-1}$ (Theis and Wirth, 1977) gives an annual emission of 4.7×10^3 t.

The total annual Hg emission from coal burning was estimated from an average Hg coal content of 0.2 $\mu g\,g^{-1}$ (Table 8.7) and an emission factor of 0.9 (Billings and Matson, 1972; Kalb, 1975) to give 0.54×10^3 t. This figure is six times smaller than previous estimates by Joensuu (1971) or Heindryckx *et al.* (1974) who assumed an unrealistically high average Hg coal content and an emission factor of 1. However, the contribution from coal combustion is still significant, being over 20% of the total anthropogenic emission of the metal.

A number of comparisons of radioactive emissions from coal-fired power plants and nuclear power plants have been made (Eisenbud and Petrow, 1964; Hull, 1971; Martin *et al.*, 1971; Jordan and Schikarski, 1973; Jaworowski *et al.*, 1974; Martin, 1974; Lave, 1975; Illyin, Kniznikov and Barkhudov, 1977; Kniznikov and Barkhudov, 1977; McBride *et al.*, 1978). Some studies have emphasized the relatively greater biological significance of radionuclides released from coal combustion. Health impacts from coal-derived radionuclides will depend on a number of variables including the activity concentrations of radionuclides in the combusted coal (^{40}K, ^{238}U decay series and ^{232}Th decay series). Some of the worst prognoses have been based on the use of coal with a very high ^{238}U content but Beck, Gogolak and Miller (1979) found that the activity concentration in coal samples varied over two orders of magnitude. The fly ash control measures adopted are also of importance as smaller, escaping fly ash particles may be considerably enriched with some decay series element. UNGA (1980) quotes fly ash atmospheric releases of between 1 and 30%.

Human health hazards. Particulate emissions from coal combustion may be inhaled by humans, and with stacks as tall as 200 m deposition may occur within a circle of 500 km radius (UNGA, 1980). As briefly discussed earlier, particle size is of major importance in determining the toxic significance of such emissions. This is because inhaled particles are deposited in different regions of the respiratory system depending on their effective aerodynamic particle size, as shown in Fig. 8.8. Larger particles are preferentially deposited

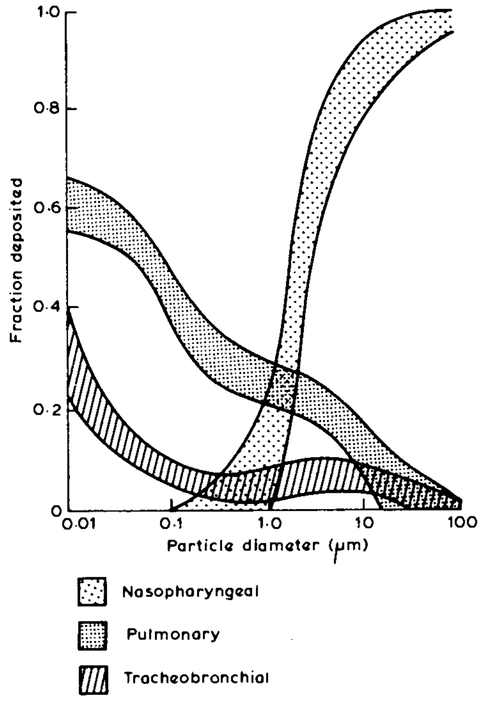


FIG. 8.8. Efficiency of particle deposition in the three respiratory tract compartments (nasopharyngeal, pulmonary and tracheobronchial) as a function of particle size (Natusch, Wallace and Evans, 1974).

in the nasopharyngeal and tracheobronchial regions and are generally removed after a short time by cilial action and consequently swallowed. Elemental uptake from such particles will thus take place in the gastrointestinal system where absorption rates are much lower than in the lung (Norberg, 1976). Smaller particles are deposited more efficiently in the pulmonary region of the lung (Natusch, Wallace and Evans, 1974).

At present, emissions from coal combustion have not been adequately characterized (Section 2.1.1), but strong increases of several trace elements with decreasing particle size have been reported (Natusch, Wallace and Evans, 1974; Campbell *et al.*, 1978; Ondov, Ragaini and Biermann, 1978; Smith, 1980). For this reason it is again emphasized that the precise particle size distribution of toxic elements in suspended fly ash is required before the significance of such emissions can be evaluated. Smith (1980) points out that volatilized elements which condense upon fly ash (As, Sb, Pb, Cd, V, Mo, Zn, Ga, Cs and U) are often emitted into the amosphere by a factor of up to 10 or more than elements not volatilized and thus have a higher probability of pulmonary deposition upon respiration. Elements which are totally volatilized and do not condense on particulate matter (halogas, Hg, some portions of the Se, B and perhaps P and Sb) will be emitted into the atmosphere in 100 times or more greater abundance than those not volatilizing. They may have a probability of × 1000 for pulmonary deposition upon respiration compared to elements not volatilized during combustion. In addition, the possible surface predominance of volatile toxic elements (Natusch, Wallace and Evans, 1974) will result in intimate contact with the alveolar membrane and further enhance the toxicity of small particles. Finally, the chemical form of deposited elements in the lung requires evaluation as this will be an important factor in determining the solubility and hence uptake into the pulmonary bloodstream.

One aspect of human health concern is the possible carcinogenic properties of coal fly ash. Evidence in favour of this view derives from the reported mutagenicity of respirable fly ash filtrates to bacteria (Chrisp, Fisher and Lammert, 1978). Mumford and Lewtas (1982) used the Ames test to investigate mutagenicity of whole particles of conventional-combustion fly ash and fly ash originating from a pressurized fluidized bed combustion miniplant. The former showed no mutagenicity, whereas FBC fly ash proved mutagenic in tester strains TA98 and TA1538. Extracts of fluidized bed combustion fly ash were found to contain direct-acting frameshift mutagens. Much lower mutagenic activity was found in extracts of conventional-combustion fly ash. Some of the mutagenic components present in very fine fly ash were inorganic in nature (Fisher, Chrisp and Raabe, 1979). In addition, Chrisp, Fisher and Lammert (1978) noted that equine serum was more efficient in extracting the mutagens than saline, suggesting that such substances should be similarly soluble in alveolar lung fluid. The tests carried out by Mumford and Lewtas (1982) on conventional-combustion and fluidized bed combustion fly ash utilized the rabbit alveolar macrophage-system. This was more sensitive for measuring cytotoxicity than the Chinese hamster ovary system.

The three elements As, Cr and Ni are present in high concentrations in fly ash particles and all possess carcinogenic forms of importance as As(III), Cr(VI) and Ni carbonyl. There is no information available on the abundance of the carcinogenic forms of As and Cr in fly ash, but Van Hook (1978) considers both are present. It is not thought that the thermolabile Ni carbonyl will be present in fly ash. Identification of the possible carcinogenic substances in fine fly ash from coal combustion, and char and other residues from coal conversion processes, is required.

Arsenic contamination from the Czech power plant burning high As coal, described earlier (Section 1.2.2), extended to humans in the area. Bencko and Symon (1977) reported that children residing up to 5 km from the plant contained elevated As levels in hair, urine and blood. Furthermore, hearing changes were reported in a group of boys living near the coal-fired power plant. Bencko and Symon (1977) tentatively assigned this to an As-induced effect, as there are known cases of auditory damage, including total deafness, caused by As poisoning. It was not clear whether the elevated As in these children was predominantly dietary in origin or resulted from respiratory uptake, as As air concentrations were extremely high near the plant, with a maximum of 69 $\mu g\,m^{-3}$ (Bencko *et al.*, 1976).

The radiological impacts of coal combustion. This has been assessed on a number of occasions (UNSCEAR, 1977; UNGA, 1979, 1980). Individuals will be exposed from several sources: suspensions of radioactive particles in the atmosphere, particles inhaled or ingested. Only the inhalation pathway was thought to be significant as a human health impact (Beck, Gogolak and Miller, 1979), but more recent work is resulting in a revision of this view (see Section 2.2, Chapter 7). Estimates of collective dose commitments (UNGA, 1980) suggest that coal-fired power plant emissions are either equal to or less than those of a nuclear power plant in normal operation and that for both the dose levels are extremely low, constituting only a negligible percentage of the dose from natural background radiation. A recent estimate of atmospheric radionuclide discharge from coal combustion is 56×10^{-3} GBq per MW(e)yr. This results in 3.7×10^{-3} man-Sv per MW(e)yr from inhalation, internal radiation and external irradiation due to the activity deposited, compared to an annual dose equivalent of 0.8×10^{6} man-Sv for the population of Europe and North America from natural irradiation.

One or two points need to be taken into consideration, however. The calculations for coal combustion are based on 1977 world coal production figures (3×10^{9} t). The lowest class of solubility in lungs is employed; bacterial mutagenicity studies (Chrisp, Fisher and Lammert, 1978) and aqueous extraction experiments (Theis and Wirth, 1977) indicate that at least some inorganic constituents will be soluble in the lung. Additionally the utilization of low-grade coals with elevated (20 $\mu g\, g^{-1}$) U content from parts of the western USA may pose significant health hazards on a local scale, not anticipated from the use of average activity concentrations.

3.2. Solid Wastes from Coal Utilization

3.2.1. Coal combustion

The solid wastes resulting from the combustion of coal in power plants are fly ash, bottom ash and slag and, if SO_2 scrubbers are employed, scrubber sludge. Fly ash is the most important waste material in modern pulverized coal power plants. Various forms of fly ash emission control devices are in use including mechanical or cyclone collectors, electrostatic precipitators, flue gas scrubbers and fabric filters. Ash retention may vary between 70 and 99% (UNGA, 1980). In the USA the EPA clean air standard to be eventually implemented corresponds to a release of about 1% of total ash. Problems of fly ash disposal primarily result from the enormous quantities collected; in the USA alone the figure was 60×10^6 t in 1976 (Scanlon and Duggan, 1979). Efforts have been made to employ fly ash for commercial purposes, but stockpiles are increasing with time. Commercial utilization of fly ash is apparently more successful in the UK than the USA, with about half the ash produced gainfully used in the UK (Brown, Ray and Ball, 1976), compared to 20% in the USA (Scanlon and Duggan, 1979). Disposal of fly ash is carried out by either wet storage in ash ponds or after removing water, in landfills. In both cases the disposed fly ash will be in close contact with adjacent soil and water systems.

Trace element content of ash. Trace element concentrations in fly ash vary widely from one sample to another, reflecting differences in the original coal composition as well as boiler-operating conditions. Reported values from two studies of fly ash from the USA are shown in Fig. 8.4. The investigation by Furr *et al.* (1977) is thought to provide representative values as levels were measured in 23 samples from 21 states in the USA.

Passage to terrestrial ecosystems. Several volatile elements in coal pose a potential hazard to the environment, but there is little evidence for any marked accumulation in soils around power plants. Similarly, several investigations have failed to detect significant enrichment of trace elements in vegetation or animals in the vicinity of power plants (Roffman, Kary and Hudgins, 1977; Strojan and Turner, 1978). The study of Klein and Russell (1973), on the other hand, reported that vegetation as well as soils near a Michigan power plant contained elevated levels of Cd, Ni and Zn. Two other studies (O'Toole, Wessels and Malaby, 1981; Wangen and Turner, 1980) have trace element enrichment in natural vegetation due to coal-burning power plants in the vicinity. As and B, F and Se concentrations in vegetation were shown to decrease with distance, downwind from the power plants.

The large variability in the trace element content of soils may be masking the input of small quantities of trace elements from coal combustion, not detectable by conventional bulk analysis. The significance of such inputs will be determined to a certain extent by the chemical state of the deposited elements. Greater solubility of an element in fly ash compared to soil will increase its biological availability and potential for bio-accumulation, but would also lead to greater mobility, resulting in rapid passage through the soil. Van Hook (1978) considers that microbial soil processes are particularly susceptible to small increments in trace element deposition, as micro-organisms are in intimate contact with trace elements, being found in the immediate vicinity of soil particles. In this context it is of interest to note that reduced litter decomposition rates have been associated with the atmospheric deposition of Cd, Pb and Zn from metal smelters (Tyler, 1976; Coughtrey *et al.*, 1979). Adverse effects on soil/litter processes may also result from the long-term deposition and accumulation of trace elements around power plants. Soils in which ambient levels of trace elements are already elevated are expected to be sensitive to such effects.

A possibility also exists that atmospheric deposition from coal combustion will ameliorate the deficiency of a particular essential trace element in the surrounding soil. Once again, it should be emphasized that the chemical state of the deposited element will determine its biological availability to vegetation. Factors affecting the element's chemical state, and therefore its solubility behaviour with the soil column, include the soil type and its pH, the amount of rainfall, the acidity of the rainfall and the oxidation-reduction conditions in the soil.

The accumulation of trace elements by terrestrial vegetation growing around ash disposal ponds or on fly ash landfills has been examined recently. Markedly elevated B levels were found in the leaves of several herbaceous species from near one disposal pond (Romney, Wallace and Alexander, 1977). The values encountered would have been high enough to cause toxic symptoms in sensitive species. Yellow sweet clover (*Melilotus officinalis*) found growing on landfilled fly ash contained higher concentrations of a range of trace elements compared to controls (Furr *et al.*, 1976), but only in the case of Se could those levels be considered elevated. In this respect it is of interest to note that small mammals trapped from this landfill site also contained greater tissue Se concentrations than controls—the levels present are not, however, considered toxic (Fleming, Gutenmann and Lisk, 1979).

The stabilization and restoration of fly ash disposal areas to reduce water and air erosion has involved the establishment of vegetation on the sites (Hodgson and Buckley, 1975). As part of a feasibility study, Scanlon and Duggan (1979) examined the elemental uptake by eight woody species planted on fly ash. After 2 years' growth, there was substantial foliar accumulation in all species of only B, Ni and Se. The foliage of plants grown on fly ash accumulated B to about twice that in plants grown on soil, although levels in the two substrates were similar. Additionally, the Se content of fly ash-grown samples was generally greater than the fly ash concentration itself, yet soil-grown plants contained lower levels than the substrate. These findings suggest that both B and Se display greater bio-availability to plants from fly ash compared to soil. Cherry and Guthrie (1979) confirmed the accumulation of Se in plants grown on fly ash and reported similar bioaccumulation of As, Cu, Hg and Zn. In the case of Ni, the elevated tissue levels of fly ash-grown plants simply reflected the higher fly ash concentrations of this metal compared to soil (Scanlon and Duggan, 1979). After 3 years' plant growth, the rates of element concentration in foliage to content in substrate decreased for both Se and B in the fly ash-grown plants. It would thus appear that there is a reduction in the available B and Se levels in fly ash with time, possibly due to weathering. Reduced availability of B with time has been reported previously for fly ash (Townsend and Gilham, 1975); several years' weathering of fly ash landfills may sufficiently reduce the B content to allow the growth of susceptible plant species.

The extent of trace element uptake by plants grown on fly ash-amended soil, or other substrates, has also been investigated, in order to assess the practical value of fly ash to ameliorate agricultural soil or its use as a high pH medium to ameliorate acid wastes like colliery spoil: a range of crops, grown on soil with 5 or 10% fly ash, were found by Furr *et al.* (1976) to contain enhanced concentrations of As, B, Cu, Hg, Ni, Sb and Se in their edible portions, compared to soil-grown controls. Jastrow *et al.* (1981) found enhanced levels of Co, Cu and Zn in the foliage of *Festuca arundinacea* (Kentucky 31) when grown on fly ash amended colliery spoil, compared to growth on topsoil or colliery spoil with additions of lime. In an acid and a calcareous soil treated with up to 8% fly ash by weight (Elseewi *et al.*, 1981), accumulation of B occurred in alfalfa (*Medicago sativa*), Bermuda grass (*Cynodon dactylon*) and white clover (*Trifolium repens*), with uptake being dependent on plant species, soil reaction and rate of fly ash application. Tissue concentrations of B were positively and linearly correlated with B from treated soils determined by saturation extracts. Elseewi, Straughan and Page (1980) suggested that there might be potential problems involved with the use of forage grown on fly ash treated soils in view of low Cu : Mo ratios in the foliage and associated indirect animal toxicity problems from excess Mo. A number of studies suggest that providing a diet to animals made up largely, or entirely, of vegetation grown on pure fly ash should be avoided. Furr *et al.* (1976, 1978) and Stoewsand, Gutenmann and Lisk (1978) found that Se was present in elevated levels in animals (guinea pig, quail, sheep and goat) fed in this way.

In a similar study (Furr *et al.*, 1977), cabbage grown on soil amended with fly ash from different regions of the USA contained concentrations of As, B and Se which were positively correlated with the respective fly ash value. Nevertheless, the levels found in cabbage were much lower than the respective values reported for woody species growing on pure fly ash (Scanlon and Duggan, 1979), even after taking account of the reduced total trace element content of the amended soils. It is possible that the high alkalinity of most fly ashes may actually be responsible for reducing the availability of trace elements from ash when mixed with soil. The finding of Phung *et al.* (1979), that increased fly ash additions to soil actually reduced the available levels of some trace elements, supports this suggestion.

Comparison with natural weathering of trace elements. In an attempt to assess the significance of trace element release from solid coal wastes in the USA, Klein *et al.* (1975) compared the quantities of elements discharged annually to ash disposal sites with data on natural weathering mobilization. Similar comparisons have also been carried out on a worldwide basis for Se (Andren, Klein and Talomi, 1975) as well as Cd and Hg (Van Hook, 1978). These studies reported that As, Cu, Ni, Pb and Zn all display mobilization rates from this source that are larger than 10 % of the natural weathering rate, while in the case of Cd, Se and U the values are equivalent to those for natural weathering. It is felt, however, that the validity of such comparisons is questionable, as many elements are efficiently contained within the ash disposal sites and show only limited migration into adjacent soil and water systems. In contrast, natural weathering rates reflect the mobilization of trace elements by the world's rivers on a global scale. Nevertheless, in spite of the low mobilization potential of most trace elements from ash disposal sites, future increases in coal utilization will place a greater strain on the sorbing capacity of adjacent soils and may result in greater contamination from such areas. Thus, the extent of mobilization of trace elements from ash disposal sites should be regularly monitored in order to prevent ground water contamination on a significant scale.

Mobilization of trace elements from ash ponds. The possible contamination of water and soils by the leaching of trace elements from fly ash disposal ponds has only recently received attention. Theis and his co-workers (Theis *et al.*, 1978; Theis and Richter, 1979) reported that although trace element concentrations in an ash pond were elevated, levels rapidly decrease in ground water and soil with distance from the pond. In accord with experimental extraction studies (Theis and Wirth, 1977), Theis *et al.* (1978) noted that the partitioning of trace elements between water and soil, as well as their attenuation around the ash pond, was most affected by the pH and the oxides of manganese and iron. Accumulation of metals in the adjacent soil was primarily due to a complex interaction between precipitation and absorption on to these metal oxides. It was pointed out by Theis *et al.* (1978) that unless these metal oxides were replenished, the absorptive capacity of the system was likely to be exhausted, resulting in a freer migration of trace elements from the ash pond. In view of its large sorptive capacity, manganese oxide additions to ash ponds may well be a useful control mechanism to reduce leaching of trace elements from such sites.

The pH of ash pond leachate also exerts an important influence upon trace element behaviour. The alkaline nature of most fly ashes (Townsend and Gilham, 1975) results in the precipitation of insoluble hydroxides of many toxic metals in the ash pond itself. Elements with anionic character, such as As, B, Cr, F, S, Mo and Se, are more soluble in the alkaline environment and significantly elevated concentrations of these elements have been reported in ash ponds (Theis *et al.*, 1978) and their effluent water (Dreeson *et al.*, 1977; Turner *et al.*, 1982). Future investigations of contamination from ash ponds into effluent waters should thus focus upon the behaviour of these elements. It should be noted, however, that attenuation of these elements in soil adjacent to the ash pond still occurs and they are probably absorbed on to positive iron hydroxide (Theis and Richter, 1979).

The direct run-off of fly ash leachate into adjacent streams has been found to result in the accumulation of several potentially toxic elements in both invertebrate and vertebrate fauna as well as aquatic vegetation (Gutenmann *et al.*, 1976; Cherry and Guthrie, 1977; Cherry *et al.*, 1979). Of particular interest was the elevated concentrations of Se and Cd in several organisms from the drainage systems receiving coal ash effluent, examined by Cherry *et al.* (1979). Decreased invertebrate densities adjacent to the ash basin were also reported, but this was ascribed to the increased turbidity, rather than any effect of toxic trace elements. However, in an aquatic bioassay study, Birge (1978) showed that simulated effluent from fly ash was particularly toxic to amphibians and fish. Moreover, LC_{50} values for several trace elements (As, Cd, Hg, Se and Zn) found in coal ash for these organisms were actually lower than the respective drainage water concentrations recorded by Cherry *et al.* (1979). In order to place the extent of this contamination in perspective, it should be noted that Cherry *et al.* (1979) found that ash discharge influence extended to only about 1 km from the disposal pond.

3.2.2. Coal conversion processes

The range of solid wastes derived from coal conversion processes not only include a bottom ash and a fly ash, analogous to the products of conventional coal combustion, but also a carbonaceous char or residue. Char from gasification may be combusted to provide energy, while liquefaction residues may undergo gasification to produce synthetic gas. Thus, a further series of solid inorganic wastes will be derived from such processes. Additionally, it is possible that char or residue will be produced in excess of demand, necessitating the disposal of these wastes. The intended combustion of synthetic liquid fuels in power plants will also produce significant quantities of ash. Such produce residues will consist of a bottom ash and, if collection devices are employed, a fly ash. Finally, spent catalysts from conversion processes which cannot be reclaimed will also require disposal.

The utilization of inorganic ashes from coal conversion, for such purposes as road construction, may occur on a commercial scale, but this is unlikely to solve the disposal problem of these wastes. Disposal is expected to take place in landfills or ash ponds adjacent to the conversion facility. It is probable that the spent catalysts and any unwanted char or residue will also be disposed of in this manner. In the case of inorganic ashes, it is expected that the mobilization and bio-accumulation of trace elements from such disposal areas will be similar to that found for fly ash derived from conventional coal combustion. Certain S compounds, absent from solid waste from combustion processes, may still be present in conversion process wastes. Attention should be paid also to the behaviour of trace elements in the fly ash produced by the Koppers–Totzek process. This is because the operating conditions of this process indicate that concentrations of several potentially toxic elements may actually be greater than found for coal combustion. The mobilization potential of trace elements present in the waste organic residues has not been investigated. It is thought unlikely that this will differ substantially from the limited migration of trace elements observed in the vicinity of fly ash disposal sites (Theis *et al.*, 1978). Contamination of water and soils by the Co, Cr, Fe, Mo and V present in spent catalysts is expected to occur adjacent to the disposal site, but the likely extent of such contamination is unknown and requires investigation.

4. Conclusions

4.1. Summary of Evidence from Existing Combustion Processes

The environmental and human health implications of trace element release from coal utilization are of current concern, notably because the quantities of coal combusted are at present very large and are planned to further increase in the near future (US National Committee for Geochemistry, 1980).

The results of investigations into the combustion behaviour of trace elements in power plants can be explained by the physicochemical properties of the elements and their compounds, and by the operating conditions within the power plant. Such studies have enabled the classification of trace elements according to the manner in which they are partitioned between the various exit streams and by their particle size distribution in suspended fly ash. These findings have been rationalized in terms of a volatilization-condensation mechanism, involving the vaporization of volatile elements during combustion and their subsequent condensation on fly ash particles. This model predicts that there will be dramatic increases in the concentration of volatilized elements on submicron particles and these elements will be preferentially enriched on the surface of such particles. These points are of significance as particles of this size are retained least efficiently by collection devices, have the longest atmospheric residence times and are deposited most efficiently in the pulmonary region of the respiratory system. Several recent investigations have, however, reported smaller enrichments of these elements than predicted by this model, leading to the development of a more complex mechanism to account for the observed behaviour of trace elements in power plants. Whatever the precise mechanism, it is essential that the extent of enrichment of several potentially toxic elements of submicron particles be clarified. More information on the surface predominance of volatile elements and their chemical form in fly ash is also required, as both factors are important in determining the toxicity and solubility of these elements. Such studies should extend to emissions from domestic heating appliances, as evidence from one investigation indicates that certain elements may also be highly enriched in the suspended particles from these units.

The full characterization of emissions from coal-fired power plants, together with an understanding of the effects of ageing on the composition of such plumes, will allow an assessment of the importance of coal combustion in contributing to the large enrichment of trace elements in urban aerosols. In particular, the significance of the high enrichment factors reported for As and Se in power plant emissions requires substantiation.

Evaluation of the potential human health hazards from coal combustion also requires the accurate characterization of trace element emissions. In the context of human health, the possible carcinogenic properties of fly ash also require further investigation, with special attention paid to the presence of the carcinogenic forms of As and Cr in respirable-sized fly ash particles.

Only in one case of As contamination has serious environmental pollution been directly linked to trace element emissions from coal combustion. The power plant responsible consumed a low-grade coal with elevated As content, and children in the vicinity of the facility exhibited symptoms associated with As toxicity. It is not known whether other power plants in the region also consume coal with high As concentrations or if the use of improved particulate control technology would significantly reduce As emissions. It is suggested that the screening of coals suspected to contain high As concentrations be carried out, and the use of such coals be avoided, especially in facilities without adequate control devices.

Estimates of the radiological impact to humans from coal combustion indicate that this source represents a very small fraction of the collective dose. However, the utilization of uraniferous coals from the western USA, or from other parts of the world, would be expected to substantially increase the radiological exposure to humans residing in the vicinity of such plants.

The importance of coal combustion as a source of atmospheric trace elements has been assessed on a global scale. Emissions of As from coal combustion apparently rival the total natural emission rate for this element.

Although the other elements considered in this report are released in significant amounts, the contribution to the total anthropogenic flux appears to be relatively small, with the exception of Hg. Earlier estimates of the global releases of Hg from coal combustion are thought to be too large, as unrealistically high Hg values were assumed. Such errors highlight the necessity for both accurate and representative trace element values in coal, to enable the significance of this source of contamination to be determined.

The mobilization of trace elements from ash disposal areas has only recently received attention. The limited data available indicate there is rapid attenuation in soils and ground water close to the disposal site. Contamination of adjacent water courses by direct run-off from disposal ponds results in decreased water quality and the accumulation of trace elements toxic to aquatic organisms. Present information suggests that such contamination is also restricted to the close vicinity of the disposal ponds. However, the expected increase in coal utilization will result in greater quantities of fly ash requiring disposal, which will place a greater strain on the absorptive capacities of soils. For this reason, control techniques, such as the addition of manganese oxide, may be needed to reduce the leaching of trace elements from ash disposal sites.

In conclusion, it is clear that at present there is insufficient information to allow an adequate evaluation of the environmental and health impacts of trace element release from current and projected coal utilization. From the evidence available it would appear that coal combustion will make a contribution, along with other anthropogenic sources, to the trace element load in the atmosphere. The significance of this contribution will be dependent upon the ambient air quality, the proximity of other anthropogenic sources and the concentrations of volatile trace elements in the coal combusted. In the case of solid wastes, the limited information available indicates that trace elements are efficiently retained within the fly ash disposal sites, but long-term studies are needed to determine leaching rates upon weathering. For this reason, it is felt unrealistic to compare the quantities of trace elements discharged in disposal sites with the natural weathering rates of these elements. However, the mobilization and bioaccumulation of trace elements in fly ash used to ameliorate agricultural soils requires further investigation.

4.2. Predicting Likely Effects of New Conversion Technologies

The assessment of trace element contamination from future coal conversion technologies was based upon information from pre-commercial conversion units, recent published material on commercial gasification and liquefaction plants, and extrapolation from studies of conventional coal-fired power facilities. In coal-fired power plants the operating conditions are known to be important in determining the partitioning of elements between the various exit streams. Unfortunately, the large differences in operating conditions between the various processes still being developed for coal conversion make predictions for future trace element impact difficult. Nevertheless, most of the conversion processes discussed in this section employ high pressures and lower temperatures than conventional coal-fired power plants. These conditions are expected to reduce the vaporization of volatile elements, resulting in a less marked partitioning between fly ash and bottom ash and a smaller enrichment of such elements on submicron particles. Thus, atmospheric emissions from coal conversion facilities are likely to exert less impact on the loading of ambient aerosols than coal combustion.

Another important factor affecting the environmental significance of coal conversion is the trace element content of feed coal. Preference may be given for the selection of coals which exhibit high hydrogenation rates. This may result in the use of coals which contain a high mineral content and elevated concentrations of certain trace elements.

From the limited information available it would appear that gasifier char retains a large proportion of the trace elements present in the feed coal. The proposed combustion of char in utility boilers may thus result in the emission of fly ash enriched in volatile trace elements.

The use of catalysts in coal conversion processes adds another dimension to the environmental implications of this technology which requires assessment. Spent catalysts which are not reclaimed will pose a disposal problem, the importance depending on the amounts of catalysts used. The metals present in these catalysts may contaminate the conversion products as well as the effluent streams.

The trace elements retained in the conversion products will also be mobilized when those products are combusted. From the limited data it would appear that liquefaction products contain trace element concentrations in the range found for coal, or with significant reductions in certain elements. The trace element composition of emissions from the burning of these oils may be no greater than that found for coal combustion. Gasification products undergo cleaning procedures which appear to remove most trace elements. However, some available evidence indicates that Hg and F may still be present in the final gas product (Oldham and Wetherold, 1977). If this is the case, then further cleaning may be required in order to remove these toxic elements.

The large range of inorganic wastes from coal conversion processes are derived from the coal conversion process itself, from the further combustion of char and gasification of residue, and from the combustion of liquefaction products. In addition, spent catalysts and unwanted chars and residues may also require disposal. In most cases it is expected that the behaviour of trace elements in sites selected for disposal will be similar to that reported for the fly ash disposal ponds associated with coal combustion. However, attention should be paid to the extent of leaching of trace elements from such sites, especially where spent catalysts have simultaneously been disposed.

Experimental, theoretical and limited commercial experience of coal conversion processes indicate that a number of environmental problems may arise as the conversion processes are developed. As far as gasification is concerned, water treatment processes for condensate water containing high concentrations of Hg, As, Sb, Pb and other metals, disposal of water treatment solids containing high toxic metal concentrations, the combustion of char material, disposal of bottom ashes and the disposal of spent catalysts and metal deposits are all areas of environmental concern that need consideration and research. Development of liquefaction processes will engender concern over elements with volatile behaviour such as Hg, Sb and As, particularly their presence in process waters and some light oils. Those treatment processes that have been shown to effectively reduce toxic metals in effluent waters produce biotreatment sludges containing high concentrations of Hg, Se, As and other elements. These sludges, and deposits from fouled process units, may produce significant disposal problems.

5. References

ABERNATHY, R. F., PETERSON, M. I. and GIBSON, F. H. (1969). Spectrochemical analyses of coal ash for trace elements. US Bureau of Mines Report Investigation No. 7281.

ANDERSON, W. L. and SMITH, K. E. (1977). Dynamics of mercury at a coal-fired power plant and adjacent cooling lake. *Environmental Science and Technology*, **11**, 75–80.

ANDERSON, G. L., HILL, A. H. and FLEMING, K. K. (1979). *Symposium Proceedings on Environmental Aspects of Fuel Conversion Technology, IV*, Hollywood, US EPA 600/7-79-217, p. 303.

ANDREN, A. W., KLEIN, D. H. and TALOMI, Y. (1975). Selenium in coal-fired steam plant emissions. *Environmental Science and Technology*, **9**, 856–8.

BARBER, D. E. and GIORGIO, H. R. (1977). Gamma-ray activity in bituminous, sub-bituminous and lignite coals. *Health Physics*, **32**, 83–8.

BECK, H. L., GOGOLAK, C. V. and MILLER, K. M. (1979). Perturbations on the utilization of coal as an energy source. *Proceedings of the Natural Radiation Environment, III.*

BENCKO, V. and SYMON, K. (1977). Test of environmental exposure to arsenic and hearing changes in exposed children. *Environmental Health Perspective*, **19**, 95–101.

BENCKO, V., SYMON, K., CHLADEK, V. and PIHRT, J. (1976). Health aspects of burning coal with a high arsenic content. II. Hearing changes in exposed children. *Environmental Research*, **13**, 386–95.

BERRY, W. L. and WALLACE, A. (1974). Trace elements in the environment: their role and potential toxicity as related to fossil fuels. A preliminary study. University of California, Los Angeles, Report No. UCLA-12-946.

BERTINE, K. K. and GOLDBERG, E. D. (1971). Fossil fuel combustion and the major sedimentary cycle. *Science*, **173**, 233–5.

BETHELL, F. V. (1962). The distribution and origin of minor elements in coal. *The British Coal Utilisation Research Association*, **25**, 401–30.

BILLINGS, C. E. and MATSON, W. R. (1972). Mercury emissions from coal combustion. *Science*, **176**, 1232–3.

BIRGE, W. J. (1978). Aquatic toxicology of trace elements of coal and fly ash. *Energy and Environmental Stress in Aquatic Systems* (ed. by J. H. Thorp and J. W. Gibbons), pp. 219–40. Symposium Augusta, Georgia 1977. US Department of Energy.

BLOCK, C. and DAMS, R. (1975). Inorganic composition of Belgian coal and coal ashes. *Environmental Science and Technology*, **9**, 146–50.

BLOCK, C. and DAMS, R. (1976). Study of fly ash emission during combustion of coal. *Environmental Science and Technology*, **10**, 1011–17.

BOLTON, N. E., VAN HOOK, R. I., FULKERSON, W., LYON, W. S., ANDREN, A. E. and EMERY, J. F. (1973). Trace element measurements at the coal-fired Allen Steam Plant. Progress Report, June 1971–January 1973. Report No. ORNL-NSF-EPL43. Oak Ridge National Laboratory, Tennessee.

BOMBAUGH, K. J., CORBETT, W. E., LEE, K. W. and SEAMES, W. S. (1980). An environmentally based evaluation of the multi-media discharge from the Lurgi coal gasification system at Kosovo. *Symposium on Environmental Aspects of Fuel Conversion Technology, V.* St. Louis, Missouri, US Environmental Protection Agency.

BOWEN, H. J. M. (1966). *Trace Elements in Biochemistry.* Academic Press, New York.

BROWN, J., RAY, N. J. and BALL, M. (1976). The disposal of pulverised fuel ash in water supply catchment areas. *Water Research,* **10,** 1115–21.

CAMPBELL, J. A., LAUL, J. C., NIELSEN, K. K. and SMITH, R. D. (1978). Separation and chemical characterization of finely-sized particles. *Analytical Chemistry,* **50,** 1032–40.

CAMPBELL, J. A., SMITH, R. D., DAVIS, L. E. and SMITH, K. L. (1979). Characterization of micron-sized fly-ash particles by X-ray photoelectron spectroscopy (ESCA). *The Science of the Total Environment,* **12,** 75–85.

CAMPLIN, W. C. (1980). *Coal-fired Power Stations—the Radiological Impact of Effluent Discharges to Atmosphere.* National Radiological Protection Board. HMSO, London.

CHATTERJEE, P. K. and POOLEY, F. D. (1977). An examination of some trace elements in South Wales coals. *Proceedings of the Australian Institute of Mining and Metallurgy,* **263,** 19–30.

CHERRY, D. S. and GUTHRIE, R. K. (1977). Toxic metals in surface waters from coal ash. *Water Resources Bulletin,* **13,** 1127–36.

CHERRY, D. S. and GUTHRIE, R. K. (1979). The uptake of chemical elements from coal ash and settling basin effluent by primary producers II. Relation between concentration in ash deposits and tissues of grass growing on the ash. *The Science of the Total Environment,* **13,** 27–31.

CHERRY, D. S., GUTHRIE, R. K., SHERBERGER, F. F. and LARRICK, S. R. (1979). The influence of coal ash and thermal discharges upon the distribution and bioaccumulation of aquatic invertebrates. *Hydrobiologia,* **62,** 257–67.

CHRISP, C. E., FISHER, G. L. and LAMMERT, J. E. (1978). Mutagenicity of filtrates from respirable coal fly ash. *Science,* **199,** 73–5.

CLARKE, M. C. and SWAINE, D. J. (1962). *Trace Elements in Coal.* CSIRO Technical Communication No. 45.

COLES, D. G., RAGAINI, R. C. and ONDOV, J. M. (1978). Behaviour of natural radionuclides in western coal-fired power plants. *Environmental Science and Technology,* **12,** 442–6.

COLES, D. G., RAGAINI, R. C., ONDOV, J. M., FISHER, G. L., SILBERMAN, D. and PRENTICE, B. A. (1979). Chemical studies of stack fly ash from a coal-fired power plant. *Environmental Science and Technology,* **13,** 455–9.

COUGHTREY, P. J., JONES, C. H., MARTIN, M. H. and SHALES, S. W. (1979). Litter accumulation in woodlands contaminated by Pb, Zn, Cd and Cu. *Oecologia (Berl.),* **39,** 51–60.

CRAUN, J. C. and MASSEY, M. J. (1979). *Proceedings of the Symposium on Potential Health and Environmental Effects of Synthetic Fossil Fuel Technologies,* US DOE CONF 780903, p. 42.

CROCKETT, A. B. and KINNISON, R. R. (1977). Mercury distribution in soils around a large coal-fired power plant. US Environmental Protection Agency Report, 6003/3-77-063.

DENSON, N. M. (1959). Uranium in coal in the western United States. *United States Geological Survey Bulletin,* No. 1055A. United States Geological Survey, Washington, D.C.

DREESON, D. R., GLADNEY, E. S., OWENS, J. W., PERKINS, B. L., WIENKE, C. L. and WANGEN, L. E. (1977). Comparison of levels of trace elements extracted from fly ash and levels found in effluent waters from a coal-fired power plant. *Environmental Science and Technology,* **11,** 1017–21.

DVORAK, A. J. (1977). The environmental effects of using coal for generating electricity. *US Nuclear Regulatory Commission* Report No. NUREG-0252. Washington, D.C.

EISENBUD, M. and PETROW, H. G. (1964). Radioactivity in the atmosphere effluents of power plants that use fossil fuels. *Science,* **144,** 288–9.

ELSEEWI, A. A., GRIMM, S. R., PAGE, A. L. and STRAUGHAN, I. R. (1981). Boron enrichment of plants and soils treated with coal ash. *Journal of Plant Nutrition,* **3,** 409–27.

ELSEEWI, A. A., STRAUGHAN, I. R. and PAGE, A. L. (1980). Sequential cropping of fly-ash emended soils: effects on soil chemical properties and yield and elemental composition of plants. *The Science of the Total Environment,* **15,** 247–59.

FIRTH, J. N. M. (1971). The Mineralogy of the South Wales Coalfield. Ph.D. thesis, University of Bristol.

FISHER, G. L., CHRISP, C. E. and RAABE, O. G. (1979). Physical factors affecting the mutagenicity of fly ash from a coal-fired power plant. *Science*, **204**, 879–81.

FLEMING, W. J., GUTENMANN, W. H. and LISK, D. J. (1979). Selenium in tissues of woodchucks inhabiting contaminated landfills. *Bulletin of Environmental Contamination and Toxicology*, **21**, 1–3.

FORNEY, A. J., HAYNES, W. P., GASIOR, S. J., JOHNSON, G. E. and STRAKEY, J. P. Jr (1974). *Analysis of Tars, Chars, Gases and Water Found in Elements from the Synthane Process.* US Bureau of Mines, Technical Progress Report TPR 76.

FURR, A. K., KELLY, W. C., BACHE, C. A., GUTENMANN, W. H. and LISK, D. J. (1976). Multielement uptake by vegetables and millet grown in pots on fly ash amended soil. *Journal of Agricultural Food Chemistry*, **24**, 885–8.

FURR, A. K., PARKINSON, T. F., HINRICHS, R. A., VAN CAMPDEN, D. R., BACHE, C. A., GUTENMANN, W. H., ST JOHN, L. E., PAKKALA, I. S. and LISK, D. J. (1977). National survey of elements and radioactivity in fly ash soil mixtures. *Environmental Science and Technology*, **11**, 1194–201.

FURR, A. K., PARKINSON, T. F., HEFFRON, C. L., REID, J. T., HASCHEK, W. M., GUTENMANN, W. H., BACHE, C. A., ST JOHN, L. E. and LISK, D. J. (1978). Element content of tissues and excreta of lambs, goats and kids fed white sweet clover growing on fly ash. *Journal of Agricultural Food Chemistry*, **26**, 847–51.

GHASSEMI, M., CRAWFORD, K. and QUINLIVAN, S. (1979). *Environmental Assessment Report: Lurgi Coal Gasification Systems for SNG.* US Environmental Protection Agency Report EPA-600/7-79-120 Washington, DC.

GIBSON, J. (1979). *Coal and the Environment.* National Coal Board, London.

GLADNEY, E. S. (1974). *Trace element emissions of coal-fired power plants: a study of the Chalk Point Electric Generating Station.* Ph.D. thesis, University of Maryland.

GLADNEY, E. S., SMALL, J. A., GORDON, G. E. and ZOLLER, W. H. (1976). Composition and size-distribution of in-stack particulate material at a coal-fired power plant. *Atmospheric Environment*, **10**, 1071–7.

GOLDBERG, E. D., HODGE, V. F., GRIFFIN, J. J., KOIDE, M. and EDGINGTON, D. N. (1981). Impact of fossil fuel combustion on the sediments of Lake Michigan. *Environmental Science and Technology*, **15**, 466–71.

GORDON, G. E., ZOLLER, W. H. and GLADNEY, E. S. (1973). Abnormally enriched trace elements in the atmosphere. *Trace Substances in Environmental Health, VII* (ed. by D. D. Hemphill), pp. 167–74. University of Missouri, Columbia, Missouri.

GUTENMANN, W. H., BACHE, C. A., YOUNGS, W. D. and LISK, D. J. (1976). Selenium in fly ash. *Science*, **191**, 966–7.

HEE, S. S. Q., FINELLI, V. N., FRICKE, F. L. and WOLNIK, K. A. (1982). Metal contents of stack emissions, coal and fly ash. *Journal of Environmental and Analytical Chemistry*, **13**, 1–18.

HEINDRYCKX, R. (1976). Comparison of the mass-size functions of elements in the aerosols of the Gent industrial district with data from other areas. Some physico-chemical implications. *Atmospheric Environment*, **10**, 65–71.

HEINDRYCKX, R., DEMUNYNCK, M., DAMS, R., JANSSENS, M. and RAHN, K. A. (1974). Mercury and cadmium in Belgian aerosols. *Problems of the Contamination of Man and his Environment*, pp. 135–48. CEC, Luxembourg.

HEINRICHS, H. (1977). Emissions of 22 elements from brown coal combustion. *Naturwissenschaften*, **64**, 479–81.

HEINRICHS, H. (1982). Trace element discharge from a brown coal-fired power plant. *Environmental Technology Letters*, **3**, 127–36.

HODGSON, D. R. and BUCKLEY, G. P. (1975). A practical approach towards establishment of trees and shrubs on pulverized fuel ash. *The Ecology of Resource Degradation and Renewal* (ed. by M. J. Chadwick and G. T. Goodman), pp. 305–29. Blackwell, Oxford.

HULL, A. P. (1971). Radiation in perspective: some comparisons of the environmental risks from nuclear and fossil fueled power plants. *Nuclear Safety*, **12**, 185–96.

ILLYIN, L. A., KNIZNIKOV, V. A. and BARKHUDOV, R. M. (1977). A relative risk estimation of excessive frequency of malignant tumours in populations due to discharges into the atmosphere from fossil-fuel and nuclear power stations. In *IV International Congress*, Paris, pp. 189–93.

Jastrow, J. D., Zimmerman, C. A., Dvorak, A. J. and Hinchman, R. R. (1981). Plant growth and trace element uptake on acid coal refuse amended with lime or fly ash. *Journal of Environmental Quality*, **10**, 154–60.

Jaworowski, Z., Bilkiewicz, J., Dobosz, E., Grzybowska, D., Kownacka, L. and Wronski, Z. (1974). Radiation hazards to the population resulting from conventional and nuclear power plants. *Environmental Surveillance Around Nuclear Installations*. IAEA Symposium, Warsaw 1973. IAEA, Vienna.

Jaworowski, Z. and Grzybowska, D. (1977). Natural radionuclides in industrial and rural soils. *The Science of the Total Environment*, **7**, 45–52.

Joensuu, O. I. (1971). Fossil fuels as a source of mercury pollution. *Science*, **172**, 1027–8.

Jordan, S. and Schikarski, W. (1973). Evaluation of radioactive and non-radioactive trace constituents emitted from fossil-fuel and nuclear power plants. *Environmental Behaviour of Radionuclides Released in the Nuclear Industry*, pp. 525–35. IAEA Publication STI/PUB/345, Vienna.

Kaakinen, J. W., Jorden, R. M., Lawasani, M. H. and West, R. E. (1975). Trace element behaviour in coal-fired power plants. *Environmental Science and Technology*, **9**, 862–9.

Kalb, G. W. (1975). Total mercury mass balance at a coal-fired power plant. In *Trace Elements in Fuel* (ed. by S. P. Babu), pp. 154–74. Symposium of the 166th Meeting of the American Chemical Society, 1973. American Chemical Society, Washington, D.C.

Klein, D. H. and Russell, P. (1973). Heavy metals: fallout around a power plant. *Environmental Science and Technology*, **7**, 357–8.

Klein, D. H., Andren, A. W., Carter, J. A., Emery, J. F., Feldman, C., Fulkerson, W., Lyon, W. S., Ogle, J. C., Talmi, Y., Van Hook, R. I. and Bolton, N. (1975). Pathways of thirty seven trace elements through coal-fired power plant. *Environmental Science and Technology*, **9**, 937–9.

Kniznikov, V. A. and Barkhudov, R. M. (1977). Comparative assessment of radiation danger to populations from TES and APS atmospheric emissions. *Atomic Energy*, no. 3, vol. 43.

Koppenhaal, D. W., Schultz, H., Lett, R. G., Brown, F. R., Booher, H. B. and Hattman, E. A. (1979). Preprints ASC Div. *Fuel Chemistry*, **24**, 299.

Kowalczyk, G. S., Choquetter, C. E. and Gordon, G. E. (1978). Chemical element balances and identification of air pollution source in Washington, D.C. *Atmospheric Environment*, **12**, 1143–53.

Kuhnert, M. (1973). Emissions and their importance for cattle breeding and human health. *Zeitschrift für die Gesamte Hygiene und Ihre Grenzgebiete*, **19**, 89–98.

Lave, L. B. (1975). Health effects of electricity generation from coal, oil and nuclear fuel. *Symposium Proceedings on Energy and the Environment: A Risk Benefit Approach*, pp. 63–85. Birmingham.

Lett, R. G., Schmidt, C. E., De Santis, R. R. and Sharkey, A. G. Jr (1977). PERC/R112, Pittsburgh Energy Technology Centre.

Lim, M. Y. (1979). *Trace Elements from Coal Combustion—Atmospheric Emissions*. Report No. ICTIS/TR05. IEA Coal Research, London.

Linton, R. W., Loh, A., Natusch, D. F. S., Evans, C. A. and Williams, P. (1976). Surface predominance of trace elements in airborne particles. *Science*, **191**, 852–4.

Lyon, W. S. (1977). *Trace Element Measurements at the Allen Coal-fired Steam Plant*. CRC Press, Cleveland, Ohio.

McBride, J. P., Moore, R. E., Witherspoon, J. P. and Blanco, R. E. (1978). Radiological impact of airborne effluents of coal-fired and nuclear power plants. *Science*, **202**, 1045–50.

Martin, Y. E. (1974). Comparative population radiation dose commitments of nuclear and fossil-fuel electric power cycles. *Proceedings of Symposium on Population Exposures*, p. 137, Health Physics Society, Knoxville, Tennessee.

Martin, J. E., Harward, E. D., Oakley, D. T., Smith, J. M. and Bedrosian, P. H. (1971). Radioactivity from fossil fuel and nuclear power plants. *Proceedings of Symposium on Environmental Aspects of Nuclear Power Stations*, pp. 352–57. International Atomic Energy Agency, Vienna.

Mason, B. (1966). *Principles of Geochemistry*. 3rd edn. Wiley, New York.

Mastradone, P. J., Mulchi, C. L., Armbruster, J. A. and Douglass, L. W. (1982). Trace metal deposition in an agricultural environment near the Chalk Point generating station. *Journal of Environmental Quality*, **11**, 203–12.

MUKHERJEE, D. K. and CHOWDHURY, P. B. (1976). Catalytic effect of mineral matter in a North Assam coal on hydrogenation. *Fuel*, **55**, 4–8.

MUMFORD, J. L. and LEWTAS, J. (1982). Mutagenicity and cytotoxicity of coal fly ash from fluidized-bed and conventional combustion. *Journal of Toxicology and Environmental Health*, **10**, 565–86.

NATUSCH, D. F. S., WALLACE, J. R. and EVANS, C. A. (1974). Toxic trace elements: preferential concentration on respirable particles. *Science*, **183**, 202–4.

NORBERG, G. F. (ed.) (1976). *Effects and Dose Response Relationships of Toxic Metals*. Elsevier Scientific Publishing, Amsterdam.

NRIAGU, J. O. (1979). Global inventory of natural and anthropogenic emissions of trace metals to the atmosphere. *Nature*, **279**, 409–10.

OLDHAM, R. G. and WETHEROLD, R. G. (1977). Assessment, selection and development of procedures for determining the environmental acceptability of synthetic fuel plants. National Technical Information Service. Report No. FE 1795–3.

ONDOV, J. M., ZOLLER, W. H., OLMEZ, K., ARAS, N. K., GORDON, G. E., RANCITELLI, L. A., ABLE, K. H., FILBY, R. H., SHAH, K. R. and RAGAINI, R. C. (1975). Elemental concentrations in the National Bureau of Standards' environmental coal and fly ash standard of reference materials. *Analytical Chemistry*, **47**, 1102–9.

ONDOV, J. M., RAGAINI, R. C. and BIERMANN, A. H. (1978). Elemental particle size emissions from coal-fired power plants: use of an inertial cascade impactor. *Atmospheric Environment*, **12**, 1175–85.

O'TOOLE, J. J., WESSELS, T. E. and MALABY, K. L. (1981). Trace element levels and their enrichment processes in terrestrial vegetation. *Journal of Plant Nutrition*, **3**, 397–407.

PATTERSON, R. K. (1980). Ambient air downwind of the Kosovo gasification complex: *Compendium Symposium on Environmental Aspects of Fuel Conversion Technology, V*. St. Louis, Missouri, U.S. Environmental Protection Agency.

PHUNG, H. T., LUND, L. J., PAGE, A. L. and BRADFORD, G. R. (1979). Trace elements in fly ash and their release in water and treated soils. *Journal of Environmental Quality*, **8**, 171–5.

QUENTON, R. H. (1980). World coal trade through the year 2000—rapid growth and political challenges. In *11th World Energy Conference, Munich*, vol. 1A, pp. 383–402.

RAASK, E. and GOETZ, L. (1981). Characteristics of captured ash, chimney solids and trace elements. *Journal of the Institute of Energy, September*, 163–73.

RAHN, K. A. (1971). Sources of Trace Elements in Aerosols—an Approach to Clean Air. Ph.D. thesis, University of Michigan.

RAHN, K. A. (1976). *The Chemical Composition of the Atmospheric Aerosol*. Technical Report. Graduate School of Oceanography, University of Rhode Island.

ROFFMAN, H. K., KARY, R. E. and HUDGINS, T. (1977). Ecological distribution of trace elements emitted by coal-burning power generating units employing scrubbers and electrostatic precipitators. *Fourth Symposium on Coal Utilization*, pp. 192–215. Louisville, Kentucky.

ROMNEY, E. M., WALLACE, A. and ALEXANDER, G. V. (1977). Boron in vegetation in relation to a coal-burning power plant. *Communications in Soil Science and Plant Analysis*, **8**, 803–7.

RUCH, R. R., GLUSKOTER, H. J. and SHIMP, N. F. (1974). Occurrence and distribution of potentially volatile trace elements in coal. Final report. *Environmental Geology Notes*, no. 72. Illinois Geological Survey, Urbana, Illinois.

SABBIONI, E., GOETZ, L., SPRINGER, A. and PIETRA, R. (1983). Trace metals from coal-fired power plants: derivation of an average data base for assessment studies of the situation in the European Communities. *The Science of the Total Environment*, **29**, 213–27.

SCANLON, D. H. and DUGGAN, J. C. (1979). Growth and element uptake of woody plants on fly ash. *Environmental Science and Technology*, **13**, 311–15.

SCHULTZ, H., GIBBON, G. A., HATTMAN, E. A., BOOHER, H. B. and ADKENS, J. W. (1977). PERC Report, PERC/RI-77/2.

SCHULTZ, H., GIBBON, G. A., HATTMAN, E. A., BOOHER, H. B. and ADKINS, J. W. (1978). *Analytical Methods for Coal and Coal Products* (ed. by C. Karr Jr.), chap. 19, vol. 1. Academic Press, New York.

SCHWITZGEBEL, K., MESEROLE, F. B., OLDHAM, R. G., MAGEE, R. A., MESICH, F. G. and THOEM, T. L. (1975). Trace elements discharge from coal-fired power plants. *International Conference on Heavy Metals in the Environment*, pp. 533–51. Proceedings of the Toronto Conference, Ontario, Canada.

SMITH, K. R. (1977). The coal-uranium breeder: uranium from coal. *Energy*, **2**, 171–7.

SMITH, R. D. (1980). The trace element chemistry of coal during combustion and the emission from coal-fired plants. *Progress in Energy Combustion Science*, **6**, 53–119.

SMITH, R. D., CAMPBELL, J. A. and NIELSON, K. K. (1979a). Characterization and formation of submicron particles in coal-fired plants. *Atmospheric Environment*, **13**, 607–17.

SMITH, R. D., CAMPBELL, J. A. and NIELSON, K. K. (1979b). Concentration dependence upon particle size of volatilized elements in fly ash. *Environmental Science and Technology*, **13**, 553–8.

STINESPRING, C. D. and STEWART, G. W. (1981). Surface enrichment of aluminosilicate minerals and coal combustion ash particles. *Atmospheric Environment*, **15**, 307–13.

STOEWSAND, G. S., GUTENMANN, W. H. and LISK, D. J. (1978). Wheat grown on fly ash: high selenium uptake and response when fed to Japanese quail. *Journal of Agricultural Food Chemistry*, **26**, 757–9.

STROJAN, C. L. and TURNER, F. B. (1978). Trace elements and sulfur in soils and plants near the Mohave Generating Station in southern Nevada. *4th Joint Conference on Sensing of Environmental Pollutants*, pp. 537–40. American Chemical Society, Washington D.C.

STYRON, C. E. (1979). An assessment of natural radionuclides in the coal fuel cycle. *Proceedings of the Natural Radiation Environment, Vol. III.*

SWAINE, D. J. (1977). Trace elements in coal. *Trace Substances in Environmental Health, Vol. XI* (ed. by D. D. Hemphill), pp. 107–16. University of Missouri, Columbia, Missouri.

SWAINE, D. J. (1979). Trace elements in Australian bituminous coals and fly ashes. *Combustion of Pulverized Coal—The Effect of Mineral Matter* (ed. by I. M. Stewart and T. F. Wall), pp. W3, 14–18. University of Newcastle, NSW.

SWANSON, V. E., MEDLIN, J. H., HATCH, J. R., COLEMEN, S. L., WOOD, G. H., WOODRUFF, S. D. and HILDEBRAND, R. T. (1976). Collection, chemical analysis and evaluation of coal samples in 1975. US Department of Interior, Geological Survey. Open file report 76–468.

THEIS, T. L. and RICHTER, R. O. (1979). Chemical speciation of heavy metals in power plants ash pond leachate. *Environmental Science and Technology*, **13**, 219–24.

THEIS, T. L. and WIRTH, J. L. (1977). Sorptive behaviour of trace metals on fly ash in aqueous systems. *Environmental Science and Technology*, **11**, 1096–1100.

THEIS, T. L., WESTRICK, J. D., HSU, C. K. and MARLEY, J. J. (1978). Field investigations of trace metals in ground water from fly ash disposal. *Journal of the Water Pollution Control Federation*, **50**, 2457–69.

THOMAS, W. C., TREDE, K. N. and PAGE, G. C. (1979). *Environmental Assessment: Source Test and Evaluation Report—Wellman-Galusha (Glen Gery) Low Btu Gasification.* US Environmental Protection Agency Report EPA-600/7-79-185. Washington, D.C.

TORREY, S. (1978). *Trace Contaminants from Coal.* Noyes Data Corporation, Park Ridge, NJ.

TOWNSEND, W. N. and GILHAM, E. W. F. (1975). Pulverized fuel ash as a medium for plant growth. *The Ecology of Resource Degradation and Renewal* (ed. by M. J. Chadwick and G. T. Goodman), pp. 287–304. The 15th Symposium of the British Ecological Society, Blackwell, Oxford.

TUREKIN, K. K. and WEDEPOHL, K. H. (1961). Distribution of the elements in some major units of the earth's crust. *Bulletin of the American Geological Society*, **72**, 175–91.

TURNER, R. R., LOWRY, P., LEVIN, M., LINDBERG, S. E. and TAMURA, T. (1982). *Leachability and Aqueous Speciation of Selected Trace Constituents of Coal Fly Ash.* EPRI, Palo Alto, California.

TYLER, G. (1976). Heavy metal pollution, phosphatase activity and mineralization of organic phosphorus in forest soils. *Soil Biology and Biochemistry*, **8**, 327–32.

UNGA (1979). United Nations General Assembly. Technologically Modified Exposures to Natural Radiation. United Nations Publication No. A/Ac.82/R.358.

UNGA (1980). United Nations General Assembly. Technologically Modified Exposures to Natural Radiation. *United Nations Scientific Committee on the Effects of Atomic Radiation.* Vienna. UN Publication No. A/AC.82/R.371.

UNSCEAR (1977). *United Nations Scientific Committee on the Effects of Atomic Radiation.* 1977 Report on the General Assembly. United Nations publication No. E.77. ix.1, New York.

U.S. NATIONAL COMMITTEE FOR GEOCHEMISTRY (1980). *Trace-Element Geochemistry of Coal Resource Development Related to Environmental Quality and Health.* Assembly of Mathematical and Physical Sciences, National Research Council. National Academy Press, Washington, D.C.

VALKOVIC, V. (1983). *Trace Elements in Coal. Volumes I and II.* CRC, Boca Raton, Florida.

VAN HOOK, R. I. (1978). Potential health and environmental effects of trace elements and radionuclides from increased coal utilization. Oak Ridge National Laboratory, Tennessee. Report No. ORNL-5367.

VINOGRADOV, A. P. (1959). *The Geochemistry of Rare and Dispersed Chemical Elements in Soils*, 2nd edn. Consultants Bureau, Inc.

WADE, N. (1979). Synfuels in haste, repent at leisure. *Science*, **205**, 167–8.

WALSH, P. R., DUCE, R. A. and FASCHING, J. L. (1979). Considerations of the enrichment, sources and flux of arsenic in the troposphere. *Journal of Geophysical Research*, **84**, 1719–26.

WANGEN, L. E. and TURNER, F. B. (1980). Trace elements downwind of a coal-fired power plant. *Water, Air & Soil Pollution*, **13**, 99–108.

WANGEN, L. E. and WILLIAMS, M. D. (1978). Elemental deposition downwind of a coal-fired power plant. *Water, Air & Soil Pollution*, **10**, 33–44.

WIERSMA, G. B. and CROCKETT, A. B. (1978). Trace elements in soil around the Four Corners power plant. Environmental Protection Agency Report, 600/3-78-079.

YAROVSKY, P. M. and AKHTAR, S. (1974). *Proceedings of the Symposium on Environmental Aspects of Fuel Conversion Technology*, St. Louis, EPA-650/2-74-118, 325–30.

ZOLLER, W. H., GLADNEY, E. S., GORDON, G. E. and BORS, J. J. (1974). Emissions of trace elements from coal-fired power plants. *Trace Substances in Environmental Health VIII* (ed. by D. D. Hemphill); pp. 167–72. University of Missouri, Columbia, Missouri.

ZUBOVIC, P., SHEFFEY, N. B. and STADNICHENKO, T. (1967). Distribution of minor elements in some coals of the Western and South Western regions of the Interior Coal Province. *US Geological Survey Bulletin*, 117-D.

9

The Environmental Significance of Coal-derived Carbon Compounds

1. Introduction 218
2. Human Health Effects of Coal-derived Carbon Compounds 220
   - 2.1. Early Studies on Coal Tar 220
     - 2.1.1. Experimental work with coal tar 220
     - 2.1.2. The discovery of chemically pure carcinogens 221
   - 2.2. Carcinogenic Polycyclic Aromatic Hydrocarbons (PAH) 222
     - 2.2.1. Carcinogenic activity and PAH structure 223
     - 2.2.2. Benzo(*a*)pyrene (BaP) 224
     - 2.2.3. Environmental sources of PAH 227
3. Polycyclic Aromatic Hydrocarbons in the Food Chain 229
   - 3.1. Atmospheric PAH 229
   - 3.2. PAH in Soils 230
     - 3.2.1. Contamination by soil treatment 230
   - 3.3. PAH Uptake by Plants 231
     - 3.3.1. PAH levels in food 232
   - 3.4. PAH in Water 233
     - 3.4.1. Freshwater reservoirs 233
     - 3.4.2. Seas and oceans 234
   - 3.5. Bacterial Degradation of PAH 236
4. Natural Origins of Polycyclic Aromatic Hydrocarbons 236
   - 4.1. Synthesis by Plants and Micro-organisms 236
   - 4.2. Volcanic Activity 236
5. Polycyclic Aromatic Hydrocarbons and Coal Combustion and Conversion Processes 237
   - 5.1. Coal Combustion 237
   - 5.2. Coal Conversion 238
     - 5.2.1. Coal gasification 238
     - 5.2.2. Coal liquefaction 238
6. Polycyclic Aromatic Hydrocarbon Body Burdens in Man 239
7. References 240

1. Introduction

Many carbon compounds, including coal derivatives, formed in manufacturing cycles, possess high biological and pathological activities. Polycyclic aromatic hydrocarbons (PAH) are of considerable interest in this respect as many are known to be mutagenic and some have been shown to be carcinogenic in animal systems.

Polycyclic aromatic hydrocarbons can originate from natural sources or be emitted in the course of a wide range of anthropogenic activities. These include automobiles and other petroleum-burning engines (including aircraft), refuse burning industrial processes, domestic heating appliances and tobacco smoking. PAH compounds are produced by the incomplete combustion and pyrolysis of fossil fuels and other organic materials. They are emitted to the atmosphere either in the gaseous phase or adsorbed on very small particles. 70–90 % of PAH in the atmosphere is in a particulate form that is in the respirable range (< 5 μm in diameter). Relatively long-term monitoring of PAH concentrations in the Netherlands (Brasser, 1979) has revealed that during the period that natural gas replaced coal and oil as a fuel for heating buildings (and to some extent in factories and power stations) there was a spectacular fall in concentrations from relatively high to negligible levels. This fact, and the data summarized by Suess (1976) and given in Table 9.1, therefore make it natural that PAH should be one focus of attention for the possible health impacts of coal combustion and coal conversion technologies.

TABLE 9.1. *Estimated benzo(a)pyrene emissions to the atmosphere (Suess, 1976)*

	BaP emission in t yr^{-1}		
Sources of formation	USA	Worldwide excluding USA	World-wide
Heating and power generation			
Coal	431	1945	2376
Oil	2	3	5
Gas	2	1	3
Wood	40	180	220
Total	475	2129	2604
Industrial processes			
Coke production	192	841	1033
Catalytic cracking	6	6	12
Total	198	847	1045
Refuse and open burning			
Enclosed incineration			
Commerce and industry	23	46	69
Other	11	22	33
Open burning			
Coal refuse	340	340	680
Forest and agriculture	140	280	420
Other	74	74	148
Total	588	762	1350
Vehicles			
Trucks and buses	12	17	29
Automobiles	10	6	16
Total	22	23	45
Grand Total	1283	3761	5044

In view of the interacting environmental factors, and range of sources of PAH emissions, the impact of coal conversion technologies is assessed against a historical background and in the light of a wide range of other anthropogenic activities.

2. Human Health Effects of Coal-derived Carbon Compounds

2.1. *Early Studies on Coal Tar*

The prominent English surgeon P. Pott described, in 1775, an occupational disease of chimney sweeps. He correctly ascribed skin cancer affecting the scrotum and thighs to the continual exposure of the tissue to soot. These observations showed that there was a close correlation between coal products and malignant growths in man. This has subsequently been confirmed as a result of studies of occupational cancer in man in other industries: for example, from contact with unrefined paraffins; in workers making briquets from coal dust (Table 9.2); in workers in continual contact with petroleum products and in other situations. Skin tumour formation appears to be directly dependent on the duration of contact with the carcinogenic agent which, in the case of the coal-briquet makers, was pitch and tar. Since the technology of briquet-making has been changed in order to eliminate skin contact, skin tumours have disappeared.

2.1.1. *Experimental work with coal tar*

Systemic application of coal tar to the skin of the ears of rabbits for a year was found to induce, initially, papillomas and then carcinomas that metastasized into the lymph glands (Yamagiwa and Ichikawa, 1918); this has been

TABLE 9.2. *Development of professional skin tumours in the briquet production industry (Teutschlaender, 1929).*

No. of years of work	No. of patients	Percentage of patients with skin tumours (papillomas, cancer)
1–5	40	17
5–10	38	21
10–15	22	36
15–20	9	67
20–25	13	69
25–30	14	71
30–35	13	77
35–40	5	80
>40	4	100

confirmed by other workers using different animal species. It was then established that coal tars differ in their carcinogenicity; for instance "Limburg coal tar" was found not to be carcinogenic at all. Coal type and the temperature of coking were found to be of importance in this respect. Coal tar fractions obtained at different temperatures had different carcinogenic properties.

Coal tar is a complex mixture of a larger number of aromatic hydrocarbons (benzene, naphthalene, anthracene, phenanthrene and their homologues), aromatic acids and nitrogen-containing compounds and it transpired that it was better to study the carcinogenic effect of separate fractions obtained by distillation at different temperatures. It became established that only the tar fractions distilled at 400–600°C and above are carcinogenic; distillation at 800–850°C results in the highest yield of carcinogenic products. It was concluded that carcinogenic products have a high boiling point, are stable when heated repeatedly, do not contain sulphur or arsenic, are neither an acid nor an alkali and are not nitrogen-containing compounds. These characteristics placed the neutral, polynuclear aromatic hydrocarbons with a high boiling point under suspicion as the carcinogenic agents. A study of the actions of sulphuric and picric acids on the different coal tars and schist resins suggested that the carcinogenic principle in tars is found amongst the higher polynuclear hydrocarbons; the treatment of certain tars with sulphuric acid was found to deprive them of carcinogenic activity.

2.1.2. The discovery of chemically pure carcinogens

Carcinogenic tars could be obtained from various materials by heating them to 560–880°C; for example, tars capable of producing skin cancer in mice were obtained by heating isoprene to 820°C or acetylene to 700°C. These substances had to be distilled in a hydrogen atmosphere and it was established that the tars obtained represented a mixture of higher hydrocarbons.

In 1930 the fluorescence of a number of known carcinogenic tars and mineral oils was described (in ultraviolet light, a spectrum with three bands at 400, 418 and 440 nm was seen). Since benz(*a*)anthracene and a number of other substances from the same group possess a similar fluorescence, this led to a range of polycyclic aromatic hydrocarbons (PAH) being investigated for carcinogenic properties, among them dibenz(*a*,*h*)anthracene (Cook *et al.*, 1932). Painting the skin of mice was a 0.3% solution of this substance in benzene resulted in almost half of the animals developing, after 6 months, firstly papillomas and then carcinomas. Injection of several milligrams of this substance under the skin of mice and rats led to the development of sarcomas in the majority of animals treated.

Comparison of the carcinogenic activity of dibenz(*a*,*h*)anthracene with that of certain well-known types of coal tar showed that the latter possessed a more pronounced action and hence contained a more powerful carcinogenic substance. After persistent and painstaking work, Cook, Hewitt and Hieger (1933) succeeded in isolating this substance from coal tar and identifying it as benzo(*a*)pyrene. This hydrocarbon induced cancer in mice in half the time taken by dibenz(*a*,*h*)anthracene and in a wider range of animal species. Benzo(*a*)pyrene (BaP) was subsequently prepared synthetically.

2.2. *Carcinogenic Polycyclic Aromatic Hydrocarbons* (*PAH*)

The names and structural formulae of several of these compounds are shown in Fig. 9.1.

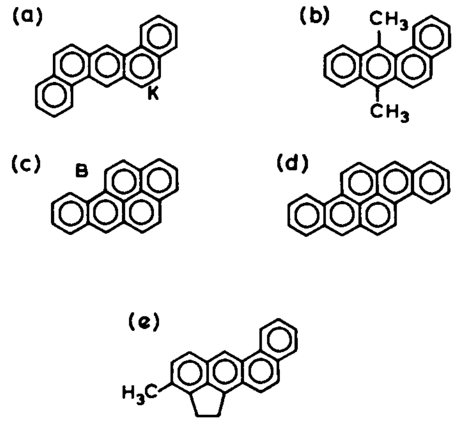


FIG. 9.1. The structural formulae of some important carcinogenic aromatic hydrocarbons (alternative names are given in parentheses) showing the "K-region" (K) and the "bay-region" (B): (a) DBahA Dibenz(*a*, *h*)anthracene (1,2-5,6-dibenzanthracene); (b) DMBA 7,12-Dimethylbenz(*a*)anthracene (9,10-dimethyl-1,2-benzanthracene); (c) BaP Benzo(*a*)pyrene (3,4-benzpyrene); (d) DBahP Dibenzo(*a*, *h*)pyrene (3,4-8,9-dibenzpyrene); (e) MCA 3-Methylcholanthrene (3-methyl-1,2-dihydrobenz(*j*)acyanthrylene).

DBahA was synthesized in 1929 and first tested in 1930–2 (Cook *et al.*, 1932). It possesses average carcinogenicity, producing tumours in many species of laboratory animals, even in animals refractory to PAH, such as the guinea pig.

Dimethylbenz(*a*)anthracene was first synthesized in 1938 independently in the USSR and in the USA, and when tested it proved to be the most important of all the carcinogenic PAHs. Painting the skin of mice with a 0.05 % solution of DMBA in benzene leads to the appearance of papillomas on or around the 40th day of treatment and, in experiments using a 0.1 % solution, papillomas appeared within 2–3 weeks. Painting the skin of the ears of rabbits with a 1 % benzene solution of DMBA produced cancer in almost all the animals in 4 months.

BaP is probably the best known and most intensively investigated carcinogenic PAH; it has been tested in a whole range of laboratory animals and its carcinogenic potency is about twice that of DBahA.

DBahP, which was synthesized in 1937, shows a carcinogenicity approximately equal to that of BaP when tested in mice.

Methylchloranthrene (MCA) was synthesized in the UK and the USA from desoxycholic acid, a normal component of human and animal bile. MCA is approximately equal to BaP in its carcinogenic properties, but when applied to the skin of mice MCA elicits a more prominent inflammatory reaction.

2.2.1. Carcinogenic activity and PAH structure

Not all polycyclic aromatic hydrocarbons are carcinogenic, and those that are differ widely in terms of potency. A great deal of effort has therefore been expended in trying to discover the scientific basis for the structure-activity relationships that exist within the hydrocarbons as a class of chemical carcinogens. In addition, the mechanism of action of PAHs as carcinogens has attracted considerable attention, not least because it was difficult to envisage quite how these compounds, which are commonly formed at high temperatures and are relatively inert chemicals, might change normal into malignant cells. So far, somewhat better progress has been made in the second of these areas of interest than has occurred in the first.

The K-regions of PAHs (Fig. 9.1), which were known to be the most chemically reactive and electron-dense regions of hydrocarbon molecules, initially attracted much attention (Pullman and Pullman, 1955) and this was followed by a period in which the metabolism of the hydrocarbons was intensively investigated (for a review see Sims and Grover, 1974). It was known that the aromatic double bonds present in the molecules could be metabolized (Fig. 9.2) by the so-called drug-metabolizing enzymes that are widely distributed throughout the animal kingdom and that enable many species to transform foreign compounds into water-soluble derivates that can be excreted. Moreover, it was also found that when PAHs were metabolized by these enzymes *in vitro* reactive intermediates were formed that could react covalently with, for example, DNA and protein. Initially these intermediates, which of course might also be formed in susceptible tissues *in vivo*, were thought to be simple epoxides. However, over the last few years it has become apparent that the reactive species are most probably diol-epoxides (Fig. 9.2). The particular diol-epoxides that are most reactive are diol-epoxides of the "bay region" type in which the epoxide moiety is adjacent to a "bay" in the molecule (Fig. 9.3), i.e. the region formed by the presence of an angular benzene ring. Metabolites of this type are currently under active investigation (see review by Grover, 1980) since differences in their properties and in their rates of formation and further metabolism may help to explain the very marked structure-activity relationships that exist within the PAHs.

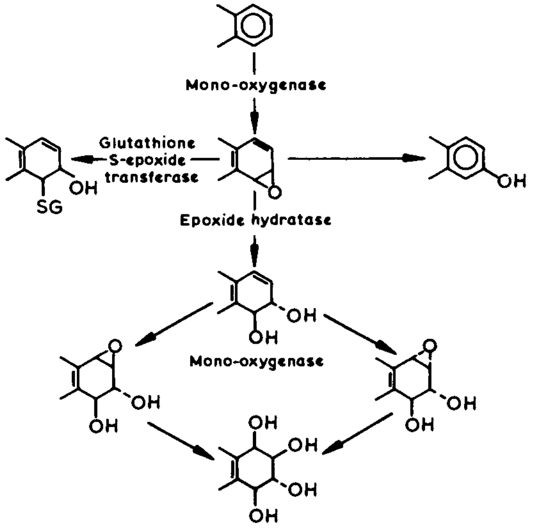


FIG. 9.2. The metabolism of an aromatic ring in a polycyclic hydrocarbon to hydroxylated derivates.

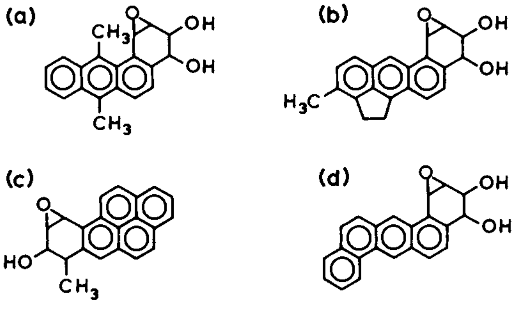


FIG. 9.3. Reactive "bay-region" diol-epoxides of (a) 7,12-Dimethylbenz(*a*)anthracene; (b) 3-Methylcholanthrene; (c) Benzo(*a*)pyrene; (d) Dibenz(*a,h*)anthracene.

2.2.2. *Benzo(a)pyrene (BaP)*

Even when present in complex mixtures, BaP can be determined quantitatively by relatively precise and easily performed methods (Shabad *et al.*, 1972; Egan *et al.*, 1979). Spectro-luminescence methods, in which fine-structured spectra are obtained at low temperatures (−196°C) (Khesina, 1979; Yanisheva and Kireeva, 1979), and methods based on gas chromatography (Grimmer and Bohnke, 1979) have frequently been employed.

BaP is the most widely distributed PAH carcinogen found in the environment. Of the several thousand samples of air, soil and water that have been examined in the USSR, there was none from which it was absent. Samples rich in BaP are usually found in the exhaust gases of automobiles, aeroplanes and motorized ships. Benzene extracts of condensates of these gases, and of soots, produce papillomas and skin cancers in mice (Shabad, 1973). Painting the skin of mice with the products of the distillation of coal tar and of petroleum containing different concentrations of BaP gives neoplasms that correspond in number to the BaP concentration used (Yanisheva, Chernickenko and Balenko, 1977).

In a study of automobile emissions (Huenigen *et al.*, 1978), a relationship was established between the concentration of BaP and the concentration of other PAH compounds that were present. It is therefore possible to calculate the approximate quantities of the other PAH compounds present from BaP estimations, although for some types of automobile exhausts the relationship does not hold.

Of late, many new forms of occupational tumours have been described. These are often due to exposure of products containing large quantities of BaP that are formed in the partial combustion of fossil fuels. Lawther and Waller (1976) found that roofing workers who had sustained contact with material containing coal tar and pitch contracted skin cancer 4 times and lung cancer 1.5 times more frequently than control groups. Fox, Lindars and Owen (1974) have reported that a higher incidence of malignant disease of the respiratory organs is seen in workers in the rubber and cable industries, and Konstantinov *et al.* (1974) have described skin cancers in workers in aluminium electrolysis plants. In all these cases the contact with PAH in general, and with BaP in particular, was over a long period. BaP was not the only carcinogenic agent present, and it probably acted in combination with other PAHs. It seems reasonable, however, to use BaP as typical of the entire group.

Testing both coal tars isolated from industrial products and wastes produced by various industries shows that carcinogenic activity is correlated with the physicochemical properties of BaP. In addition, the greater the BaP concentration present in the product studied, the higher seem to be the number of neoplasms that occur in treated animals, although it is too early yet to assume a simple linear dose-response curve or to assume zero repair. The tumours induced malignant tumours capable of metastasizing (Tables 9.3 and 9.4).

BaP can remain unchanged in the soil for long periods and laboratory and field experiments have shown that the time required for complete destruction (99 %) of BaP in the soil is from 3 to 5 years. The exact fate of BaP released to the atmosphere is unclear. It was, for some time, assumed that it was extremely stable, but photochemical decomposition is now an accepted fate (NAS, 1972; Grimmer, 1979). However, recently, stabilization of benzo(*a*)pyrene when adsorbed on to coal fly ash has been investigated (Korfmacher *et al.*, 1980). A number of adsorbed polycyclic aromatic hydrocarbons will spontaneously oxidize in the absence of light. The decomposition rate appears to be fly ash-dependent.

TABLE 9.3. *Development of tumours in mice ($CC_{57}W$ strain) painted with treated products of coal tar and petroleum with different BaP contents (Yanisheva, Chernichenko and Balenko, 1977; Yanisheva and Kireeva, 1977)*

			No. of animals with tumours						
			Skin			Other organs			
Material studied	Mean BaP content (%)	No. animals surviving till the first	Total	Malignant	With metastases	Total	Malignant	With metastases	Systemic lesions (leukaemia)
Coal tar pitch	0.24	43	38	35	9	24	1	1	0
Petro-bituminous compounds from cracking residue									
BH-5	0.0138	49	9	6	3	11	9	0	2
BH-4	0.0027	42	4	2	0	4	0	0	0
Petro-bituminous compounds from petroleum tars									
BH-5	0.00002	43	2	1	0	5	3	0	0
BH-2	0	40	0	0	0	1	1	0	0

TABLE 9.4. *Carcinogenic properties of waste products of the coal processing industry (Yanisheva, 1939)*

Coal tar under study	Months to appearance of the first tumour	Percentage of animals with tumours		BaP concentration in coal-tar (%)
		Malignant	Benign	
From coke gas exudate from	2	95.4	4.6	0.1
coke furnaces	4	86.6	13.4	0.06
From atmospheric dust (near coking factory)	6	44.4	38.9	0.03
From phenol sediment collectors	2	52.7	19.1	0.03

A comparative study of different coal and wood tars that have been employed in the preparation of tar ointments (Shabad *et al.*, 1970) showed that their carcinogenicity in mice corresponds to their BaP content, the strongest carcinogenic activity being seen after painting the skin of mice with lianthrol (CIBA, Switzerland), used in the preparation of "Locacorten-Tar". However, this straightforward relationship has recently been questioned.

Investigations (Klipinitser *et al.*, 1977) showed that the anthracene fraction of coal tar contains especially high quantities of PAH and BaP in particular (up to 27,000 $\mu g\,kg^{-1}$). However, BaP content is markedly decreased in SANT-H tar and other products obtained by continuous combustion. This is an example of the advantages that may be achieved by improved technology which are essential since polycyclic organic matter (POM), derived from the combustion of fossil fuels, mainly coal, may cause 5–10 cases of lung cancer per 100,000 males yr^{-1}. An enhancement of 1 μg 1000 m^{-3} in atmospheric POM gives a 50% increase in deaths from lung cancer. In some situations, BaP concentration can provide a good indication of POM concentrations (Shabad, 1978), but as BaP concentrations do not necessarily represent the carcinogenicity of a PAH sample, these correlations are no longer accepted (Grimmer, 1979).

2.2.3. *Environmental sources of PAH*

The main anthropogenic sources of carcinogenic hydrocarbons are heating systems, emissions from industrial and power plants, automobile emissions and aviation exhausts and these are considered separately below.

Heating systems. In 1948 a study was begun of atmospheric PAH in three cities in the USSR: Moscow, Ivanovo and Grozny. These cities were chosen because coal was the main fuel used for heating in Moscow, wood predominated in Ivanovo, whilst liquid fuels were the main heat source in the third city. It was found that the quantity of BaP in the atmosphere depends not so much on the type of fuel as on the methods of combustion used and that improved combustion decreases the quantity of BaP emitted into the atmosphere. These studies established, therefore, a basis for prophylactic measures.

Improvements in combustion and in smoke control can be gained by replacement of individual heating systems with regional and central heating installations combined with electricity generation. The use of gas in place of other types of fuel, increased electricity generation and neighbourhood heating schemes all represent real measures that can be taken against atmospheric pollution. The centralized heat supply of cities by large thermo-electric stations has become a decisive measure in enabling the closure of thousands of small boilers with unfavourable conditions for coal burning and the substitution for them of a few thermo-electric stations with favourable coal-burning conditions and better emission control. Rational city planning, which separates industrial zones from residential areas, and the creation of "green zones" also bring improvements and much has been achieved in Moscow, and in other large cities, through improvements such as these.

Industry. For certain cities and regions, it is industrial emissions and automobile exhausts rather than domestic heating systems that make the greatest contribution to atmospheric pollution. A pitch coking plant in the village of Buroz, for example, was responsible for much of the atmospheric pollution. Changes in plant design, giving improved combustion characteristics, decreased the quantity of smoke, pitch and BaP emitted from this plant (Shabad and Dikun, 1959).

Investigations of a number of metallurgic, oil refining, pitch coking and briquet manufacturing plants confirmed the dependence of the quantity of carcinogenic hydrocarbons emitted on the combustion characteristics (Yanisheva, Chernichenko and Balenko, 1977). The main measures to be taken to reduce atmospheric pollution by carcinogenic substances must therefore be technological improvements in plant design and operation. A considerable decrease in atmospheric pollution in London has been brought about by improvements in the design of heating systems and industrial plants, and by restrictions that have been placed on the types of fuel that may be used (Lawther and Waller, 1976). The quantity of BaP present in the atmosphere in London has decreased tenfold in a 25-year period.

Automobile emissions. The study of automobile exhaust gases is of significance to the assessment of coal conversion effects, as these can be an important source of atmospheric pollution with carcinogenic substances in cities. Waller (1952) showed that automobile exhaust gases contain BaP and in the USSR carcinogenic substances have been detected in soot and in exhaust gases from automobiles by many workers (Gurinov, Tugarinova and

Vasilieva, 1962; Gofmekler *et al.*, 1963; Shabad, Khesina and Khitrovo, 1968; Zabezhensky, 1968; Varshavsky *et al.*, 1970). Atmospheric BaP concentrations on highways exceed the background concentration by a factor of at least 1.5 and it can be shown that, with a slight lag period, BaP concentration is positively correlated with traffic density.

The mean concentration of BaP in the air on main roads is approximately 3 μg 100 m^{-3}. If the volume of air inhaled by a man in 24 hours is 20 m^3, then the inhabitants of a large city can inhale about 0.6 μg BaP day^{-1} from automobile exhaust gases. Roughly the same quantity of BaP is contained in the smoke of 50 cigarettes. Studies in a small town in Austria showed that from 1958 to 1970 mortality from cancer among people living near the highway was 9 times higher than that occurring amongst people living some way away from it, in these particular studies. Soil near the highway also had a PAH content that was much higher than that present in soil distant from it. Improvements in engine design are required in order to reduce PAH emissions in exhaust gases (Heunigen *et al.*, 1978; Shabad, Khesina and Khitrovo, 1968). Epidemiological evidence from a study of residents in a Swiss town in relation to the proximity of their habitation to the roadway (Blumer, Jaumann and Reich, 1972) showed that death from cancer was 9 times as frequent for residents near the roadway. The reduction of lead in gasoline is likely to lead to increased PAH emissions from this source (Candeli *et al.*, 1974).

Aviation exhausts. The presence of carcinogenic hydrocarbons in the exhaust gases of automobile engines suggested that other internal combustion engines using the same fuel should be investigated. It has been shown that the soot from exhaust pipes of aviation engines and the exhaust gases themselves contain BaP (Shabad and Smirnov, 1976). The gas turbine engines of modern aircraft emit exhaust gases with 2–4 mg BaP min^{-1} and many times more is present during take off (up to 40 mg).

Extracts from soots formed by aviation engines induce malignant tumours in mice in 100 % of cases, and soots from piston engines induce neoplasms 2 weeks earlier than do soots from turbo-jets. All the tumours formed initially were papillomas and these became transformed later into epidermoid cancers; in some cases carcinosarcomas and sarcomas were seen.

BaP has also been detected in the plants and soil around Moscow airport, the quantities reducing with increasing distance away from the centre of the airport. Dust collected from the runway itself contained 182 μg kg^{-1} of BaP.

3. Polycyclic Aromatic Hydrocarbons in the Food Chain

3.1. Atmospheric PAH

The sources of atmospheric PAH have already been outlined. Aspiration samples taken in cities in the USSR show that concentrations vary considerably. Generally, the larger the city, the higher the concentration and levels of PAH, and concentrations in industrial regions exceed those present in rural areas. Especially high concentrations of PAH are found associated with coal and petroleum utilization.

In the USSR, PAH levels are generally higher in Winter than Summer, with the exception of certain large cities, where seasonal variations in traffic densities (higher in Summer) override this trend.

Certain factors can help in removing PAH from the atmosphere. Photo-oxidation, thermal decomposition, the action of nitrogen oxides, ozone, sulphur oxides and the action of other oxidants have been said to cause the destruction of carcinogenic PAH, and particulate and aerosol sedimentation also result in their removal (Shabad and Dikun, 1959; Shabad, 1973). However, the products of some of these reactions may be more toxic than PAH and thus present increased health hazards (NAS, 1972; Nikolau, Masclet and Mouvier, 1984). One example of this is the action of nitrogen oxides in adding to, substituting or oxidizing PAH. Butler (1981) and Butler and Crossley (1981) suggested that an atmospheric chemical reaction converted BaP into a non-carcinogenic 6-nitro derivative. On the other hand, Pitts and Katzenstein (1981) demonstrated the production from BaP of both weak and strong mutagens (for example, 3-nitrobenzo(*a*)pyrene).

3.2. *PAH in Soils*

Rainout, washout and sedimentation lead to the addition of carcinogenic hydrocarbons to the soil, particularly in the vicinity of large industrial towns. Shabad and Dikun (1959) demonstrated the presence of benzo(*a*)pyrene in a park zone in Leningrad. Blumer (1961), Borneff and Fabian (1963), Mallet and Heros (1962), and Zdrazil and Picha (1966) have similarly demonstrated the presence of BaP in soils in the USA, the FRG, France and Czechoslovakia respectively. Yanisheva and Kireeva (1977) were able to demonstrate that there was a relationship between soil BaP concentration and distance from industrial sources, particularly coke-making plants.

Losses of BaP from the soil take place mainly during Spring and Summer following increased deposition during the Winter. Leaching occurs from the upper down to the lower layers of the soil (Table 9.5).

Plant uptake, through the roots, can also account for some reduction in soil concentration and, in addition to decomposition by ultraviolet radiation, bacterial breakdown also occurs.

3.2.1. *Contamination by soil treatment*

The use of irrigation water from contaminated sources can add BaP to soils but by far the most noticeable effects result from the treatment of soil with certain hydrocarbon products in order to prevent erosion. In the USSR,

TABLE 9.5. *Distribution of benzo(a)pyrene in soil layers, in μg kg^{-1} (Shabad, 1973)*

	Distance from oil-refining plant		
Depth of layer (cm)	250 m	500 m	1500 m
2.5	11.90	1.20	0.12
5.0	12.97	1.24	0.11
10.0	11.08	1.95	0.13
20.0	17.34	1.62	0.09
30.0	14.53	1.12	0.008
50.0	13.53	0.081	0.001
80.0	2.77	0.18	0.002
110.0	5.40	0.002	0.0005
150.0	–	0.002	–

TABLE 9.6. *BaP content (μg kg^{-1}) in soil samples treated with Nerosine in May 1969, in (a) Cotton–Uzbek SSR and (b) Spring wheat–Kazak SSR (Shabad, 1973)*

(a)

Soil depth (cm)	Control		Nerosine 1.4 t ha^{-1}		Nerosine 2.1 t ha^{-1}	
	May	Sept	May	Sept	May	Sept
0–10	4.7	8.2	64.0	47.0	235.0	153.0
10–20	4.9	2.8	15.0	47.0	46.0	46.0
20–30	4.4	5.0	32.0	54.0	9.0	79.0

(b)

Soil depth (cm)	Control				Nerosine-treated			
	1969			1970	1969			1970
	June	July	Aug.	Aug.	June	July	Aug.	Aug.
10–10	0.8	1.0	3.5	1.0	238.0	186.0	169.0	37.0
10–20	0.34	1.0	1.2	0.8	95.0	42.0	14.0	7.2
20–30	0.1	0.3	1.2	0.1	9.0	25.0	21.0	5.2

"Nerosine", an Estonian oil shale product, is added to soil as an anti-erosion treatment and the BaP content of this product may vary from 30 to 100 ppm. It has been shown that Nerosine is carcinogenic in mice. After application to the soil surface, BaP from the product rapidly penetrates deeper soil layers (Table 9.6).

3.3. PAH Uptake by Plants

Plants grown on soils with high PAH concentrations absorb BaP through the roots and translocate it to the shoots. Asters (*Aster nova-belgii*) and nasturtiums (*Tropaeolum majus*) grown on soil highly polluted with BaP contained 6.3 times as much BaP as the same species grown on control soils. However, no significant elevation of BaP levels was found in the grain of cereals (Winter and Spring wheat, rye, barley or maize) grown on Nerosine-treated soil, but treatment does result in BaP accumulation in sunflower (*Helianthus annuus*) seeds.

3.3.1. *PAH levels in food*

There is evidence of relatively high concentrations of PAH (particularly BaP) in some food products (Fritz *et al.*, 1977). Potatoes may contain up to 1 $\mu g\,kg^{-1}$ and even higher amounts are present in samples from some particular localities (Gräf and Diehl, 1966) usually in the skins. In other food products from Georgia (particularly vegetables and fruit), BaP concentrations were seldom above background levels (Ilnitsky, Belitsky and Shabad, 1975). Levels in soil in excess of 100 $\mu g\,kg^{-1}$ may give rise to significant increases in BaP concentrations in vegetables.

Table 9.7 gives details of BaP levels in soils and vegetable products in relation to locality. The effects of proximity to a highway are evident. The concentrations present in vegetables depend not only on location but also on deposition characteristics, soil factors and the crop species concerned. The highest concentrations were found in cabbages (*Brassica oleracea*) and the lowest in pumpkins (*Cucurbita pepo*). Particulate deposition on the leaves of cabbages accounted for a high proportion of the concentration.

The mean cumulative dose of PAH to a 70-year-old human might be 3 mg from food (excluding vegetables), 2 mg from vegetables and 0.3–0.4 mg from drinking-water. In addition, some PAH might accrue from food packaging, processing and preparation.

TABLE 9.7. *BaP content ($\mu g\,kg^{-1}$) in soils and vegetables grown in different areas of the Ukraine (Yanisheva and Kireeva, 1977)*

Population point	In the soil		In vegetables		Dose of benzo(*a*)-pyrene entering the human body in vegetables over 70 years (mg)
	Near the highway	Among houses	Near the highway	Among houses	
Large administrative centre	1937 ± 306	249 ± 70	0.67 ± 0.09	0.17 ± 0.04	3.1–12.1
Average industrial town	348	15 ± 5.5	0.21 ± 0.08	0.11 ± 0.04	2.0 ± 3.8
Small town	160.6	38 ± 11.4	–	0.16 ± 0.02	2.9
Settled agricultural area	32.0	18.7 ± 4.5	–	–	–
Uninhabited area	5.0 ± 1.6	–	–	0.10 ± 0.02	1.8

3.4. PAH in Water

3.4.1. Freshwater reservoirs

Systematic studies of the fate of BaP and other PAH compounds in freshwater reservoirs have not been undertaken, although some information is available (Ilnitsky, Voronin and Solenova, 1974; Ilnitsky, Belitsky and Shabad, 1975; Shabad, 1973). The main sources of PAH in reservoirs are: exhaust gases from the engines of boats, effluents from industrial plants, urban run-offs from cities and seepage of sub-soil water.

Comparison of BaP content in plankton, bottom sediments and higher water plants in two water reservoirs near Moscow showed that the reservoir where the passage of boats was authorized had 2–10 times more BaP than the one where such activities were forbidden.

Table 9.8 shows the significance of sewage water for pollution of a large river with BaP.

Table 9.9 shows the effect of the input from a large city into a river. Concentrations in the water, in aquatic plants and in bottom sediments are many times higher than they are in the river upstream of the city, the contamination presumably resulting from industrial emissions and urban run-off.

As an indication of PAH contamination in reservoirs, BaP concentrations were used. BaP possessed a very low solubility in water (about 0.1 $\mu g\,l^{-1}$), but organic solvents (acetone, petroleum and petroleum products, detergents) increase this solubility; the stability of BaP in water is sufficient to enable it to

TABLE 9.8. *BaP concentrations in the water of a large river in 1971–4 (Lembik, 1979)*

Position	No. of samples	BaP content ($\mu g\,l^{-1}$)
26 km upstream from the inflow of sewage water	32	0.0130 ± 0.0002
2 km upstream from the inflow of sewage water	36	0.0110 ± 0.0008
500 m downstream from the inflow of sewage water	36	0.0250 ± 0.0008
10 km downstream from the inflow of sewage water	36	0.0120 ± 0.0005

TABLE 9.9. *BaP concentration in a river upstream from and downstream from a large city (Shabad, 1973)*

	Upstream	Downstream
Water ($\mu g\,l^{-1}$)	0.001	0.04
Plankton ($\mu g\,kg^{-1}$)	–	475
Aquatic plants ($\mu g\,kg^{-1}$)	0.0–6.0	75
Bottom sediment ($\mu g\,kg^{-1}$)	18	146–403

spread considerable distances. After 30–45 days' storage, 5–20 % of the initial concentration of 10 $\mu g l^{-1}$ remained in both tap- and lake-water.

The BaP concentration in reservoirs in industrial regions is higher than in rural districts, and a study of several dozen bodies of water (rivers, lakes and reservoirs) in the USSR permitted a number of significant trends to be recognized (Table 9.10).

TABLE 9.10. *Degree of pollution by BaP in water bodies (Shabad, 1973)*

	Background level	Degree of pollution with BaP		
		Moderate	Considerable	High
Water ($\mu g l^{-1}$)	Up to 0.0003–0.0007	Up to 0.005	Up to 0.01	Above 0.01
Bottom sediments ($\mu g kg^{-1}$)	Up to 1.0–3.0	Up to 20–30	Up to 100	Above 100
Higher water plants ($\mu g kg^{-1}$)	Up to 1.0–3.0	Up to 15–20	Up to 50	Above 50

The pollution of freshwater reservoirs with PAH raises the question of preventing such substances from reaching drinking-water supplies. It is important to remove tars and other products that contain PAH from the effluents coming from industrial plants and cities. Further, it is necessary to employ both biological methods of purification and filtration through activated charcoal. After these initial stages of purification, water must also be subjected to the action of chlorine or ozone.

Ilnitsky *et al.* (1968) concluded that chlorination of water is relatively ineffective as far as PAHs (especially BaP) are concerned. Comparison with the actions of ultraviolet light and ozone showed that the latter destroys BaP and a number of other PAHs many times more effectively and more completely than ultraviolet light which, in turn, is more effective than chlorination. Burleson, Caulfield and Pollard (1979) found that ozonization removes mutagenic and carcinogenic effects that are due to the presence of PAH and cyclic amines in water, as well as inactivating certain mutagenic pesticides.

3.4.2. Seas and oceans

The growing use of seas and oceans as a source of potable water (after distillation) emphasizes the need to consider their pollution with PAH. The pollution of sea and ocean waters emanates both from general sources (shipping, aviation, atmospheric particulates) and from specific or local sources (specific catastrophes or outflows from industrial plants engaged, for example, in coal conversion or in shale and oil refining). With regard to environmental catastrophes, a modern oil tanker in a limited sea area can deliver instantaneously several kilograms of BaP and other PAHs. The polluting effect of a shore industry can be clearly seen from estimations of BaP in sand, aquatic plants and other littoral organisms, and Gortalum (1972) found higher levels of pollution by BaP in the Baltic than in the Black Sea, in the region of Yevpatoriya. Andelman and Suess (1970) reported on BaP concentrations in marine plants and also found considerable quantities of BaP in fish, molluscs, oysters and mussels. According to Gortalum (1972), BaP accumulates in especially large quantities in jellyfish (over 71.7 $\mu g\,kg^{-1}$ dry weight) and this is thought to be due to PAH absorption from the sea water.

Ermakov (1979) took 300 samples from different seas in and around the USSR. Special attention was paid not only to the bottom layer and to water taken at some depth, but also to the so-called border layer. This is a film forming a border between two phases (atmosphere and hydrosphere) where different pollutants accumulate. BaP was found in varying quantities with considerable differences in mean pollution levels from different seas becoming evident (Table 9.11). The Bering Sea concentrations can be considered as background levels, and Baku Bay and the estuary of the River Purtse were found to be the two most polluted. This is undoubtedly due to the presence of the oil industry and its refineries and to the shale oil industry, respectively. The Bay of Finland is considerably more polluted than the central part of the Baltic Sea, as it is adjacent to industrial areas.

On the basis of the study it is possible to assess, semi-quantitatively, the different levels of marine pollution using BaP as an indicator of total PAH. Such an assessment is given in Table 9.12.

TABLE 9.11. *Mean concentration ($\mu g\,l^{-1}$) of BaP in seas in and around the USSR (Ermakov, 1979)*

Object of study	Water	Surface layer	Bottom sediment
Bering Sea (central area)	0.025 ± 0.003	0.125 ± 0.003	–
Baltic Sea (central area)	0.051 ± 0.004	–	16.1 ± 1.17
Baltic Sea (Bay of Finland)	0.15 ± 0.01	2.24 ± 0.1	76.8 ± 3.25
Baltic Sea (estuary of the River Purtse)	0.27 ± 0.07	5.0 ± 0.25	7500 ± 125
Caspian Sea (Bay of Baku)	0.10 ± 0.02	10.6 ± 0.39	8030 ± 146

TABLE 9.12. *Pollution levels ($\mu g\,l^{-1}$) in the marine environment by BaP (Ermakov, 1979)*

Degree of pollution	Concentration of BaP in water	Concentration of BaP in the surface layer	Concentration of BaP in the bottom sediment
High	> 0.1	> 0.1	> 100
Average	0.01–0.1	0.1–0.1	20–100
"Background"	< 0.01	< 0.1	< 20

3.5. *Bacterial Degradation of PAH*

The decomposition of polycyclic aromatic hydrocarbons was first demonstrated many years ago by Tauson and Veselov (1973) and this has been confirmed more recently in work on the degradation of crude oil in sea water by micro-organisms (Zo Bell, 1969). Isolates of soil bacteria (*Bacterium megaterium* and *B. sphericus*) have proved capable of destroying BaP in soil by metabolizing it, and it has also been shown that this leads to a reduction in carcinogenic activity in mice. In soil, the seasonal variations in soil BaP content can be related to seasonal changes in the activity of soil micro-organisms. Cultures of *B. mutilate*, which actively break down BaP, act similarly on a whole range of polycyclic aromatic hydrocarbons.

In addition, bacteria isolated from sewage effluent, fresh-water systems and the sea have all been found to be capable of PAH degradation (Ermakov, 1979; Lembik, 1979; Shabad *et al.*, 1979).

4. Natural Origins of Polycyclic Aromatic Hydrocarbons

4.1. *Synthesis by Plants and Micro-organisms*

There has been some dispute as to whether or not plants and micro-organisms can synthesize PAH (Blumer, 1961; Gräf and Diehl, 1966). In order to exclude the possibility of exogenous PAH penetration, experiments were carried out in which a specially constructed, hermetically sealed chamber was used into which air was introduced after rigorous filtration. Every precaution was taken in germinating the seeds and in growing the resulting plants in the chamber to avoid contamination. The plants were harvested and subsequently analysed for BaP content. The results given in Table 9.13 seem to provide evidence that BaP is in fact formed.

TABLE 9.13. *Mean contents of benzo(a)pyrene with standard deviations, in three crop species, as $\mu g\,kg^{-1}$ (Ilnitsky et al., 1979)*

Crop	Initial concentration	Shoots	Roots
Wheat	1.58 ± 0.08	19.73 ± 2.79	8.44 ± 2.70
Rye	0.74 ± 0.07	8.01 ± 0.61	1.60 ± 0.10
Oats	1.27 ± 0.06	14.30 ± 0.78	5.05 ± 0.38

4.2. *Volcanic Activity*

Volcanic activity represents a powerful pyrogenic process and the production of natural PAH is a likely outcome. Ilnitsky, Belitsky and Shabad (1975) collected samples of fresh volcanic ash, of soil and of plants near volcanoes, and analysed them for BaP. The results are given in Table 9.14.

TABLE 9.14. *Benzo(a)pyrene content ($\mu g\,kg^{-1}$) in soil, vegetation and volcanic mud in the vicinity of volcanoes (Ilnitsky, Belitsky and Shabad, 1975)*

	Soil			Vegetation			Volcanic mud		
Location	1	2	3	1	2	3	1	2	3
Burlyaschy Volcano	11	1.1	0.4– 3.1	3	7.4	5.4– 9.8	–	–	–
Uzon Volcano	5	4.2	0.9– 7.1	10	3.6	0.7–11.7	12	2.8	0.5–7.5
Geyser Valley	5	1.3	0.3– 0.4	4	4.7	1.1– 8.4	–	–	–
Kliuchevskayasopka Volcano	13	3.4	0.4–14.6	6	7.4	1.4–16.4	–	–	–
Kurile islands: Kunashir	2	5.9	0.7–11.2	1	4.5	–	–	–	–

1 = no. of samples; 2 = mean content; 3 = confidence limits ($p = 0.05$).

Calculations on the annual input of BaP to the biosphere from volcanoes suggest that this amounts to only several tons. This appears to be an insignificant quantity in relation to man-made emissions which equal about 1300 tons $annum^{-1}$ in the USA (Selkirk, 1977) and about 5000 tons globally each year (Suess, 1976).

5. Polycyclic Aromatic Hydrocarbons and Coal Combustion and Conversion Processes

5.1. Coal Combustion

Junk and Ford (1980) give a list of 47 PAH compounds resulting from coal combustion residing in fly ash, grate ash or the stack emissions. 25 % of the compounds occur in all three. However, large, efficient coal-burning electricity generating plants, with high combustion temperatures, emit relatively low total amounts of PAH. Coal-burning residential units emit, in terms of gross heat input, at least three orders of magnitude more BaP than coal-burning power plants. On the same criteria, coal units are usually greater emitters of PAH than oil or natural gas units (Smith, 1984).

Coal-fired power plants contribute very little to PAH emissions when properly operated. For example, three Scandinavian coal-fired power plants had emissions of PAH less than $0.5\,\mu g\,kg^{-1}$ of coal charged. Table 9.15 summarizes emissions for power plants, industrial and commercial boilers using coal, oil, gas, peat or wood. A fluidized bed combustor is seen to give rise to large PAH emissions. The unit in question is said to have been under faulty operation although in this respect it is interesting that Robertson, Groth and Blasko (1980) report that PAH levels are higher in exhaust fumes from low sulphur fuels. Some workers have confirmed that POM emissions are higher from FBC and PFBC units than from conventional coal combustion (Hobbs *et al.*, 1980a, 1980b; Abel *et al.*, 1981). However, others have found similar emissions for FBC units compared with conventional coal-fired units of a similar size (Nielsen, 1983) and fluidized bed coal combustion units appear

TABLE 9.15. *Emission factors ($\mu g\ kg^{-1}$) for power and heat generation using different fuels (Ramdahl, Alfheim and Bjørseth, 1983a, 1983b)*

Facility	PAH		BaP	
	Representative value	range	Representative value	range
Pulverized coal-fired power plant	19	0.5–32	1.6	1.1–2.7
5 MW fluidized bed plant	3600	–	150	–
Coal-fired industrial boilers	41	–	0.93	–
Oil-fired industrial boilers	23	5.3–100	1.1	–
Oil-fired commercial boilers	820	–	40	–
2 MW gas-fired intermediate hot water boiler	–	490–1100	10	–
Peat-fired hot water boiler (2 MW)	15 000	–	–	–
Wood-fired hot water boiler (2 MW)	2000	1890–3390	22	10–36

only to be suspect during faulty operation (build up of slag in the bed and consequent temperature reduction) or during start-up or shut-down. Different firing methods in conventional coal-fired power plants give variations in particulate PAH emissions (Zelenski, Pangaro and Hall-Enos, 1980) of at least an order of magnitude.

5.2. *Coal Conversion*

5.2.1. *Coal gasification*

So far there is no information that coal gasification plants give rise to atmospheric emissions of PAH that are high enough to be a cause for concern. At the Lurgi coal gasifier at Kosovo in Yugoslavia (Josephson, 1980) PAH emissions to the atmosphere were said to be similar to those in "a typical urban American atmosphere". Of more concern are PAH residues in the tar and oil wastes (Bradley, Ferrell and Felder, 1983) but it must be remembered these are often recycled. PAH levels in aqueous wastes may include BaP at harmful concentrations (Gangwal, 1981; Gangwal, Cleland and Truesdale, 1981) and these will need careful treatment (Dark, 1981).

5.2.2. *Coal liquefaction*

Although PAH compounds have been detected in gaseous streams of coal liquefaction plants (Gasper and Rosenberg, 1981), these were estimated to be well below any harmful concentration. Solid and aqueous wastes are of more concern and a number of PAH compounds have been identified in solid wastes (Maskarinec *et al.*, 1982; Gasper and Rosenberg, 1981) and in aqueous wastes

(Shields *et al.*, 1979; Dileo and Nicholas, 1980; Woodbridge, Kim and Hanson, 1981; Robbins *et al.*, 1981; Gasper and Rosenberg, 1981; Gasamtverband, 1982). Coal liquefaction plants operate under pressure and control measures will need to be directed towards solid and aqueous wastes but phenolic compounds are regarded by some (Smith, 1984) as a greater hazard.

Palmer (1979) investigated the mortality experience of 50 workers with occupational exposures to the products of coal hydrogenation processes, but there is a paucity of epidemiological evidence in relation to the commercial operation of coal gasification and liquefaction plants because of the low number of fully-operational units. The information on health effects is discussed in Chapter 10.

6. Polycyclic Aromatic Hydrocarbon Body Burdens in Man

It is now well established that the effect of chemical carcinogens is directly related both to the dose and to the duration of contact with the carcinogenic agent. It is thus important to know the "body load" to which man is exposed during his lifetime. Man is exposed to PAH from the atmosphere, from food and from water. Over a 70-year lifespan, food intake of PAH is likely to be about 5 mg. Inhabitants of large cities will, in addition, gain 12–16 mg from inhalation (Shabad and Dikun, 1959). Intake of PAH in water will normally be extremely low. Much of the PAH in water will be excreted, and Shabad and Dikun (1959) found that the tissues of elderly town-dwellers contained 1000 times less BaP than might have been expected from intake figures. Linnik, Smirnov and Shabad (1971) showed that the activities of enzyme systems in the body lead to the formation of oxy-derivatives which are no longer carcinogenic in activity. Gorelova and Cherepanova (1970) confirmed this by carrying out experiments using cows, hens, ducks, geese and mice fed on food containing considerable quantities of BaP. They emphasized the suitability for human consumption of meat, milk and even eggs obtained from these animals, even though the food that had been fed to these animals was contaminated. The PAHs appear in general to be more readily detoxified and excreted by a variety of species than are some of the halogenated pesticides and it appears unlikely, therefore, that PAHs will accumulate in food chains.

There has been considerable effort applied to the determination of maximum allowable concentrations (MAC) of PAH in the atmosphere, since this represents the major exposure source to which man is subjected. BaP levels are often taken as indicators for the establishment of MAC values and the standard can be set by reference to background levels that are continuously monitored. In addition, experiments utilizing animals that have been subjected to different routes of BaP administration (dermal, oral, intratracheal) can give some useful indications. Thirdly, occupational exposures must be assessed. Standards set usually allow only 0.1 μg 100 m^{-3}. For higher figures, a 6-hr working day and period of extended leave are stipulated. Levels in water reservoirs are set at 0.005 $\mu g\, l^{-1}$ and levels in soil at 20 $\mu g\, kg^{-1}$.

It is not easy at this time to predict the levels of pollution by PAH that might result from increased coal combustion and the large-scale implementation of coal gasification and liquefaction operations. As these procedures become more widely used, monitoring of the working environment and of general environmental levels will have to be undertaken. The current standards can be taken as reasonable guidelines at the present time, but it must be borne in mind that any increase in dose and sustained contact may lead to increased disease and an increased incidence of malignant tumours. A comprehensive study involving adequate monitoring procedures will be required as a basis for the prophylactic measure that should accompany the increased utilization of coal.

7. References

ABEL, W. T., RICE, R. L., SHANG, J. Y., TUREK, D. G. and AYERS, W. J. (1981). *Combustion of Western Coal in a Fluidized Bed.* DOE/METC/R1-178, Morgantown Energy Technology Center, West Virginia.

ANDELMAN, J. B. and SUESS, M. J. (1970). Polynuclear aromatic hydrocarbons in the water environment. *Bulletin of the World Health Organisation,* **43,** 479–508.

BLUMER, M. (1961). Benzpyrene in soil. *Science,* **134,** 474–5.

BLUMER, W., JAUMANN, R. and REICH, T. (1972). Motorisierungwichtigste Ursache für Krebsentstehung? *Schweizerische Rundschau fuer Medizin (Praxis),* **61,** 514–8.

BORNEFF, I. and FABIAN, B. (1963). Kanzerogene Substanzen in wasser und boden XIX. Weitere untersuchungen über polyzyklische aromatische kohlenwasserstoffe in erdcproben. *Archiv für Hygiene und Bakteriologie,* **147,** 401–12.

BRADLEY, A. A., FERRELL, J. K. and FELDER, R. M. (1983). Polycyclic aromatic hydrocarbons in the condensed phases associated with coal gasification. In *Polynuclear Aromatic Hydrocarbons: Formation, Metabolism and Measurement* (ed. by M. Cooke and A. J. Dennis). Battelle, USA.

BRASSER, L. J. (1979). PAH concentrations in the Netherlands. *Polycyclic Aromatic Hydrocarbons.* Colloquium Summaries. VDI, Kommission Reinhaltung der Luft.

BURLESON, G. R., CAULFIELD, M. J. and POLLARD, M. (1979). Ozonation of mutagenic and carcinogenic polyaromatic amines and polyaromatic hydrocarbons in water. *Cancer Research,* **39,** 2149–54.

BUTLER, J. D. (1981). Author's reply—Reactivity of polycyclic aromatic hydrocarbons adsorbed on soot particles. *Atmospheric Environment,* **15,** 1783–4.

BUTLER, J. D. and CROSSLEY, P. (1981). Reactivity of polycyclic aromatic hydrocarbons adsorbed on soot particles. *Atmospheric Environment,* **15,** 91–4.

CANDELI, A., MASTRANDREA, V., MOROZZI, G. and TOCCACELI, S. (1974). Carcinogenic air pollutants in the exhaust from a European car operating on various fuels. *Atmospheric Environment,* **8,** 693–705.

COOK, J. W., HEWITT, C. L. and HIEGER, I. (1933). The isolation of a cancer producing hydrocarbon from coal-tar. *Journal of the Chemical Society,* **1,** 395–405.

COOK, J. W., HIEGER, I., KENNAWAY, E. L. and MAYNEORD, W. V. (1932). The production of cancer by pure hydrocarbons. *Proceedings of the Royal Society,* **B111,** 455–84.

DARK, W. A. (1981). Determination of aromatic hydrocarbons in biologically treated water from a coal gasification process. In *Analysis of Waters Associated With Alternative Fuel Production.* Proc. of Symp., Philadelphia, Pa. American Society for Testing and Materials, USA.

DILEO, A. J. and NICHOLAS, D. M. (1980). Characterizing wastewater and monitoring its treatment in SRC-I. *Quarterly Technical Report, October–December, 1980.* International Coal Refining Company, Allentown.

EGAN, H., CASTERGNARO, M., BOGOVSKI, P., KUNTE, H., WALKER, E. A. and DAVIS, W. (eds.) (1979). *Environmental Carcinogens: Selected Methods of Analysis*, vol. 3, *Analysis of Polycyclic Aromatic Hydrocarbons in Environmental Samples*. IARC Publications no. 39. Lyon.

ERMAKOV, E. A. (1979). Certain data on the distribution of carcinogenic hydrocarbons in the bottom sediment, the surface layer of seas and the atmosphere. *Carcinogenic Substances in the Environment*, pp. 43–8. Gydrometeoizdat, Moscow.

FOX, A. S., LINDARS, O. C. and OWEN, P. A. (1974). Survey of occupational cancer in the rubber and cable making industries: results of five-year analysis, 1967–1971. *British Journal of Industrial Medicine*, **31**, 140–51.

FRITZ, W., ENGST, R., SHABAD, Z. M., DIKUN, P. P. and KHESINA, A. Ya. (1977). Vergleichende Untersuchungen zur kontamination von lebensmitteln met krebserzengenden kohlenwasserstoffen in der UdSSR und der DDR. *Arch. Geschwulstforsch.* **46/7**, 561–6.

GANGWAL, S. K. (1981). Gas chromatographic investigation of raw wastewater from coal gasification. *Journal of Chromatography*, **204**, 439–44.

GANGWAL, S. K., CLELAND, J. G. and TRUESDALE, R. S. (1981). *Evaluation of Relative Environmental Hazards from a Coal Gasifier*. Final Report PB-81-217648. Research Triangle Institute, North Carolina.

GASPER, J. R. and ROSENBERG, S. E. (1981). *Public Health Hazards of Lurgi/Fischer-Tropsch Coal Liquefaction*. ANL/EES-TM-153, Argonne National Laboratory, Argonne.

GERSAMTVERBAND (1982). *Umweltvertraeglichkeit von Anlagen zur Vergasung und Verfluessigung von Steinkohle in der Bundesrepublik Deutschland*. Gersamtverband des deutschen Steinkohlenbergbaus, Essen.

GOFMEKLER, V. A., MANITA, M. D., MANUSADZHANTS, Zh. I. and STEPANOV, Z. Z. (1963). The correlation between the concentration of 3,4-benzpyrene and carbon monoxide in exhaust gases of automobiles. *Gygiena i Sanitariya*, **8**, 3–8.

GORELOVA, N. D. and CHEREPANOVA, A. I. (1970). On benzo(*a*)pyrene in cow's tissue and milk after its administration by fodder. *Voprosy Onkologii*, **16**, 659–73.

GORTALUM, G. M. (1972). The circulation of polycyclic aromatic hydrocarbons in the marine environment. In *Investigations into the Prophylactic Treatment of Pollution of Man's Environment by Carcinogenic Substances*, pp. 30–32. Tallin.

GRÄF, W. and DIEHL, H. (1966). Über den naturbedingten normalpegel kenzerogener polyzyklischer aromatic und seine ursache. *Archiv für Hygiene und Bakteriologie*, **150**, 49–59.

GRIMMER, G. (1979). Messtechnische Erfassung von PAH durch Profil-Analyse. *Umwelt*, **2**, 123–5.

GRIMMER, G. and BOHNKE, H. (1979). Gas chromatographic profile analysis of polycyclic aromatic hydrocarbons in (I) high protein foods, (II) fats and vegetable oils and (III) plants, soils and sewage sludge. In *Environmental Carcinogens: Selected Methods of Analysis*, vol. 3, *Analysis of Polycyclic Aromatic Hydrocarbons in Environmental Samples* (ed. by Egan, H., Castegnaro, M., Bogovski, P., Kunte, H., Walker, E. A. and Davis, W.). IARC Publications no. 29, pp. 63–173. Lyon.

GROVER, P. L. (1980). The metabolic activation of the polycyclic aromatic hydrocarbons, *VDI Berichte*, **358**, 257–71.

GURINOV, B. P., TUGARINOVA, V. N. and VASILIEVA, O. I. (1962). Carcinogenic properties of exhaust soot of motortransport. *Gygiena i Sanitariya*, **2**, 19–24.

HOBBS, C. H., CARPENTER, R. L., NEWTON, G. J., CLARKE, C. R., HENDERSON, R. F., HANSON, R. L., WEISSMAN, S. H., GRIFFIS, L. C. and HILL, J. O. (1980a). Physical, chemical and biological characterization of atmospheric pressure fluidized bed coal combustion effluents. In *Proceedings of the 6th International Conference on Fluidized Bed Combustion, Volume III*. Department of Energy, Washington, D.C.

HOBBS, C. H., CARPENTER, R. L., NEWTON, G. J., KOVACH, J. J., HANSON, R. L., WEISSMAN, S. H., CLARK, C. R., HILL, J. O. and SHANG, J. Y. (1980b). Characterization of effluents from an experimental atmospheric pressure fluidized bed combustor. In *Proceedings of the Second U.S. Department of Energy Environmental Control Symposium, Volume I. Fossil Energy*. Department of Energy, Washington, D.C.

HUENIGEN, E., PRIETSCH, V., NAUMANN, M., JASKULLA, N., SHABAD, L. M., KHESINA, A. Y. and SMIRNOV, G. A. (1978). Reduction of air pollution caused by carcinogenic substances from exhaust gases of automobiles. In *Atmospheric Protection*. The co-ordinating centre on the problems of atmospheric protection from pollution by harmful substances. Dresden, GDR.

Ilnitksy, A. P., Belitsky, G. A. and Shabad, L. M. (1975). Carcinogenic polycyclic aromatic hydrocarbon benzo(*a*)pyrene in volcano exhausts. *Seriia Biologicheskaya, Dokladi Akademii Nauk USSR*, **255**, 214–6.

Ilnitksy, A. P., Khesina, A. Ya., Cherkinsky, S. N. and Shabad, L. M. (1968). The influence of ozonation of the aromatic carcinogenic hydrocarbons. *Gygiena i Sanitariya*, **3**, 8–11.

Ilnitsky, A. P., Mischenko, V. S. and Shabad, L. M. (1979). The natural sources of carcinogenic hydrocarbons. In *Carcinogenic Substances in the Environment*, pp. 25–9. Gydrometeoizdat, Moscow.

Ilnitsky, A. P., Voronin, V. I. and Solenova, L. G. (1974). Carcinogenic effects of benzo(*a*)pyrene and its combination with other hydrocarbons during their oral intake. *Voprosy Onkologii*, **20**(10), 71–5.

Josephson, J. (1980). Toxic by-products of coal conversion. *Environmental Science and Technology*, **14**, 1283–6.

Junk, G. A. and Ford, C. S. (1980). *A Review of Organic Emissions from Selected Combustion Processes*. Report IS-4727, Ames Laboratory, Ames.

Khesina, A., Ya. (1979). Determination of benzo(*a*)pyrene in extracts by spectroluminescence. In *Environmental Carcinogens: Selected Methods of Analysis*, vol. 3, *Analysis of Polycyclic Aromatic Hydrocarbons in Environmental Samples* (ed. by Egan, H., Castegnaro, M., Bogovski, P., Kunte, H., Walker, E. A. and Davis, W.). IARC Publications no. 29, pp. 215–9. Lyon.

Klipinitser, I. L., Ninin, V. K., Kuznetsova, T. A., Shabad, L. M., Khesina, A. Y. and Serkovskaya, G. S. (1977). An investigation of benzo(*a*)pyrene in the products of the anthracene fraction of coal tar. *Gygiena Truda i Profzabolevanyi*, **3**, 47–9.

Konstantinov, V. G., Somakhina, P. G., Gotlieb, E. V. and Kuzminykh, A. I. (1974). On the question of carcinogenic danger in aluminium electrolysis workshops. In *Occupational Cancer*, pp. 87–91. Moscow.

Korfmacher, W. A., Natusch, D. F. S., Taylor, D. R., Mamantor, G. and Whery, E. L. (1980). Oxidative transformations of polycyclic aromatic hydrocarbons absorbed on coal fly ash. *Science*, **207**, 763–5.

Lawther, P. J. and Waller, R. (1976). Coal fires, industrial emissions and motor vehicles as a source of environmental carcinogens. *Environmental Pollution and Carcinogenic Risks* pp. 27–40 (ed. by Rosenfeld, C. and Davis,,W.). INSERM Symposium Series, 52, IARC Scientific Publications no. 13, Paris.

Lembik, J. L. (1979). On certain natural factors of benzo(*a*)pyrene destruction in fresh water reservoirs. *Carcinogenic Substances in the Environment*. Gydrometeoizdat, Moscow.

Linnik, A. B., Smirnov, G. A. and Shabad, L. M. (1971). The study of the carcinogenic activity of soot from aviation engines in experiments on animals. *Bulletin Experimentalnoy Biologhii i Meditsinii*, **2**, 83–7.

Mallet, L. and Heros, M. (1962). Pollution des terres vegetales par les hydrocarbures polybenzeniques due type benzo-3,3-pyrene. *Compte Rendu Hebdomadaire des Seances de l'Academie des Sciences, Paris*, **245**, 958–60.

Maskarinec, M, P., Brazell, R. S., Harvey, R. W. and Brown, D. K. (1982). Determination of selected organics in treated sludges and associated lechates from coal conversion facilities. *Analytica Chimica Acta*, **139**, 257–66.

NAS (1972). *Particulate Polycyclic Organic Matter*. National Academy of Sciences, Washington, D.C.

Nielsen, P. (1983). PAH-emission from a 4.7 MW coal-fired fluidized-bed plant. In *Polynuclear Aromatic Hydrocarbons: Formation, Metabolism and Measurement* (ed. by M. Cooke and A. J. Dennis). Battelle, USA.

Nikolaou, K., Masclet, P. and Mouvier, G. (1984). Sources and chemical reactivity of polynuclear aromatic hydrocarbons in the atmosphere—a critical review. *The Science of the Total Environment*, **32**, 103–32.

Palmer, A. (1979). Mortality experience of 50 workers with occupational exposures to the products of coal hydrogenation processes. *Journal of Occupational Medicine*, **21**, 21–4.

Petrov, N. I. (1961). *Manual of General Oncology*. Medghiz, Leningrad.

Pitts, J. N. and Katzenstein, Y. A. (1981). Reactivity of polycyclic aromatic hydrocarbons adsorbed on soot particles. *Atmospheric Environment*, **15**, 1782–3.

Pullman, A. and Pullman, B. (1955). Electronic structure and carcinogenic activity of aromatic molecules. *Advances in Cancer Research*, **3**, 117–69.

RAMDAHL, T., ALFHEIM, I. and BJØRSETH, A. (1983a). Sources and emission factors of polycyclic aromatic hydrocarbons. In *Polycyclic Aromatic Hydrocarbons in the Air*. OECD, Paris.

RAMDAHL, T., ALFHEIM, I. and BJØRSETH, A. (1983b). PAH emission from various sources and their evolution over the last decades. In *Polycyclic Organic Matter from Exhaust Gases* (ed. by D. Rhondia and M. Cooke). Reidel, Dordrecht.

ROBBINS, W. K., SEARL, T. D., WASSERSTROM, D. H. and BOYER, G. T. (1981). Determination of polynuclear aromatic hydrocarbons in wastewater from coal liquefaction processes by the gas chromatography-ultraviolet spectrometry technique. In *Analysis of Waters Associated with Alternative Fuel Production*. American Society for Testing and Materials, Pittsburgh.

ROBERTSON, D. J., GROTH, R. H. and BLASKO, T. J. (1980). Organic content of particulate matter in turbine engine exhaust. *Journal of the Air Pollution Control Association*, **30**, 261–6.

SELKIRK, J. K. (1977). Benzo(*a*)pyrene cancero-genesis: a biochemical selection mechanism. *Journal of Toxicology and Environmental Health*, **2**, 1245–58.

SHABAD, L. M. (1973). *Circulation of Carcinogens in the Environment*. Meditsina, Moscow.

SHABAD, L. M. (1978). The International Symposium of air pollution and cancer in Stockholm, 1977. *Gygiena i Sanitariya*, **2**, 121–2.

SHABAD, L. M. and DIKUN, P. P. (1959). *Air Pollution by Carcinogenic Substances: 3,4-Benzpyrene*. Medghiz, Leningrad.

SHABAD, L. M., KHESINA, A. YA. and KHITROVO, S. S. (1968). On carcinogenic hydrocarbons in the exhaust gases of motor car engines and their possible elimination. *Vestnik Akademii Meditsinski Nauk USSR*, **23**(6), 12–20.

SHABAD, L. M., KHESINA, A. YA., LINNIK, A. B. and SERKOVSKAYA, G. S. (1970). Possible carcinogenic hazards of several tars and of Locacorten-tar ointment: spectro-fluorescent investigations and experiments on animals. *International Journal of Cancer*, **6**, 314–8.

SHABAD, L. M., KHESINA, A. Y., SERVOSKAYA, G. S., OGLOBLINA, A. I. and FRIDMAN, Y. S. (1972). Determination of 3,5-benzpyrene in protein fodder and in raw material employed during its obtention. In *Medicobiological Investigations of Hydrocarbon Yeasts* (ed. by Pokrovskii, A. A.). Nauka, Moscow.

SHABAD, L. M., KHESINA, A. YA., TSIBAN, A. V., PANOV, G. V. and MIROSCHNICKENKO, N. M. (1979). Microbial degradation of carcinogenic polycyclic hydrocarbons in the marine environment. *Proceedings of the 14th Pacific Ocean Scientific Congress*, Khabarovsk.

SHABAD, L. M. and SMIRNOV, G. A. (1976). Aviation and environmental benzo(*a*)pyrene pollution. *INSERM*, **52**, 53–60.

SHIELDS, K. J., HOPKINS, H. T., WEIR, E. E. and THOMPSON, C. (1979); *Environmental Assessment Report: Solvent Refined Coal (SRC) Systems*. EPA-600/7-79-146, Industrial Environmental Research laboratory, Research Triangle Park, N.C.

SIMS, P. and GROVER, P. L. (1974). Epoxides in polycyclic aromatic hydrocarbon metabolism and carcinogenesis. *Advances in Cancer Research*, **20**, 165–274.

SMITH, I. (1984). *PAH from Coal Utilization–Emissions and Effects*. Report no. ICTIS/TR 29, IEA Coal Research, London.

SUESS, M. J. (1976). The environmental load and cycle of polycyclic aromatic hydrocarbons. *Science of the Total Environment*, **6**, 239–50.

TAUSON, V. O. and VESELOV, I. (1973). On the bacterial degradation of cyclical compounds by sulphate reduction. *Mikrobiologiya*, **3**, 360–9.

TEUTSCHLAENDER, O. (1929). Uber den Pechkrebs der briket-arbeiter auf frund von fabrikseuchen in Baden und Sud Wales. *Zeitschrift Krebsforschung*, **28**, 283–300.

VARSHAVSKY, I. L., DOZENKO, V. I., IGNATOVICH, N. I. and KLYATSKIN, V. B. (1970). On methods of investigation of the content of carcinogenic hydrocarbons in the exhaust gases of auto-transport. *Gygiena i Sanitariya*, **4**, 75–7.

WALLER, R. (1952). The benzpyrene content of town air. *British Journal of Cancer*, **6**(1), 8–21.

WOODBRIDGE, D. D., KIM, J. I. and HANSON, D. M. (1981). Chemical and biological characteristics of waste streams of the SRC-II pilot plant. In *Coal Conversion and the Environment: Chemical Biomedical and Ecological Considerations* (ed. by D. D. Mahlem, R. H. Gray and W. D. Felix). US Department of Energy, Technical Information Center, Oak Ridge, Tennessee.

YAMAGIWA, K. and ICHIKAWA, K. (1918). The experimental study of the pathogenesis of carcinomas. *Cancer Research*, **3**, 1–29.

YANISHEVA, N. YA., CHERNICHENKO, I. A. and BALENKO, N. V. (1977). *Carcinogenic Substances and their Hygienic Control in the Environment*. Kiev.

YANISHEVA, N. YA. and KIREEVA, I. S. (1977). On the hygienic assessment of elements of the environment participating in the formation of the total dose of benzo(*a*)pyrene entering the human body. *Gygiena Naselënnoy Mestnosti*, **16**, 28–32.

YANISHEVA, N. Ya. and KIREEVA, I. S. (1979). Determination of benzo(*a*)pyrene in air using quasi linear luminescence. In *Environmental Carcinogens: Selected Methods of Analysis*, vol. 3, *Analysis of Polycyclic Aromatic Hydrocarbons in Environmental Samples* (ed. by Egan, H., Castegnaro, M., Bogovski, P., Kunte, H., Walker, E. A. and Davis, W.). IARC Publications no. 29, pp. 63–173. Lyon.

YANISHEVA, N. Ya. (1939). On the comprehensive approach to the study of benzo(*a*)pyrene in the environment. In *Problemy Hygieny*, pp. 107–18 (ed. by Gorski, T. and Pliss G.). Society of Hygienists of Poland, Warsaw.

ZABEZHENSKY, M. A. (1968). City air pollution with carcinogenic substances released by motorcar engines. *Voprosy Onkologii*, **41**(1), 87–8.

ZDRAZIL, I. and PICHA, F. (1966). The occurrence of the carcinogenic compounds 3,4-benzpyrene and arsenic in soil. *Neoplasma*, **13**, 49–55.

ZELENSKI, S. G., PANGARO, N. and HALL-ENOS, J. M. (1980). *Inventory of Organic Emissions from Fossil Fuel Combustion for Power Generation*. Report EPRI-EA-1394, GCA Corporation, Bedford, MA.

ZO BELL, C. E. (1969). Microbiological modification of crude oil in the sea. *Proceedings of the Conference of Prevention and Control of Oil Spills*. PAI Publications.

10

Occupational and Environmental Health Problems from Coal Utilization and Conversion

1. Introduction 245
2. Coal Combustion 246
3. Coal Conversion 248
    3.1. Introduction 248
    3.2. Occupational Health 250
        3.2.1. Plant design and work environment standards 250
        3.2.2. Hazard evaluation 257
        3.2.3. Process data 262
    3.3. Communities in the Plant Vicinity 269
    3.4. Product Users 276
4. Control and Planning 276
5. References 279

1. Introduction

Health risks from coal arise from all stages of the coal fuel-cycle: prospecting, mining, beneficiation, transport, storage and utilization (direct combustion or following conversion to liquid or gaseous fuel). Health risks are part of the general environmental impacts that occur to natural, semi-natural and man-managed environmental systems. These include long-term and far-reaching impacts such as the effect the global emissions of carbon dioxide from coal burning will have on ambient temperatures and hence climate; the impacts of a local and regional nature arising from sulphur and nitrogen oxides emissions. Both will affect natural ecosystems (oceans, lakes and terrestrial ecosystems) and semi-natural ones such as grassland and forests as well as man-managed agricultural, horticultural and silvicultural ones. These impacts are considered in Chapter 11.

Health effects due to the coal fuel-cycle are experienced by human populations either as part of the workforce (occupational health risks) or as the general public (public health risks). Whereas it is usually a relatively simple matter to determine the size of the working population that is at risk, it is less easy to estimate with any accuracy the size of the exposed general population that runs the risk of some degree of hazardous exposure. Exposure commitment models are discussed in Chapter 7 and an attempt is made at comparative risk assessment of health effects of the coal and nuclear fuel-cycles. The impacts of trace elements and hydrocarbon emissions are considered specifically in Chapters 8 and 9 respectively. This chapter looks at the potential health problems that the adoption of coal conversion technologies may engender. In spite of the limited experience and information available in this field, it is prudent to explore this topic in order to gain insights into the factors that need to be monitored, the control technologies that may require incorporation into plant design and the environmental standards that will need to be established. This is done against a background of the general health effects of present coal use technologies.

2. Coal Combustion

Coal combustion may result in a wide range of substances being emitted to the atmosphere: particulates, SO_2, NO_x, CO, polycyclic organic matter (POM) and trace metals. Many of these are potential human health hazards. The air pollution health disasters that occurred up to the 1950s, such as those in the Meuse Valley, Belgium, Donora, Pennsylvania, and the city of London, were associated with episodes of heavy atmospheric loading of particulate matter and sulphur oxides in combination. These episodes led to atmospheric pollution control activity that included the use of clean fuels and/or higher combustion temperatures, the installation of better emission control equipment, the construction of better chimneys, the use of an intermittent episode control strategy and better land-use planning. Thus *acute exposure*, that in the past led to many excess deaths over a short time periods, is now of less interest. Concern has switched to *chronic exposure* and the difficult task of estimating the number of deaths in future years, and the spectrum of morbidity, that is attributable to present low level pollution exposure. This exposure is characterized by being the result of emissions from many remote sources, long-range transport from these, combining to produce exposure of populations even thousands of miles distant from the point sources.

Estimating exposure commitment under such conditions is subject to many difficulties. These include the complication due to chemical transformations that occur during transport in the atmosphere over such long distances. Thus NO_2 emissions may contribute to ozone exposure at distant sites. There is also the question of threshold doses to a population that undoubtedly will include a wide variation in sensitivity to low level doses. Synergistic effects from mixtures of atmospheric pollutants must be considered. The existence of long lag periods between exposure and effect makes relationships difficult to detect.

Attempts to derive an atmospheric sulphate-health damage function (Morgan *et al.*, 1978) utilized indications from animal toxicology, epidemiology and correlation analyses (Amdur, 1976; EPA, 1974; Lave and Seskin, 1977) that suggested sulphates were the harmful agents in a sulphate–

particulates mix and that the linear no threshold model (LNMT) held for increased mortality rates with increased exposure to sulphate. It was thought that chronic exposure increased the incidence of acute respiratory infection in children and this, in turn, led to an increased incidence of chronic heart and respiratory disease in adults and thus a higher eventual rate of mortality. The shortening of the life-span in deaths so attributable was put at 5–15 years.

Relationships between sulphate exposure and human health risk have recently been reviewed (Morris, 1983; Fischer *et al.*, 1983; Hamilton, 1984). They point out that the epidemiology linking health damage to exposure to sulphate has come into question (Committee of Science and Technology, 1976). Recent chemical work does not support effects at existing levels of exposure (Avol *et al.*, 1979; Schlesinger, Lipman and Albert, 1978; Wolff *et al.* 1979). The frequent $(NH_4)_2SO_4$ is of lower potency than the free H_2SO_4 aerosol that occurs at much lower levels. It is now suggested that a better index for estimating air pollution health effects may be a fine-particle health damage function (Özkaynak *et al.*, 1985). Sulphates make up a high proportion of fine particles and sulphate measurement gives the best correlation with overall fine particle concentration than any other yet available.

Health damage functions for NO_2 are the next most important in relation to coal combustion (CO is generally only of local importance). NO_2, being an oxidant, acts quite differently from the reducing sulphur–particulate mix. It is not highly correlated with sulphate incidence. Hamilton (1984) discussed in detail health damage functions in a comparative way (coal vs. oil, natural gas, wood), public vs. occupational exposure, the contribution from electricity generation, industrial-commercial sources, residential heating contribution. Some of the preliminary estimates given in that work are shown in Table 10.1 for sulphate and NO_2 health effects and in Table 10.2 various end uses of coal.

TABLE 10.1. *Upper estimates of health effects in the USA mortality (GW(e)yr output) due to sulphates and NO_2 emissions (Fischer et al., 1983)*

	Electricity generation	Industrial/ commercial	Residential heating
Sulphates:			
Coal	140	330	87
Oil	120	14	0.60
Natural gas	0.15	0.06	0.039
Wood	22	11	4.8
NO_2:			
Coal	130	54	36
Oil	63	16	9
Natural gas	150	19	19
Wood	42	22	13

TABLE 10.2. *Health effects (fatalities per GW(e)yr) for electricity generation, industrial or commercial use or residential heating in the USA (Fischer et al., 1983)*

	Occupational disease	Occupational injury	Public injury
Electricity-generation:			
Coal	0.26–0.63[a]	0.84–1.6	0.33–0.57
Oil	–	0.11–0.51	–
Natural gas	–	0.05–0.19	–
Wood[b]	–	9.0	0.21
Industrial/Commercial:			
Coal	0.12–0.28	0.30–0.96	0.14–0.24
Oil	–	0.01–0.42	–
Gas	–	0.33–0.093	–
Wood	–	4.2	0.12
Residential uses:			
Coal	0.16–0.39	0.19–1.4	0.19–0.33
Oil	–	0.036–0.13	–
Gas	–	0.020–0.087	–
Wood	–	6.0	6.0

[a] Ranges given are 95% confidence limits.
[b] Hand-cutting of lumber; unrealistic for some uses.

3. Coal Conversion

3.1. Introduction

Because the coal conversion industry is in its infancy there are few occupational or environmental health studies available. Apart from early studies of gas workers (Doll *et al.*, 1965, 1972), there is one detailed study of the health hazards of coal hydrogenation (Sexton, 1960a, 1960b; Ketchum and Norton, 1960; Weil and Condra, 1960). A group of 359 workers was examined for skin cancer over a period of 5 years (1955–60) at a coal liquefaction plant in West Virginia, USA, designed to process 273 t day^{-1} of incoming coal. Of this group, ten men developed skin cancer. Occupational exposure varied from several months to 23 years, although these longer exposures did not all accrue from work in the liquefaction plant, but all significant lesions were found in workers with less than 10 years' exposure. The reported incidence was 16–37 times the incidence of skin cancer expected in the chemical industry. Precancerous lesions were also found in the skin of forty additional workers (Sexton, 1960b).

Analyses of the liquid materials produced in the plant identified a number of carcinogenic materials. An increase in carcinogenicity with increasing liquid boiling point, usually with respect to liquids with boiling points > 260°C, was observed in laboratory animals (Weil and Condra, 1960). This relationship was particularly evident for the oil, produced in the plant, which was recycled to slurry the incoming coal. In addition, air measurements in working areas revealed benzo(*a*)pyrene concentrations often in excess of 0.05 mg m^{-3} with a peak value of 18 mg m^{-3} (Ketchum and Norton, 1960). Benzo(*a*)pyrene has been identified as strongly carcinogenic (Chapter 9). These concentrations are in contrast to urban levels of 0.003–0.006 μg m^{-3} in the USA (NAS, 1972). However, higher concentrations have been quoted for main roads in urban areas elsewhere (Shabad and Dikun, 1959; Shabad, Khesina and Khitrovo, 1968).

In 1977 a follow-up study was instituted to identify and trace the original fifty workers with skin lesions (Palmer, 1979). After a latency period of 18–20 years, the cancer incidence did not appear to be marked. It is well to note that none of the deaths had been autopsied (NIOSH, 1978a). It was not intended that the review by Palmer (1979) should be a basis for drawing firm conclusions about cancer incidence save for the highly selected group studied. An epidemiological study was recommended to identify the remainder of the work-force who were exposed to the coal hydrogenation plant discharges but who did not develop skin cancer in the 1955–60 interval. The possibility exists that some of the workers who had not developed skin lesions during that period may have developed more serious organ cancers after the surveillance programme was discontinued in 1960 (Palmer, 1970). It has also been reported that since the end of December 1958 the workers have been followed and that at first there were one or two new cases of skin cancer every year, but after the plant stopped operating the incidence was about one new case every 5 years (NIOSH, 1978b).

Mortality or morbidity data at other coal hydrogenation facilities have not been systematically collected or analysed, at least in the published literature. In place of such specific health data, regulatory agencies in the USA have supported testing programmes in existing plants to determine which hazardous materials are formed and need to be controlled (Lee *et al.*, 1981; Page, 1978; Murin, Sipes and Page, 1980). Even these data tend to be limited by confidentiality restrictions imposed by governments or individual companies (see, for example, the limitations noted by Clausen and Zee, 1981). A very complete set of measurements has recently become available from Lurgi plant at Kosovo in Yugoslavia (Lee *et al.*, 1981) and somewhat more limited information from the Texaco process (Konkol *et al.*, 1982; Schlinger and Richter, 1980). Also, as an aid in evaluating the potential toxicity of various process streams, the results of a number of biological screening tests on plant liquids and solids have been reported. These have included mutagenic tests on bacterial systems (for example Ames testing); *in vitro* cytotoxicity assays (for example, with Chinese hamster ovary or rabbit alveolar macrophage cells); and *in vivo* mammalian tests on rats, rabbits and guinea pigs (Lee *et al.*, 1981; Cowser *et al.*, 1981). The relationship of such biological tests to human toxicity is, however, still not well defined.

Process emission data and biological screening information which has become available recently is reviewed here. A summary of earlier work was made by Wadden and Trabert (1981). Also discussed are some of the approaches which have been proposed for environmental and occupational health protection.

3.2. *Occupational Health*

3.2.1. *Plant design and work environment standards*

Although synthetic gas and liquefaction plants have been operating in Europe and South Africa for many years, there is a dearth of health information on workers and other exposed groups. Apparently there have been no systematic studies of worker health, nor has there been emphasis on gathering information which would lead to such studies. A case in point, the SASOL I Lurgi coal liquefaction plant in South Africa, has been operating for over 23 years and employs approximately 5600 workers. The plant doctor has indicated that skin cancer has not been observed among the gasifier and gas clean-up crews as had been reported for a pilot plant in the USA (Sexton, 1960b). However, record keeping does not include chronic diseases such as cancer, and worker follow-up studies have not been undertaken. In addition, the quality of health surveillance differs considerably between white and black workers; to date, no workplace monitoring has been reported (Harris *et al.*, 1980). Many coal conversion projects have multinational support. For instance, SASOL was involved in the design of the Great Plains plant in North Dakota (Kuhn, 1982). The lack of information on historical exposure and health effects is therefore a difficult problem for public agencies responsible for control.

In general, there is recognition that air, water and solid waste emissions from coal conversion plants are qualitatively different from those associated with petroleum refining or other sectors of the chemical industry. Existing data indicate the presence of phenols and cyanides in waste water streams; ammonia, hydrogen cyanide, hydrogen sulphide, various mercaptans, and polycyclic hydrocarbons in gas streams; heavy metals in solid wastes; and polycyclic hydrocarbons in retained tar and oils (Lee *et al.*, 1981; Cowser *et al.*, 1981; Schlinger and Richter, 1980). Commercially viable plants may approach the rate of coal use of some larger power plants (14,000 tons day^{-1} for the Great Plains project) and exceed coke ovens or other coal handling processes. This emphasizes the need for careful control of hazards.

In the USA the approach has been to require coal processing plants to meet existing air, water and solid waste quality standards. In-plant exposures would be limited to TVL-threshold limit value (ACGIH, 1980) although this alone would not guarantee zero health risk. Furthermore, not all potentially hazardous discharges are covered under these standards. Consequently, various performance standards have been recommended for operation and occupational health monitoring in coal gasification (NIOSH, 1978a, 1978b) and liquefaction plants (Harris *et al.*, 1980; NIOSH, 1981). Release of contaminants outside the plant grounds would be covered under a variety of regulations, some of which are summarized in Table 10.3. In addition, air emission guidelines for sulphur and hydrocarbons have been suggested for coal gasification plants by the Environmental Protection Agency (EPA) and are shown in Tables 10.4 and 10.5 (Gibson and Page, 1979).

At present there are guidelines for health and safety maintenance in coal processing plants (NIOSH, 1978a). However, few maintenance plans are reported before plants begin operation. A notable exception is the approach to health and environmental evaluation adopted by the Tennessee Valley Authority (TVA) at their coal-to-ammonia plant which recently started up and which uses the Texaco process (HP, 1982). Table 10.6 summarizes their plans for environmental monitoring. In particular, the medical surveillance and industrial hygiene programmes recognize the need to "... gain knowledge to answer the persistent questions concerning the health and safety of workers exposed to the coal gasification environment in general", as well as to protect the workers at this specific plant (Williamson, 1981). The medical programme requires a preplacement examination of candidate workers to determine initial health conditions which might be affected by work in the coal gasification plant. Particular emphasis is placed on the condition of the skin, respiratory system and genitourinary tract. Colour photographs were made of the exposed skin of hands, face, neck and any suspicious lesions. Periodic, 12-month, examinations are planned, as well as follow-up examinations of former employees, in order to develop health statistics appropriate for a continuing epidemiological study (Williamson, 1981).

Various hazard rating schemes have also been developed to evaluate the relative toxicity of individual process streams. One approach is the EPA's Source Analysis Model (SAM) which is based on multi-media environmental goals (MEGs) (Cleland and Kingsbury, 1977; Kingsbury and White, 1979). However, the toxicological and human health effect basis for such measures is quite unclear as well as the applicability of the rating scheme (Hogan, 1982).

An experimental approach of a similar type is to test the effects of process fluids on bacterial and animal systems. The Ames test for mutagenicity is based on the ability of mutant strains of the bacteria *Salmonella typhimurium* to revert from histidine dependence to prototrophy (the ability to synthesize histidine) upon exposure to various mutagens and carcinogens. Results are reported as the number of revertants per plate adjusted for controls. Cytotoxic assays use mammalian cells in culture to measure cell death or metabolic impairment upon exposure to toxic materials. Commonly used cell systems are rabbit alveolar macrophages (RAM) and Chinese hamster ovary fibroblasts (CHO). Results are generally reported as effective concentrations of the toxic material (or stream) at which 50 % of the cells die (EC_{50}). Whole body animal tests will generally be reported as LD_{50} (the dose at which 50 % of the

TABLE 10.3. *Summary of waste streams, applicable EPA regulations, and control technology requirements for low/medium-Btu gasification plants (Gibson and Page, 1979)*

Waste stream	Source	Characteristics	Federal regulations to be considered[a]	Necessity of control technology application
Air				
Dust emissions	Coal pretreatment (storage, handling and crushing/sizing)	These emissions are primarily coal dust. Amounts will vary from one site to another depending on wind velocities and coal size	NAAQS[b]; PSD[c]; NSP[d]; NESHAP[e]; SIP[f]; Off-set Ruling; Performance Guidelines for Lurgi Gasification Plants	Yes
Vent gases	Control pretreatment (coal drying, partial oxidation, and briquetting)	The emissions will contain coal dust and combustion gases along with organic compounds liberated as a result of coal devolatilization reactions. Data on the characteristics of these organic species are lacking	Same as above	Further data on the characteristics of of these emissions are necessary to determine if control is neeeded and, if so, the types of applicable control techniques
Vent gases	Coal gasification (coal feeding device)	These vent gases will contain hazardous species found in the raw product gas exiting the gasifier	Same as above	Yes; control methods that may be applicable include recycling to the gas for inlet air or flaring

Vent gases	Coal gasification (ash removal device)	No data are yet available. These emissions may contain potentially hazardous species	Same as above	Yes; however, adequate control methods cannot be determined until these emissions have been characterized
Start-up vent stream	Coal gasification (coal gasifier)	Depending on the coal feedstock, tar and oil aerosols, sulphur species, and cyanides may be present	Same as above	Yes
Fugitive emissions	Coal gasification (coal gasifiers)	These emissions will contain hazardous species (e.g. hydrogen sulphide, carbon monoxide, and hydrogen cyanide) which are present in the raw product gas	Same as above	Yes
Tail gases	Gas purification (acid gas removal)	Few data are available. These streams will contain sulphur species, hydrocarbons, trace elements, and solvent carry-over	Same as above	Yes
Liquid				
Water runoff	Coal pretreatment (storage, handling, and crushing/sizing)	This stream may contain dissolved and suspended organics and inorganics	NSPS[d] Effluent Guidelines and Standards Pretreatment Standards Oil and Hazardous Substance Rules National Interim Primary Drinking Water Reg. Underground Injection Controls	Yes; however, adequate control methods cannot be determined until this stream has been further characterized

TABLE 10.3. *(continued)*

Waste stream	Source	Characteristics	Federal regulations to be considered[a]	Necessity of control technology application
Spent ash quench or sluice water	Coal gasification (ash removal device)	Limited data exist. This stream may contain dissolved and suspended organics and inorganics	Same as above	Same as above
Spent quench liquor	Gas purification (gas quenching and cooling)	Data are insufficient. There are significant quantities of suspended and dissolved organics (primarily phenols) from tars and oils, and inorganics in this stream	Same as above	Yes; control techniques will vary depending on the quantity and composition of this effluent stream
Process condensates, sorbents and reactants	Gas purification (acid gas removal)	No data have been reported. These streams may contain hazardous species such as cyanides, heavy metals, organics	Same as above	Yes; however, adequate control methods cannot be determined until this stream has been characterized further
Solid				
Solid wastes from crushing and sizing	Coal pretreatment (storage, handling, and crushing/sizing)	This stream consists of rock and mineral matter rejected from crushing and sizing coal. There are few data on the trace components, and their potential to contaminate surface and groundwaters is not known	RCRA[g] Regulations	This waste has been disposed of in landfills. Leaching tests are necessary to determine if control is necessary

Ash or slag	Coal gasification (ash removal device)	Data are limited, especially regarding the amount of unreacted coal, trace elements and organics	Same as above	Leaching tests are necessary to determine whether further treatment is necessary before final disposal. The organic content of the ash quench liquor may affect ash disposal
Collected particulate matter	Gas purification (particulate removal)	Limited data are available. This stream may contain unreacted carbon, organics and trace elements	Same as above	Further data are necessary to determine if control is needed or if this stream could be used as fuel (data indicate the presence of large amounts of unreacted carbon)

---

[a] The Toxic Substances Control Act regulations may potentially affect all waste streams and by-product streams.

[b] National Ambient Air Quality Standards.

[c] Prevention of Significant Deterioration Guidelines.

[d] New Source Performance Standards.

[e] National Emission Standards for Hazardous Air Pollutants.

[f] State Implementation Plans.

[g] Resource Conservation and Recovery Act.

TABLE 10.4. *Recommended standards of performance for coal gasification processes (Gibson and Page, 1979)*

Emission	Standard
Sulphur	1. Shall not exceed the mass rate, E, as determined by: $E = 0.025\ (A \times B)^{0.5}$ ($E = 0.019\ (A \times B)^{0.5}$) where E = kg h^{-1} (lb h^{-1}) sulphur emissions, calculated as elemental sulphur (mol. wt = 32.06 kg kg^{-1} mole), A = kg h^{-1} (lb h^{-1}) coal sulphur to gasifier, B = 10^6 kcal h^{-1} (10^6 Btu h^{-1}) higher heating value of coal to gasifier. 2. Shall not exceed 500 ppm dry, by volume, in any exhaust gas stream
Hydrocarbons (excluding ethane and methane)	1. Shall not exceed 0.01 10^6 kg kcal (0.06 10^{-6} lb Btu) higher heating value of coal to the gasifier 2. In any exhaust stream, shall not exceed 100 ppm dry, by volume

TABLE 10.5. *Performance guidelines for Lurgi gasification plants (Gibson and Page, 1979)*

Emission	Standard
Sulphur dioxide	$ES = 0.07\ (S_c)^{0.85}\ (HHV_c)^{0.15}$ for System I[a] $ES = 0.032\ (S_c)^{0.75}\ (HHV_c)^{0.25}$ for System II[b] where ES = total sulphur emissions, S_c = coal sulphur input, HHV_c = coal heat input.
Non-methane hydrocarbons	$EHC = 0.07\ HHV_c$, EHC = emissions of non-methane hydrocarbons, HHV_c = coal heat input.

[a] System I is an emission control system consisting of a Stretford sulphur recovery plant and a Claus sulphur recovery plant.

[b] System II is an emission control system consisting of a Stretford sulphur recovery plant.

animals die or LC_{50} (the concentration at which 50 % of the animals die). Other measures, such as tumour incidence, are also used.

There is some evidence that measures of lethal dosage for rats are directly related to the maximum dosages of anticancer drugs tolerated in man (Freireich *et al.*, 1966); and to dosages of organophosphate insecticides having no effect on humans as measured by erythrocyte cholinesterase inhibition (Hogan, 1982). The relationship of LDs for other materials to human health has not been measured. The Ames test appears to respond positively to many carcinogens; and the *in vitro* tests have yet to be linked to specific human response. Consequently, the biological testing procedures presently in use are only approximate characterizations of human toxicity. However, such tests do provide some guidance about process streams which are too chemically complex to adequately characterize and for fluids which contain known but unregulated pollutants.

TABLE 10.6. *Environmental and occupational evaluation programme TVA coal-to-ammonia plant (Williamson, 1981)*

Study area	Sample	Analyses performed
Gaseous emissions monitoring and characterization	Sulphur recovery tail gas	Sulphur species Nitrogen species Hydrocarbons Particulates Trace elements
Liquid effluent monitoring and characterization	Treated effluent Accumulator discharge to wastewater treatment	Priority pollutants (129) Trace elements Other[a]
Solid waste monitoring and characterization	Gasifier slag Solids to landfill (from wastewater treatment) Background Monitoring wells	Trace elements Hazardous waste extraction
Radiological characterization	Coal Gasifier slag Sulphur recovery tail gas Accumulator discharge to wastewater treatment Treated effluent/disposal pond and monitoring wells Solids to landfill (from wastewater treatment)	Ra-226 Ra-228
Medical surveillance	Operating personnel (individual) Maintenance personnel (individual)	Preplacement physical examinations Periodic physical examinations Transfer/termination physical examinations Follow-up physical examinations
Basic industrial hygiene	Operating personnel (individual) Maintenance personnel (individual) Employee work stations (ambient air)	CO H_2S COS Particulates Aromatic hydrocarbons

[a] NH_3, NO_2 and NO_3, organic N, TDS, TSSS VSS, BOD_5, alkalinity, COD, S^-, anide, TOC, formate.
Also may include Ca, Mg, SO_4, SiO_2, PO_4.

3.2.2. *Hazard evaluation*

Describing and eliminating the health hazards of coal processing requires an understanding of the nature of the raw materials, products, by-products, the type of pollutants produced, the design of the plant and its operating methods (Wadden, 1976). The structure of coal was described in Chapter 6 and is thought to be a highly linked amorphous polymer consisting of aggregates connected by relatively weak cross-links. If the cross-links are broken by pyrolysis, the resulting fragments are closely related to, if not the same as, coal liquids. Figure 10.1 indicates a schematic structure and points at

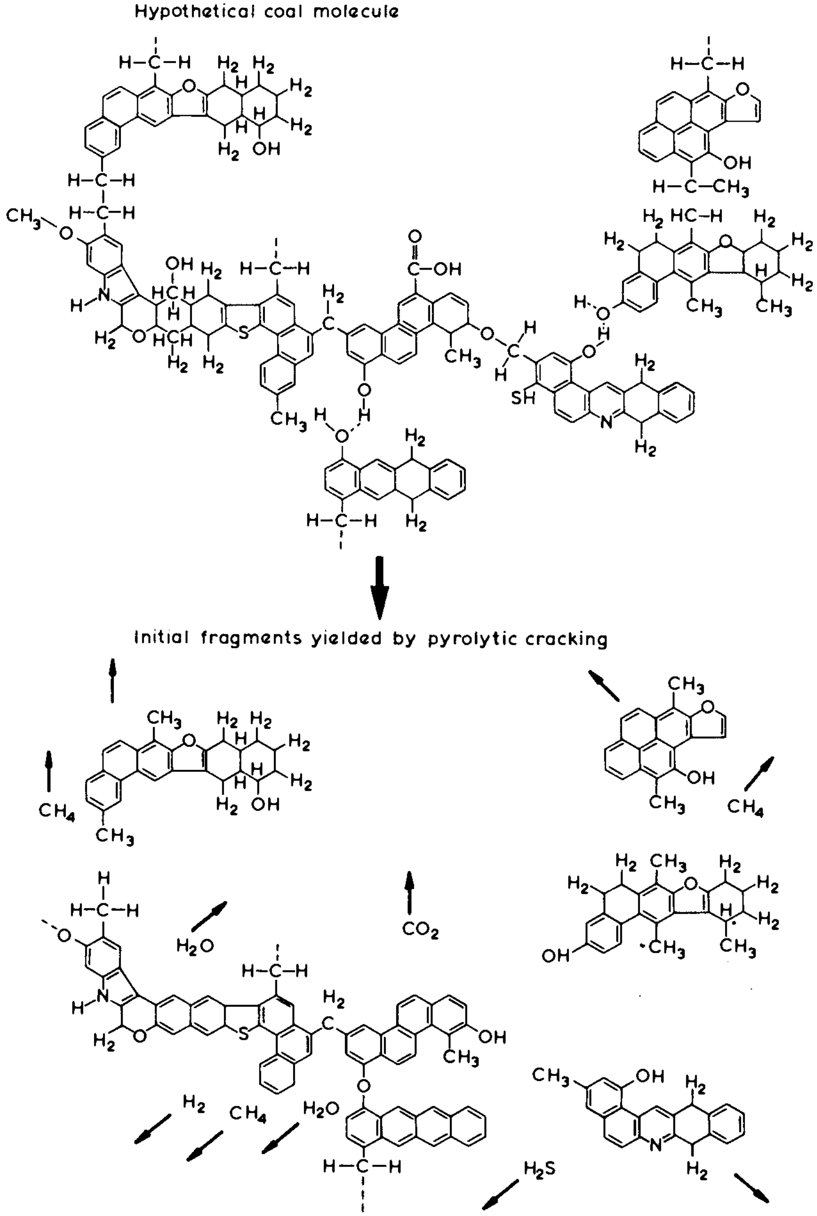


FIG. 10.1. Coal structure and the effect of pyrolitic cracking (adapted from Haggin, 1982). Reprinted with permission from *Chem. & Engin. News* 17–26, Aug. 9, 1982. Copyright © 1982 American Chemical Society.

which the bonds may separate. Further processing of the coal to gas or liquid then involved hydrogenation (Fig. 10.2). One of the probable results of hydrogen addition is also the production of aromatics, which may account for the greater aromatic content of coal liquids compared to those from oil or natural gas (Haggin, 1982). Such a difference in structure is illustrated in Fig. 10.3. Since the structure of coal is highly complex it is difficult to predict the exact nature of the by-products and end products from processing. In addition to this problem, aromatics, heavy metals and various sulphur

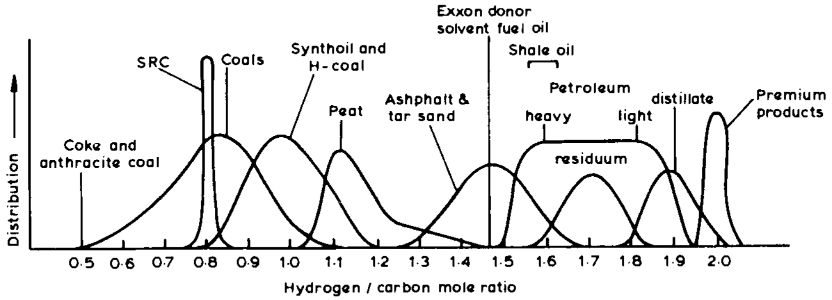


FIG. 10.2. The H:C ratio of various hydrocarbons (Haggin, 1982). Reprinted with permission from *Chem. & Engin. News* 17–26, Aug. 9, 1982. Copyright © 1982 American Chemical Society.

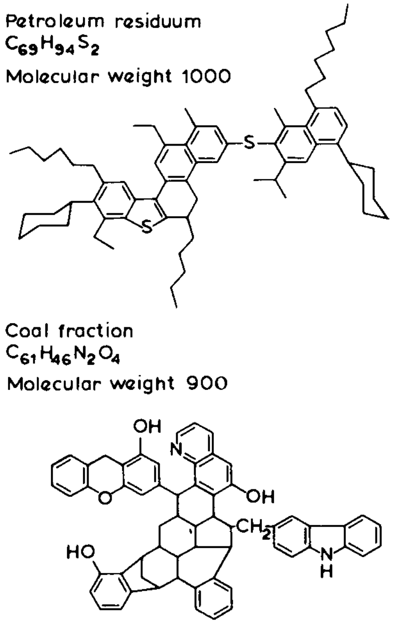


FIG. 10.3. The structure of a heavy fraction of a petroleum residuum with an asphaltol fraction compared with a coal fraction from Kentucky bituminous coal (Haggin, 1982). Reprinted with permission from *Chem. & Engin. News* 17–26, Aug. 9, 1982. Copyright © 1982 American Chemical Society.

compounds have been reported at relatively high levels in gas, liquid and solid products and by-products from coal processing plants (Wadden and Trabert, 1981). Under certain conditions CO_2 levels may be relatively high. All these considerations emphasize the necessity for monitoring worker health.

The nature of the gasification or liquefaction process will have an important effect on the types of pollutants produced and the operating characteristics of a particular plant. For instance, fixed or moving bed gasifiers like the Lurgi design result in considerable coal pyrolysis. This leads to the formation of large quantities of tars and oils, many of which contain carcinogenic materials. Table 10.7 summarizes polynuclear aromatic concentrations in the gas stream of the Kosovo (Yugoslavia) Lurgi plant (Bombaugh *et al.*, 1981). The benzo(*a*)pyrene concentration of 670 $\mu g\,m^{-3}$ in the LP Coal Lock Vent gas was evidently due to entrained oil and tar from the gasifier. The measured value related well to estimates made using the BaP concentration of oil or tar and the particulate loading of this exhaust stream (Table 10.8). Table 10.9 also indicates significant cytotoxicity for oil and tar. It has been suggested that tar can be recycled to extinction in the gasifier, thereby avoiding the disposal of solid waste (Clark, 1979). However, the basic design of the Lurgi gasifier is that of a batch process requiring the opening of the gasifier to add and remove coal. Some gas with associated particulate matter is inevitably released.

The Texaco process, including its modifications, and the Koppers–Totzek design are basically entrained bed systems (Clark, 1978; Koh, 1982). They therefore require the coal to be fed as a fine particle dust, or, more reliably, as a water slurry, in order to prevent the break-through of oxygen and consequential risks of explosion. It is difficult to convert the coal completely. In the

TABLE 10.7. *Concentrations ($\mu g\,m^{-3}$) of selected hazardous polynuclear aromatics in Kosovo gaseous discharge streams (Bombaugh et al., 1981)*

Source	BaA[a]	BaP[b]	DBahA[c]	BkF[d]
LP coal lock vent	163	670	52	670
Ammonia stripper vent	85	20	<2.1[e]	12
Naphtha storage tank vent	<0.06	0.085	0.06	0.11
Start-up vent	–[f]	139	<2.1	–
Tar tank vent	–	252	<10	–
Phenolic water bank vent	–	<50	<50	–
Medium oil tank vent	–	<6.5	<6.5	–
H_2/s-rich waste gas	–	<0.6	<0.6	–
CO_2-rich waste gas	–	<0.7	<0.7	–

[a] B(*a*)A, Benz(*a*)anthracene

[b] B(*a*)P, Benzo(*a*)pyrene.

[c] dB(*ah*)A, Dibenz(*a,h*)anthracene.

[d] B(*k*)F, Benzo(*k*)fluoranthene.

[e] All (<) values are calculated from a minimum detectable concentration of 0.1 ppm in the measuring solution.

[f] –not determined.

TABLE 10.8. *Hazardous PAHs (μg g-1) in Kosovo light tar and medium oil (Lee et al., 1981)*

	Light tar	Medium oil
7,12-Dimethylbenz(*a*)anthracene	1100	62
Benz(*a,h*)anthracene	490	160
Benzo(*b*)fluroanthene	310	120
Benzo(*a*)pyrene (BaP)	210	68
Benz(*a,b*)anthracene	23	7
3-Methylcholanthrene	26	NF[a]
252 Group	950[b]	280[b]

[a] Not found.

[b] BaP, 24% of 252 Group in both by-products.

TABLE 10.9. *A summary of the Kosovo Lurgi plant bioassay results (Lee et al., 1981)*

Description	Ames[a]	In vitro cytotoxicity[b] EC_{50}[c]	Rodent[d] test/ control
Ash	Negative	>1000	1/2
Naphtha	Negative	0.68	7/2
Medium oil	Negative	0.11	10/2
Tar	Positive	0.03	7/2
Heavy tar	Positive	0.07	3/1
Phenolic water	Positive	37	1/2
Phenosolvan wastewater (outlet)	Negative	98	0/2
ASTM ash leachate	Negative	>600	1/0
ASTM heavy tar leachate	Negative	120	3/0

[a] Highest concentration tested varied depending on sample toxicity.

[b] All samples tested in CHO assay except the ash sample which was tested in the RAM assay.

[c] EC_{50}s given in $\mu l\ ml^{-1}$ or $\mu g\ ml^{-1}$ (EC = effective concentration).

[d] Number of dead mice in test group over control group. Ten animals were used in each group.

Koppers–Totzek process about half the ash from the gasifier leaves in the form of fine particles (still containing unreacted carbon) in the exit gas (Koh, 1982). The Texaco gasifier overhead also contains substantial particulate matter (Schlinger and Richter, 1980). The use of slurries with a high coal content, or the elimination of water with a dry dust feed system, is economically advantageous. However, such streams are very abrasive to equipment: piping, heat exchangers, vessels and pumps. Regular replacement and maintenance are necessary to prevent leaks or plugging and subsequent fires or explosions (Evans and Verden, 1982).

Similarly, operation at high pressure and temperature puts greater stress on containment materials, and leaves less margin for operating errors which develop into hazardous conditions. However, cost considerations encourage the move to higher pressures and greater throughput. The Koppers–Totzek gasifier is an example. Normally operating at atmospheric pressure and around 1900 °C, this low pressure process has a relatively low coal throughput and high compression requirement (Koh, 1982). Further development of such processes will emphasize operation at greater pressures.

The large size of installations introduces other operating and environmental uncertainties. For instance, the largest Stretford sulphur removal process to be built in the USA (105 ton day^{-1}) has been contracted for the Great Plains gasification process (Stantberg, 1982). However, the Stretford process will not remove COS, and the presence of HCN will result in unregenerable compounds in the Stretford solution. The fate of mercaptans in the process is also uncertain (Page, Corbett and Magee, 1981). It is likely that such compounds will be present in the feed stream as has been demonstrated by the composition of the H_2S-rich waste gas stream in the Kosovo Lurgi plant (H_2S-rich waste gas in Table 10.10).

3.2.3. *Process data*

Because the Lurgi and Texaco gasification processes are currently being utilized in large installations in the USA, some of the most pertinent test data from each of these processes are summarized here. The Kosovo Lurgi plant converts approximately 2000 tons of coal per day to medium Btu gas. Schematics of the overall process and key process streams are shown in Figs. 10.4 and 10.5. For comparison, the USA Great Plains block flow

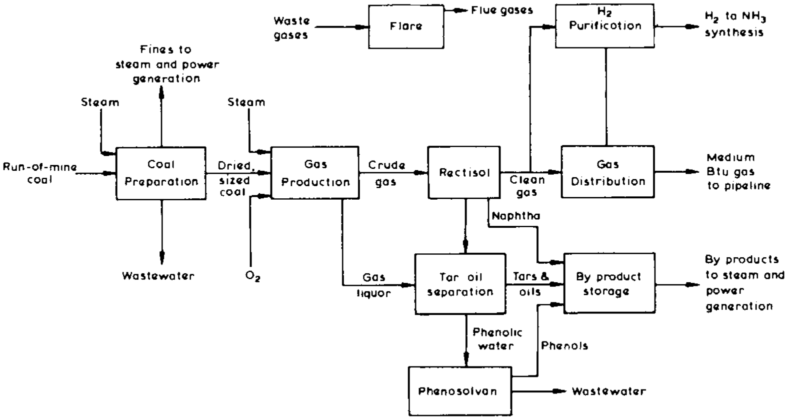


FIG. 10.4. Simplified flow diagram of the Kosovo coal preparation/gasification plant operations (Lee et al., 1981).

TABLE 10.10. *Component concentrations in Kosovo gaseous streams (Lee et al., 1981)*

Plant section:	Fleissner drier	Gas production			Rectisol	
Sample point:	Autoclave vent	Low pressure coal lock vent	Gas liquor tank vent	High pressure coal lock (flare feed stream)	H_2S-rich Waste gas (flare feed stream)	Co_2-rich waste gas
Component ($\mu g\ m^{-3}$ at 25 °C)						
Fixed gases						
H_2	Tr[a]	$3.05\ 10^{7}$	$9.74\ 10^{6}$	$2.64\ 10^{7}$	$8.98\ 10^{4}$	Tr
O_2	$1.83\ 10^{8}$	$3.53\ 10^{6}$	$1.81\ 10^{8}$	$3.14\ 10^{6}$	Tr	Tr
N_2	$6.41\ 10^{8}$	$2.06\ 10^{6}$	$6.35\ 10^{8}$	$1.60\ 10^{6}$	Tr	Tr
CH_4	Tr	$5.64\ 10^{7}$	–	$6.88\ 10^{7}$	$2.82\ 10^{7}$	$7.84\ 10^{6}$
CO	Tr	$1.67\ 10^{8}$	$2.97\ 10^{7}$	$1.37\ 10^{8}$	$1.26\ 10^{7}$	Tr
CO_2	$5.21\ 10^{8}$	$6.56\ 10^{8}$	–	$7.55\ 10^{8}$	$1.58\ 10^{9}$	$1.69\ 10^{9}$
Sulphur species						
H_2S	$3.34\ 10^{6}$	$1.80\ 10^{7}$	$2.02\ 10^{6}$	$4.87\ 10^{6}$	$6.32\ 10^{7}$	$5.43\ 10^{4}$
COS	$7.36\ 10^{4}$	$2.70\ 10^{5}$	–	$2.95\ 10^{5}$	$1.03\ 10^{6}$	$1.52\ 10^{5}$
CH_2SH	$6.68\ 10^{6}$	$8.25\ 10^{5}$	–	$9.04\ 10^{5}$	$4.13\ 10^{6}$	$1.67\ 10^{4}$
C_2H_SSH	$5.33\ 10^{6}$	$5.57\ 10^{5}$	–	$5.35\ 10^{5}$	$1.98\ 10^{6}$	$1.12\ 10^{4}$
Hydrocarbons						
C_2H_6	Tr	$2.96\ 10^{6}$	–	$5.16\ 10^{6}$	$1.01\ 10^{7}$	$1.97\ 10^{7}$
C_2H_4	NF[b]	Tr	–	Tr	Tr	Tr
$C_3{}^1s$	$5.40\ 10^{5}$	$2.52\ 10^{6}$	–	$4.50\ 10^{6}$	$1.14\ 10^{7}$	$5.04\ 10^{6}$
$C_4{}^1s$	$7.14\ 10^{5}$	$1.19\ 10^{6}$	–	$2.61\ 10^{6}$	$7.60\ 10^{6}$	Tr
$C_5{}^1s$	NF	Tr	–	$2.95\ 10^{5}$	$1.18\ 10^{6}$	Tr
$C_6{}^+$	$3.43\ 10^{5}$	$4.22\ 10^{6}$	–	$2.82\ 10^{6}$	$7.39\ 10^{6}$	Tr
Benzene	$5.44\ 10^{4}$	$2.43\ 10^{6}$	–	$1.76\ 10^{6}$	$3.51\ 10^{5}$	$3.19\ 10^{3}$
Toluene	$2.56\ 10^{4}$	$8.28\ 10^{5}$	–	$3.76\ 10^{5}$	$3.00\ 10^{4}$	Tr
Xylene 6 Ethylbenzene	$1.82\ 10^{4}$	$3.25\ 10^{5}$	–	$1.65\ 10^{5}$	NF	Tr
Phenols	–	$2.19\ 10^{4}$	Tr	$9.61\ 10^{3}$	Tr	NF
Nitrogen species						
NH_3	–	$1.73\ 10^{6}$	$4.80\ 10^{5}$	NF	$1.53\ 10^{6}$	$3.20\ 10^{4}$
HCN	–	$6.62\ 10^{5}$	–	$1.88\ 10^{6}$	$2.21\ 10^{5}$	$1.44\ 10^{4}$
Dry gas flow rate (m^3 gasifier hr^{-1} at 25 °C)	57.8	21	44	230	2300	3600

[a] Tr, trace.

[b] NF, not found.

TABLE 10.10 (*continued*)

Plant section:	Tank separation					Phenosolvan		By-product storage
Sample point:	Tar tank vent	Medium oil tank vent	Condensate tank vent	Tar separation waste gas (flare feed stream)	Phenolic water tank vent	NH_3 stripper vent	Cooler tank vent	Naptha storage
Component ($\mu g\ m^{-3}$ at 25 °C)								
Fixed gases								
H_2	Tr[a]	Tr	$1.20\ 10^{7}$	$8.98\ 10^{6}$	Tr	NF[b]	–	NF
O_2	$2.48\ 10^{8}$	$5.88\ 10^{6}$	$2.16\ 10^{8}$	Tr	$1.70\ 10^{8}$	–	–	$3.40\ 10^{7}$
N_2	$8.87\ 10^{8}$	$1.26\ 10^{7}$	$6.95\ 10^{8}$	Tr	$4.46\ 10^{8}$	–	–	$9.61\ 10^{8}$
CH_4	$1.04\ 10^{6}$	$4.98\ 10^{7}$	$7.84\ 10^{6}$	$2.29\ 10^{7}$	$1.31\ 10^{6}$	Tr	–	NF
CO	Tr	$6.75\ 10^{7}$	NF	$1.26\ 10^{7}$	NF	NF	–	NF
CO_2	$1.55\ 10^{7}$	$1.01\ 10^{9}$	$1.11\ 10^{8}$	$1.40\ 10^{9}$	$6.29\ 10^{8}$	$9.89\ 10^{8}$	–	$1.53\ 10^{7}$
Sulphur species								
H_2S_3	$9.61\ 10^{6}$	$3.62\ 10^{7}$	$8.61\ 10^{6}$	$1.25\ 10^{7}$	$1.75\ 10^{7}$	$2.72\ 10^{7}$	NF	NF
COS	$2.70\ 10^{5}$	$2.36\ 10^{5}$	–	$2.94\ 10^{5}$	$1.01\ 10^{5}$	NF	–	NF
CH_1SH	$7.66\ 10^{5}$	$1.02\ 10^{7}$	$4.11\ 10^{5}$	$4.91\ 10^{6}$	$4.13\ 10^{6}$	$5.70\ 10^{5}$	–	$5.11\ 10^{6}$
C_2H_5SH	$6.09\ 10^{6}$	$5.33\ 10^{6}$	$1.82\ 10^{5}$	$4.06\ 10^{6}$	$1.83\ 10^{7}$	$2.54\ 10^{5}$	–	$2.46\ 10^{7}$
Hydrocarbons								
C_2H_4	Tr	$4.18\ 10^{6}$	$8.57\ 10^{5}$	$4.05\ 10^{6}$	$2.46\ 10^{5}$	Tr	–	Tr
C_2H_4	–	Tr	–	Tr	–	–	–	–
$C_3{}^1s$	$1.80\ 10^{5}$	$5.40\ 10^{6}$	$9.01\ 10^{5}$	$7.39\ 10^{6}$	$3.60\ 10^{5}$	Tr	–	$1.80\ 10^{5}$
$C_4{}^1s$	Tr	$5.94\ 10^{6}$	$7.13\ 10^{5}$	$9.74\ 10^{6}$	$4.75\ 10^{5}$	Tr	–	$1.66\ 10^{6}$
$C_5{}^1s$	Tr	$2.65\ 10^{6}$	$1.18\ 10^{6}$	$2.65\ 10^{6}$	$1.77\ 10^{6}$	Tr	–	$2.36\ 10^{6}$
$C_6{}^+$	$1.30\ 10^{7}$	$8.45\ 10^{7}$	–	$4.58\ 10^{7}$	$6.34\ 10^{7}$	NF	–	$1.87\ 10^{8}$
Benzene	$6.38\ 10^{6}$	$2.44\ 10^{7}$	$1.66\ 10^{7}$	$3.06\ 10^{7}$	$3.51\ 10^{7}$	Tr	–	$1.20\ 10^{8}$
Toluene	$3.61\ 10^{6}$	$5.27\ 10^{6}$	$1.13\ 10^{7}$	$4.52\ 10^{6}$	$8.66\ 10^{6}$	–	–	$7.15\ 10^{6}$
Xylene 6 Ethylbenzene	$9.52\ 10^{5}$	$6.06\ 10^{5}$	–	$6.51\ 10^{5}$	$1.21\ 10^{6}$	Tr	–	$2.60\ 10^{5}$
Phenols	$2.19\ 10^{5}$	$1.73\ 10^{5}$	Tr	$1.62\ 10^{4}$	Tr	$2.38\ 10^{7}$	Tr	Tr
Nitrogen								
NH_3	$1.81\ 10^{6}$	$1.32\ 10^{4}$	NF	$1.34\ 10^{7}$	$8.35\ 10^{6}$	$2.91\ 10^{8}$	$5.71\ 10^{7}$	NF
HCN	$1.44\ 10^{5}$	$6.28\ 10^{4}$	$1.88\ 10^{5}$	$7.05\ 10^{4}$	$4.20\ 10^{4}$	$5.30\ 10^{6}$	–	$1.21\ 10^{6}$
Dry gas flow rate (m^3 gasifier hr^{-1})	0.55	1.7	3.38	28	5.5	260	4.4	4.5

[a] Tr, trace.

[b] NF, not found.

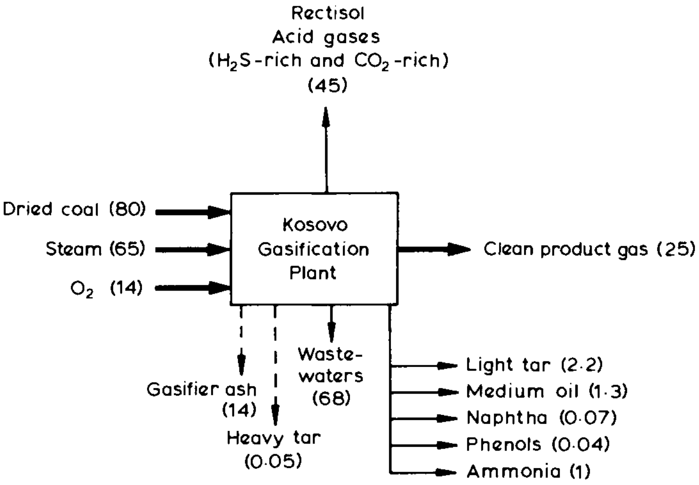


FIG. 10.5. Design flow rates of key streams in the Kosovo gasification plant (Lee et al., 1981). Values in t hr^{-1} based on 5 gasifiers in service.

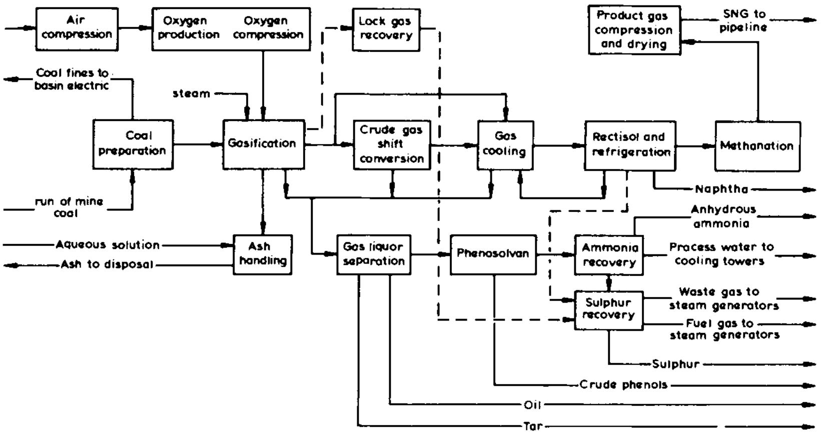


FIG. 10.6. Process block flow diagram of Great Plains coal gasification project (Kühn, 1982). Reproduced by permission of the American Institute of Chemical Engineers.

diagram is shown in Fig. 10.6. While the products and some of the process streams differ, many are common to both plants.

The composition of the Kosovo gas streams is given in Table 10.10. Note that flow rates are normalized on the basis of one operating gasifier. None of these compositions includes particulate loadings. These are given in Table 10.11. Polycyclic hydrocarbon levels in the tars and oils, which make up the condensed fraction of the total particulate matter, are shown in Table 10.8. The composition of the gas entering and leaving the Rectisol

TABLE 10.11. *Particulate concentration and flow rate data for Kosovo gaseous streams (Lee et al., 1981)*

	Autoclave vent	Coal room vent	Low pressure coal lock vent	Gasifier start-up vent	High pressure coal lock vent	Tar/oil separation waste gas	Combined gases to flare[a]
Dry gas flow rate (m^3 gasifier hr^{-1} at 25 °C)	57.8	7200	21	–	230	28	1330
Total particulate (mg m^{-3} at 25 °C)	1080	90	8100	9450	960	920	410
Condensed organics (tars and oils) (mg m^{-3})	480	–	7300	8980	660	660	310
Dissolved solids (mg m^{-3})	320	–	650	400	240	230	54
Filtered solids (mg m^{-3})	280	–	220	61	61	29	47

[a] The streams that make up this category are discussed individually here since in other plants different streams may be sent to the flare.

cleaning plant is shown in Table 10.12. The composition of aqueous streams is given in Table 10.13. Table 10.9 shows bioassay results.

Although more stringent control procedures can be implemented for new plants, the Kosovo tests provide significant information on uncontrolled discharges. It was also found that the gasifier ash was not very toxic (Table 10.9); that the organic by-products (light tar, medium oil and naphtha) were the most toxic of materials tested in rodent tests; and that the Phenosolvan process significantly reduced the biological activity of the wastewaters. In general, the discharge of hazardous metals did not appear to be remarkable, the organic composition of waste streams being a much greater potential health hazard. However, as noted in Table 10.12 mercury was found in the phenolic water of the Phenosolvan process. The Rectisol process was effective in cleaning up the product gas but its use in other plants will depend on the availability of satisfactory control technology for the H_2S-

TABLE 10.12. *Comparison of product gas composition entering and leaving the rectisol gas cleaning plant (Lee et al., 1981).*

Composition	Crude product gas	Clean product gas
Fixed gases (vol. %)		
H_2	38.1	60
O_2	0.36	0.44
N_2	0.64	0.38
CH_4	11.5	16
CO	15	22
CO_2	32	0.02
Sulphur species (ppmv)		
H_2S	6000	NF[a]
COS	97	0.17
CH_3SH	590	1.1
C_2H_5SH	200	1.0
Hydrocarbons		
Ethane (vol. %)	0.47	0.15
Ethylene (vol. %)	0.04	Tr[b]
C_3 (vol. %)	0.19	Tr
C_4 (vol. %)	0.074	Tr
C_5 (vol. %)	0.044	Tr
C_6 + (vol. %)	0.064	0.03
Benzene (ppmv)	750	–[c]
Toluene (ppmv)	230	–
Xylene and ethylbenzene (ppmv)	100	–
Phenols (ppmv)	Tr	Tr
Higher aromatics (ppmv)	–	–
Nitrogen species (ppmv)		
NH_2	3.3	Tr
HC	320	–

[a] NF, not found, <0.01 vol. % fixed gases and <1 ppmv for all other species.
[b] Tr, trace, –0.01 vol. % for fixed gases and –1 ppmv for all other species.
[c] No data available.

TABLE 10.13. *Kosovo aqueous stream data (Lee et al., 1981)*

Plant section:	Gas production	Phenosolvan	
Sample point:	Gasification section wastewater	Phenolic	Phenosolvan outlet water
Flow rate (m^3 gasifier hr^{-1})	3.0	>13	13
pH	8.1	9.2	9.6
Temperature (°C)	–[a]	60	33
Solids analysis (mg l^{-1})			
Total solids	10,900	2230	1350
Suspended solids	8760	150	190
Dissolved solids	2100	2170	1160
Water quality parameters			
COD (as mg O_2 l^{-1})	1460	18,900	7910
Permanganate (mg l^{-1})	8060	14,200	4040
BOD$_2$ (as mg O_2 l^{-1})	90	9030	2350
TOC	–	4970	1470
Aqueous composition data (mg l^{-1})			
Total phenols	–	2120	230
Volatile phenols	0.17	–	130
Free ammonia	Tr[b]	3510	Tr
Fixed ammonia	1.9	250	205
Cyanide	0.01	<1	0.019
Nitrites	0.40	–	Tr
Nitrates	4.8	<1	11.4
Pyridines	–	140	–
Chlorides	28	–	60
Fluorides	0.91	–	Tr
Mercury	–	0.14	–
Total sulphur	–	–	84
Sulphites	Tr	–	–
Sulphates	495	–	110
Sulphides	Tr	–	–
Thiocyanates	0.26	>75	<75
PNA analysis (mg l^{-1})			
Benz(*a*)anthracene	–	0.92	NF[c]
7,12-dimethylbenz(*a*) anthracene	–	0.23	NF
Benzo(*a*)fluoranthrene	–	0.68	NF
Benzo(*a*)pyrene	–	0.19	–
3-methylcholanthrene	–	<0.004	NF
Dibenz(*a*,*h*)anthracene	–	0.02	NF
252 Group (as B*a*P)	–	1.3	0.19

[a] —, not analysed.

[b] Tr, trace.

[c] NF, not found.

rich and CO_2-rich waste gases (H_2S and CO_2-rich waste gas streams in Table 10.10 (Lee *et al.*, 1981).

The Texaco process converts coal to medium-Btu synthesis gas. The process is shown schematically in Fig. 10.7. The composition of various output streams from a 15 ton day^{-1} pilot plant is given in Tables 10.13–10.16. Phenols and other organics in excess water, slag water and slag, H_2S, COS, cyanide and polynuclear hydrocarbons in the fuel gas should be noted (Schlinger and Richter, 1980). These data give only a limited indication of pollutant discharges from typical processes. This is because typical flow rates are not available and because the pilot plant is relatively small. The TVA's coal-to-ammonia plant should supply further operating information.

3.3. *Communities in the Plant Vicinity*

Both normal and hypersusceptible populations need to be considered in evaluating community exposure. Groups hypersusceptible to hydrogenation discharges include children, the elderly and those with illness or genetic dysfunction which would make them more vulnerable to the stress of exposure.

Collection and periodic review of cancer, disease and birth defect registries are necessary requirements of community health and surveillance. Monitoring for pollutants in air and water in the plant vicinity would also be prudent. When pollutant quantities discharged from the plant can be determined, methods are available for approximating community exposures. Considerable research effort is required before the exposure commitment methodology can be applied to such situations in practice.

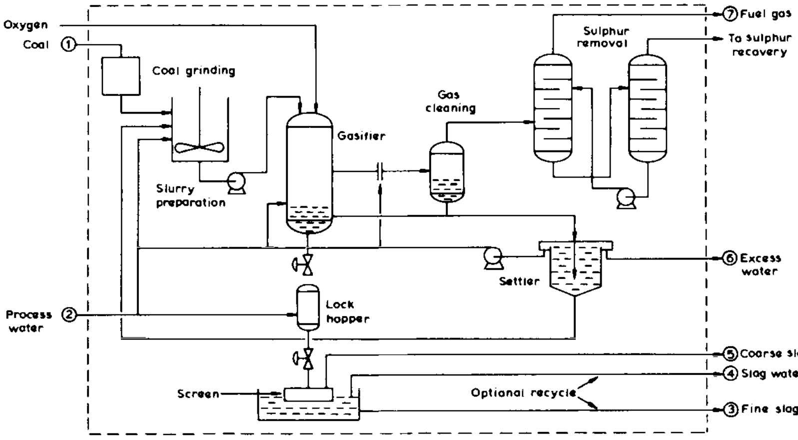


FIG. 10.7. Schematic flow diagram of the Texaco coal gasification process (Schlinger and Richter, 1980).

TABLE 10.14. *Water quality data from 15 ton day^{-1} Texaco gasification process (Schlinger and Richter, 1980)*

	Water quality Eastern coal				Water quality Western coal		
Stream no.	2 Process water	4 Slag water	6 Excess water	Stream no.	2 Process water	4 Slag water	6 Excess water
pH	NR[a]	8.0	7.7	pH	5.3	8.9	8.8
TDS, ppm	0.0	355	1708	TDS, ppm	<5	28	420
Conductivity	NR	1650	7100	Conductivity	<10	410	10 800
COD, ppm	NR	221	405	COD, ppm	<5	48	760
TOC, ppm	1	28	215	TOC, ppm	0.4	38	360
Ammonia, ppm	NR	380	1270	Ammonia, ppm	<1	130	2 700
Anions, ppm				Anions, ppm			
Bromide	<1	<1	<1	Bromide	<0.05	<0.05	<0.05
Chloride	29	146	740	Chloride	<0.1	42	7.5
Fluoride	<1	31	175	Fluoride	<0.01	<0.1	28
Cyanide	NR	<1	8	Cyanide	<0.2	0.4	11
Formate	<1	94	522	Formate	<0.1	56	1 200
Nitrate	<1	3	9	Nitrate	<0.5	<0.5	2.0
Sulphide	0.3	6	316	Sulphide	<0.5	<0.5	61
Sulphate	25	38	21	Sulphate	1.3	5	47
Thiocynate	NR	2	8	Thiocynate	<4	<4	29
Thiosulphate	NR	5	NR				
Trace organics, ppb				Trace organics, ppb			
Benzene		<0.5	3.0	Phenols	NR	1.2	1.2
Toluene		<0.5	2.0	Unidentified	NR	7.0	2.8
Anthracene		1.0	0.5				
Fluoranthene		2.0	0.5				
Naphthalene		–	2.0				
Pyrene		0.6	0.7				
Phenanthrene		0.2	0.02				
Phenols		NR	<10				

Antimony	<0.001	0.016	<0.001	Antimony	<0.001	<0.001	<0.001
Arsenic	0.002	0.07	0.022	Arsenic	<0.001	0.015	0.016
Barium	0.03	10	54	Barium	0.004	0.05	0.90
Beryllium	0.0004	0.005	0.018	Beryllium	<0.001	<0.001	0.002
Boron	–	–	–	Boron	0.2	0.3	1.4
Cadmiun	0.010	0.003	0.003	Cadmium	<0.006	<0.001	<0.001
Chromium	0.002	0.003	0.006	Chromium	<0.001	<0.001	<0.001
Cobalt	0.005	10	0.021	Cobalt	<0.001	<0.001	<0.001
Copper	0.01	0.017	0.010	Copper	0.031	0.008	0.005
Fluorine	<1	31	175	Fluorine	<0.1	<0.01	29
Lead	0.005	0.045	0.054	Lead	<0.005	0.011	<0.005
Manganese	0.13	0.08		Manganese	0.002	0.003	0.024
Mercury	<0.0001	<0.0001	<0.0001	Mercury	0.002	0.002	0.002
Molybdenum	<0.02	0.03	<0.02	Molybdenum	<0.001	0.003	0.015
Nickel	0.03	0.07	0.03	Nickel	<0.01	<0.01	<0.01
Selenium	0.005	0.033	0.010	Selenium	<0.001	0.025	0.460
Silver	0.0006	0.002	0.002	Silver	<0.002	<0.002	<0.002
Thallium	0.002	0.006	0.01	Thallium	<0.002	<0.002	0.003
Vanadium	<0.04	<0.04	<0.04	Vanadium	<0.01	<0.01	0.03
Zinc	0.195	0.046	0.03	Zinc	<0.01	<0.01	<0.01

---

[a] NR, not recorded.

TABLE 10.15. *Slag and leachate composition from 15 ton day^{-1} Texaco gasification process (Schlinger and Richter, 1980)*

	Slag analyses					Properties of deionized water leachate ASTM Method (Proposed)			
Stream no.	5	4	5	4					
	Eastern coal		Western coal			Eastern coal		Western coal	
	Coarse slag	Fine slag	Coarse slag	Fine slag		Coarse slag	Fine slag	Coarse slag	Fine slag
Major components, Wt. %									
Ash	99.30	65.14	99.91	81.52	pH	6.0	7.1	9.6	8.9
Carbon	1.79	32.39	0.40	17.21	TDS, ppm	NR[a]	NR	250	310
Hydrogen	0.023	0.31	0.16	0.01	Conductivity	100	600	53	135
Nitrogen	0.06	0.49	0.08	0.26	COD, ppm	NR	NR	45	28
Sulphur	0.83	2.08	0.0	0.09	TOC, ppm	2	3	15	9.1
					Ammonia, ppm	1.2	4.6	NR	NR
Trace elements, ppm (wt)					Anions, ppm				
Antimony	0.9	0.9	<0.5	<0.5	Bromide	<0.1	<0.1	–	–
Arsenic	15.0	20.5	30	22	Chloride	1.5	2.2	–	–
Barium	378	301.5	3200	2650	Fluoride	4.9	8.7	–	–
Beryllium	7.5	6.6	2.2	2.3	Cyanide	<0.001	<0.001	–	–
Boron	–	–	63	40	Formate	<1	<1	–	–
					Nitrate	0.4	0.5	–	–
Chlorine	10	44	6	300	Nitrite	<0.1	<0.1	–	–
Cadmium	5.0	6.7	0.32	0.40	Sulphate	26.9	3.3	–	–
Chromium	1790	842	2100	1450					
Cobalt	125	85.5	13	9.7					
Copper	81	90	49	48					
Fluorine	45	210	80	230					
Lead	30	47	0.9	2.8					
Manganese	614	525	380	300					
Mercury	0.15	0.095	0.18	0.28					
Molybdenum	64	98.5	2.3	7.6					
Nickel	229	267	39	34					
Selenium	14	17.5	0.76	4.0					
Silver	0.1	0.2	<0.3	0.6					
Thallium	0.3	0.3 1.0							
			8.4						
Vanadium	340	280	180	140					
Zinc	65	425	13	26					

TABLE 10.15 (*continued*)

	Trace compounds in slag components					Properties of deionized water leachate ASTM Method (Proposed)			
Stream no.	5	3	5	3					
	Eastern coal		Western coal			Eastern coal		Western coal	
	Coarse slag	Fine flag	Coarse slag	Fine slag		Coarse slag	Fine slag	Coarse slag	Fine slag
Compound[b], ppb					Element, ppm				
Naphthalene	ND[c]	ND	150	240	Antimony	0.003	0.005	–	–
Alkyl naphthalenes	NR	NR	ND	191	Arsenic	0.007	0.019	<0.01	<0.01
Phenanthrene[d]	12	610	230	18	Barium	<0.38	<0.38	0.04	0.2
Acenaphthene	ND	ND	10	21	Beryllium	<0.001	<0.001	–	–
Fluorene	ND	ND	15	ND	Boron	–	–	–	–
Pyrene	45	860	23	ND	Cadmium	0.004	0.0006	–	–
Anthraquinone	NR	NR	210	ND	Chromium	<0.002	0.002	0.015	0.003
Fluoranthene	63	840	NR	NR	Cobalt	0.029	0.028	<0.002	0.001
					Copper	<0.003	0.016	0.03	<0.006
					Fluorine	4.9	8.7	–	9.0
					Lead	0.005	0.008	–	–
					Manganese	0.105	0.212	0.002	0.03
					Mercury	<0.005	<0.005	0.01	<0.002
					Molybdenum	<0.9	<0.9	–	–
					Nickel	0.875	0.131	<0.02	–
					Selenium	<0.009	0.027	<0.002	0.05
					Silver	<0.002	<0.002	–	–
					Thallium	<0.006	<0.006	–	0.001
					Vanadium	<1.8	<1.8	0.14	0.16
					Zinc	0.098	0.052	<0.09	<0.09

---

[a] NR, not reported.

[b] No other compounds on the EPA Priority Pollutant List detected.

[c] ND, not detected.

[d] Includes anthracene.

TABLE 10.16. *Fuel gas composition from 15 ton day^{-1} Texaco gasification process (Schlinger and Richter, 1980)*

	Fuel gas composition			Trace components in fuel gas stream	
Stream no.	7	7	Stream no.	7	7
	Eastern coal	Western coal		Eastern coal	Western coal
Major components, vol. %			Component mg SCM^{-1}		
Hydrogen	36.8	36.2	Ammonia	3.5	<0.5
Carbon monoxide	44.5	44.4	Formate	64	NR[b]
Carbon dioxide	18.2	18.7	Cyanide	0.03	7.2
Argon	0.09	0.16	Thiocyanate	<0.1	NR
Nitrogen	0.08	0.36	Chloride	23	NR
Sulphur species, ppm (vol.)[a]			Fluoride	4.4	<0.02
			NO_x	<0.002	<0.001
Hydrogen sulphide	1300	520			
Carbonyl sulphide	200	32	Trace elements, μg SCM^{-1}		
Methyl mercaptan	–	1	Antimony	<0.2	<0.2
Ethyl mercaptan	–	1	Arsenic	2.1	<0.2
Dimethyl mercaptan	–	1	Barium	25	3
Carbon disulphide	–	1	Beryllium	0.14	<0.2
			Boron	NR	0.7
Hydrocarbons, ppm (vol.)					
			Cadmium	2	<0.2
Methane	28	570	Chromium	2	<0.2
Ethane	<0.1	<1	Cobalt	1	1.6
Ethylene	<0.1	<1	Copper	5	<0.4

Acetylene	<0.1	<1
Propane	0.9	<1
Propene	<0.1	<1
Butane	<0.1	<1
Pentanes	<0.1	6
Hexanes	<0.1	<1

Trace organics in fuel gas

Stream no.	7 Eastern coal	7 Western coal
Component $\mu g\ SCM^{-1}$		
Benzene	139	210
Alkyl benzenes	16	54
Naphthalene	2	15
Anthracene	0.2	0.12
Fluoranthene	1.1	–
Indene	–	3.2
Pyrene	0.3	–

Lead	0.8	<0.4
Manganese	<1.7	<0.2
Mercury	3	2
Molybdenum	<5.8	52
Nickel	10	5
Selenium	0.6	<0.2
Silver	0.5	<0.4
Thallium	0.5	<0.4
Vanadium	14	<2
Zinc	5	<2

---

[a] Operating conditions of the pilot unit selected to remove 90% of the sulphur. Higher efficiency can be achieved by altering operating conditions.

[b] NR, not recorded.

3.4. Product Users

The chemical composition of liquid and gaseous products from coal hydrogenation is different from the composition of equivalent materials extracted from petroleum or natural gas. Product oils are likely to have high aromatic contents (Table 10.17) and significant amounts of polynuclear hydrocarbons, some of which are carcinogenic (Table 10.8). It has been reported that Russian data indicate that gasoline from coal produces skin rashes while gasoline from petroleum does not produce a rash under similar circumstances (Brown and Witter, 1979). Burning characteristics may also be different.

As indicated in Table 10.18, the hydrogen content of product gas from coal gasification will be much higher in H_2 than natural gas. Whether this higher hydrogen content will be significant in terms of hydrogen embrittlement of existing pipelines and gas-holders, or a hazard in industrial and domestic burners, remains to be determined. Table 10.18 also shows higher CO concentrations in low-Btu product gas than in natural gas. Evaluation of the potential environmental health consequences to users of these and other hydrogenation products requires more detailed information about the composition and variability of such products and the chemical and physical changes they may undergo during combustion or further chemical processing.

4. Control and Planning

Most of the problems of control in hydrogenation plants are probably solvable, providing that the nature and source of the pollution are properly described. In addition, low-sulphur refined fuels produced by such processes

TABLE 10.17. *Organic composition of Lurgi oil produced at the Westfield, Scotland facility (Ghassemi, Crawford and Quinlivan, 1979)*

Compound/class	Concentration (wt.%)
Paraffins	10.71
Aromatics	89.30
Percentage aromatics as:	
Benzene	19.56
Toluene	28.40
Xylene and ethyl benzene	14.70
Ethyl toluene	2.69
Trimethyl benzenes	11.80
Styrene	1.07
Indane	1.43
1,2-benzofuran	1.09
Indene	5.37
Naphthalene	1.40
Thiophenes	1.77

TABLE 10.18. *Coal gasification product gas composition (vol. %)*

		Wellman–Galusha[b]	Lurgi[c]	Koppers–Totzek[d]	Hygas Pilot Plant[e]	Estimated Lurgi[e]
	Natural gas[a]	(Dry product gas) Glen Cery, York, Pa., USA	(After CO conversion) Usti, Czechoslovakia	(Shifted stream) Modderfontein, South Africa	Chicago, USA	(Shifted methanized stream)
CH_4	99.2	0.2	9.9	0.1	67.4	92.9
H_2	—	16.3	39.3	54.6	15.5	4.2
CO_2	0.2	5.5	32.7	41.3	0.0	1.8
CO	—	25.5	13.4	3.0	0.0	0.01
$N_2+(A)$	0.6	51.5	2.2	1.0	17.1	1.1
C_2H_6, C_2H_4, etc.	—	—	0.7	—	0.0	—
H_2S	—	690 ppm	1.5	—	—	—
CSO	—	93 ppm	—	—	< 1000 ppm	—
CS_2	—	< 1 ppm	—	—	—	—
O_2	—	—	0.3	—	—	—
Lower heating value						
kcal m^{-3} at 0°C	8940	1207	—	1508	6168	8720
(Btu ft^{-3})	(953)	(136)	—	(169)	(693)	(980)

[a] Sinor (1979).

[b] Thomas, Trede and Page (1979).

[c] Mácek, Prášek and Šimánek (1976).

[d] Franzen and Goeke (1976).

[e] Ghassemi, Crawford and Quinlivan (1978).

are much more acceptable in urban areas than raw coal or heavy, high-sulphur oils. However, because of the large quantities and complex nature of the conversion process, there must be a recognition that these new types of operations may produce kinds of pollutants with which we are unfamiliar.

Available process stream data for full-scale and pilot plants, although meagre, show significant concentrations of a number of pollutants which are recognized hazards to lung and skin, and some which are known carcinogens. A major effort is needed to further characterize the wastes and products of the hydrogenation process, particularly in terms of toxic materials which may appear in low concentrations. Complete quantification of pollutant discharges requires long-term operation of large-scale pilot plants. (This recommendation is particularly important because of the way pilot plants are usually operated. Due to time and economic restrictions there is a tendency to change operating conditions frequently.)

NIOSH (1978a) have stressed the importance of medical monitoring, including a pre-employment medical examination; regular check-ups (more frequent and in more detail for high-risk groups); long-term follow-up of high-risk individuals after transfer to other job categories or termination of employment; full record keeping, including work history and exposure data.

Protective procedures should include: the prompt clean-up of spilled material; prevention of leaks and regular maintenance of pollution-control equipment; proper protective clothing (Ketcham and Norton, 1960); provision of washing facilities, with time allowed for their use; specific and detailed operating instruction and work practices for preventing hazardous exposures, particularly during shut-down, start-up and vessel and tank clean-out; thorough worker education on the potential hazards of the hydrogenation process; and regular and frequent monitoring of hazardous materials in work areas.

Another important concern is one of educating plant and regulatory personnel to occupational and environmental problems which are caused by release of hazardous materials from coal processing. The most effective control device available still is turned on or off by a human being whose judgement is subject to error. Chemical plant managers and operators are used to periodic upset conditions with consequent discharges to the environment. Their major concern is to return the plant to productive operation, sometimes without sufficient concern for the types and quantities of emissions. There is a need to build into both the design and operation of such plants a consciousness of minimizing or eliminating the environmental release of any stream which has a reasonable potential for containing toxic substances.

Considerable effort should be expended to review, monitor and protect the occupational health of plant workers, particularly in view of the lack of any long-term studies on occupational health effects (NIOSH, 1978b). Retrospective epidemiological investigations on identified groups of workers at pilot or commercial-sized installations where medical records exist (pre- and post-employment medical examinations) would be extremely useful in trying to better delineate long-term health implications. Continuous monitoring of pollution levels in the worker, user and community setting, as well as perspective epidemiological evaluations and the development and maintenance of pertinent disease registries on these populations, is a necessary concomitant to responsible development of coal resources.

Control of pollutants from an established technology is often difficult and costly. It is much more desirable to identify and quantify hazards and develop appropriate characterization and control methods before a new technology is launched. Designers, operators and regulators must be aware and alert to the problems of coal hydrogenation in order to properly protect the health of the worker and the public.

5. References

ACGIH (1980). *Documentation of the Threshold Limit Values*, 4th ed. American Conference of Governmental Industrial Hygienists, Cincinnati.

AMDUR, M. O. (1976). Toxicological guidelines for research on sulfur oxides and particulates. 4th Symposium on *Statistics and the Environment*, National Academy of Sciences, Washington, D.C.

AVOL, E. L., JONES, M. P., BAILEY, R. M., CHANG, N. N., KLEINMAN, M. T., LINN, W. S., BELL, K. A. and HACKNEY, J. D. (1979). Controlled exposures of human volunteers to sulfate aerosols. *American Review of Respiratory Diseases*, **120**, 319–27.

BOMBAUGH, K. J., LEE, K. W., OLDHAM, R. G. and KAPOR, S. (1981). Characterization of process liquids and organic condensates from the Lurgi coal gasification plant at Kosovo, Yugoslavia. *EPA Symposium, Environmental Aspects of Fuel Conversion Technology, VI*, Denver.

BROWN, R. and WITTER, A. (1979). *Health and Environmental Effects of Coal Gasification and Liquefaction Technologies.* A Workshop Summary and Panel Reports for the Federal Inter-Agency Committee on the Health and Environmental Effects of Energy Technologies. Department of Energy Report DOE/HEW/EPA-03, MTR-79W00137, Mitre Corporation, McLean, Virginia.

CLARK, E. L. (1979). Coal gasification—the state of the art. *Symposium: Ammonia from Coal.* Tennessee Valley Authority Bulletin Y-143, Muscle Shoals, Alabama.

CLAUSEN, J. F. and ZEE, C. A. (1981). Modderfontein Koppers-Totzek sources test results. *Symposium Proceedings: Environmental Aspects of Fuel Conversion Technology, V.* EPA Report No. EPA-600/9-81-006.

CLELAND, J. G. and KINGSBURY, G. L. (1977). *Multimedia Environmental Goals for Environmental Assessment, Vols. I and III.* EPA Report No. EPA-600/7-77-136a.

COMMITTEE ON SCIENCE AND TECHNOLOGY (1976). *Community Health and Environmental Surveillance System (CHESS): An Investigative Report.* U.S. House of Representatives, 94th Congress, Washington, D.C.

COWSER, K. E., EPLER, J. L., GEHRS, C. W., GUERIN, M. R. and KLEIN, J. A. (1981). Health and environmental studies of H-coal process. *EPA Symposium, Environmental Aspects of Fuel Conversion Technology, VI*, Denver.

DOLL, R., FISHER, R. E. W., GAMMON, E. J., GUNN, W., HUGHES, G. O., TYREE, F. H. and WILSON, W. (1965). Mortality of gasworkers with special reference to cancers of the lung and bladder. *British Journal of Industrial Medicine*, **22**, 1–12.

DOLL, R., VESSEY, M. P., BEASLEY, R. W. R., BUCKLEY, A. R., FEAR, E. C., FISHER, R. E. W., GAMMON, E. J., GUNN, W., HUGHES, G. O., LEE, K. and NORMAN-SMITH, B. (1972). Mortality of gasworkers: final report of a prospective study. *British Journal of Industrial Medicine*, **29**, 394–406.

EPA (1974). *Health Consequences of Sulfur Oxides: A Report from CHESS, 1970–71.* EPA Report No. EPA-650/1-74-004.

EVANS, J. M. and VERDEN, M. L. (1982). Safety considerations for synfuel plants. *Chem. Eng. Prog., April*, 69–71.

FISHER, H., HAMILTON, L. D., MORRIS, S. C., MOSKOWITZ, P. D., NAGY, J. and ROWE, M. D. (1983). *Health Effects of Conventional Energy Sources; A Health and Environmental Effects Document.* Brookhaven National Laboratory, Upton, N.Y.

FRANZEN, J. E. and GOEKE, E. K. (1976). Gasify coal for petrochemicals. *Hydrocarbon Processing*, **55** (11), 134–8.

FREIREICH, E. J., GEHAN, E. A., RALL, D. P., SCHMIDT, L. H. and SKIPPER, H. E. (1966). Quantitative comparison of toxicity of anti-cancer agents in mouse, rat, hamster, dog, monkey and man. *Cancer Chemotherapy Reports*, **50**, 219–44.

GHASSEMI, M., CRAWFORD, K. and QUINLIVAN, S. (1978). *Environmental Assessment Data Base for High Btu Gasification Technology: Vol. II, Appendices A, B and C.* US Environmental Protection Agency Report EPA-600/7-78-186b, Washington, D.C.

GHASSEMI, M., CRAWFORD, K. and QUINLIVAN, S. (1979). *Environmental Assessment Report: Lurgi Coal Gasification Systems for SNG.* US Environmental Protection Agency Report EPA-600/7-79-120, Washington, D.C.

GIBSON, E. D. and PAGE, G. C. (1979). Low/medium Btu gasification: A summary of applicable EPA regulations. In *Symposium: Ammonia from Coal.* Tennessee Valley Authority Bulletin Y-143, Muscle Shoals, Alabama.

HAGGIN, J. (1982). Interest in coal chemistry intensifies. *Chem. Eng. News*, 9 August, 17–26.

HAMILTON, L. D. (1984). Health and environmental risks of energy systems. In *Risks and Benefits of Energy Systems*, IAEA, Vienna.

HARRIS, L. R., GIDEON, J. A., BERARDINELLI, S., REED, L. D., DOGGIN, R. D., EVANS, J. M., TELESCA, D. R. and TAMITA, R. K. (1980). *Coal Liquefaction: Recent Findings in Occupational Safety and Health.* NIOSH Special Technical Report DHHS (NIOSH), No. 80–122.

HOGAN, T. J. (1982). Validity of predicting human no-effect dosages from non-human indicators of toxicity. Ph.D. Thesis, University of Illinois, Chicago.

KETCHAM, N. H. and NORTON, R. W. (1960). The hazards to health in the hydrogenation of coal, III. The industrial hygiene studies. *Archives of Environmental Health*, **1**, 194–207.

KINGSBURY, G. L. and WHITE, J. B. (1979). *Multimedia Environmental Goals for Environmental Assessment, Vol. III. MEG Charts and Background Information Summaries.* EPA Report No. EPA-600/7-79-176a.

KOH, K. K. (1982). What's happening with coal gasification technology? *Hydrocarbon Processing, June 1982*, 85–8.

KONKOL, W., RUPRECHT, P., CORNILS, D. R. and LANGHOFF, J. (1982). Autothermal coal gasification. *Hydrocarbon Processing, March 1982*, 97–102.

KUHN, A. K. (1982). The Great Plains gasification project. *Chemical Engineering Progress, April 1982*, 64–8.

LAVE, L. B. and SESKIN, E. P. (1977). *Air Pollution and Human Health.* John Hopkins University Press, Baltimore.

LEE, K. W., SEAMES, W. S., COLLINS, R. V., BOMBAUGH, K. J. and PAGE, G. C. (1981). *Environmental Assessment Report: Source Test and Evaluation Report—Lurgi-type (Kosovo, Yugoslavia) Medium-Btu Gasification.* Radian Corporation, DCN 81-218-001-69.

MACEK, J., PRASEK, K. and SIMANEK, K. (1976). Ecological aspects of gas production from high-sulphur lignite. Paper IGU/87-76, 13th World Gas Conference, London.

MORGAN, M. G., MORRIS, A. K., MEIER, A. K. and SHENK, D. L. (1978). A probabilistic methodology for estimating air pollutant health effects from coal-fired power plants. *Energy Systems and Policy*, **2**, 287–309.

MORRIS, S. C. (1983). Health risks from coal and energy technologies. In *Health Risk of Energy Technologies* (ed. by C. C. Travis and E. L. Etnier). AAAS Symposium Series No. 82, Westview, Boulder.

MURIN, P., SIPES, T. and PAGE, G. C. (1980). *Environmental Assessment Report: Wellman-Galusha Low-Btu Gasification Systems.* Environmental Protection Agency Report No. EPA-600/7-80-093.

NAS (1972). *Particulate Polycyclic Organic Matter.* National Research Council Committee on

Biologic Effects of Atmospheric Pollutants, National Academy of Sciences, Washington, D.C.

NIOSH (1978a). *Recommended Health and Safety Guidelines for Coal Gasification Pilot Plants.* National Institute for Occupational Safety and Health (78-120), EPA Report No. 600/7-78-007.

NIOSH (1978b). *Criteria for a Recommended Standard. Occupational Exposures in Coal Gasification Plants.* National Institute for Occupational Safety and Health, Publication 78-191, Cincinnati, Ohio.

NIOSH (1981). *Occupational Hazard Assessment. Coal Liquefaction Vols. I & II.* National Institute for Occupational Safety and Health, 81-131 and 81-132.

PAGE, G. C., CORBETT, W. E. and MAGEE, R. A. (1981). Application of Kosovo (Lurgi) gasification plant test results to pollution control process design. *EPA Symposium, Environmental Aspects of Fuel Conversion Technology, VI,* Denver.

PAGE, G. C. (1978). *Environmental Assessment: Source Test and Evaluation Report—Chapman Low-Btu Gasification.* EPA Report No. EPA 600/7-78-202.

PALMER, A. (1979). Mortality experience of 50 workers with occupational exposures to the products of coal hydrogenation processes. *Journal of Occupational Medicine,* **21,** 41-4.

SCHLESINGER, R. B., LIPPMAN, M. and ALBERT, R. E. (1978). Effects of short-term exposures to sulfuric acid and ammonium sulfate aerosols upon bronchial airway function in the donkey. *Journal of the American Industrial Hygiene Association,* **39,** 275-85.

SCHLINGER, W. G. and RICHTER, G. N. (1980). Process pollutes very little. *Hydrocarbon Processing, October,* 66-70.

SEXTON, R. J. (1960a). The hazards to health in the hydrogenation of coal, I. An introductory statement on general information, process description, and a definition of the problem. *Archives of Environmental Health,* **1,** 181-6.

SEXTON, R. J. (1960b). The hazards to health in the hydrogenation of coal, IV. The control programme and the clinical effects. *Archives of Environmental Health,* **1,** 208-31.

SHABAD, L. M. and DIKUN, P. P. (1959). *Air Pollution by Carcinogenic Substances: 3,4-Benzpyrene.* Medghiz, Leningrad.

SHABAD, L. M., KHESINA, A. Ya. and KHITROVO, S. S. (1968). On carcinogenic hydrocarbons in the exhaust gases of motor car engines and their possible elimination. *Vestnik Akademii Meditsinski Nauk USSR,* **23**(6), 12-20.

SINOR, J. E. (1979). Reconsider coal conversion processing. *Hydrocarbon Processing,* **58**(7), 159-62.

STANTBERG, P. S. (1982). Parsons developing largest Stretford unit in US. *Hydrocarbon Processing, June,* 11.

THOMAS, W. C., TREDE, K. N. and PAGE, G. C. (1979). *Environmental Assessment: Source Test and Evaluation Report—Wellman-Galusha (Glen Gary) Low Btu Gasification.* US Environmental Protection Agency Report EPA-600/7-79-185, Washington, D.C.

WADDEN, R. A. (1976). Coal hydrogenation and environmental health. *Environmental Health Perspectives,* **14,** 201-6.

WADDEN, R. A. and TRABERT, A. L. (1981). Potential health problems from coal conversion technologies. In *Proceedings of Seventh National Conference on Energy and the Environment* (ed. by J. P. Reynolds, W. N. McCarthy and L. Theodore). Ann Arbor Science.

WEIL, C. S. and CONDRA, N. I. (1960). The hazards to health in the hydrogenation of coal, II. Carcinogenic effect of materials on the skin of mice. *Archives of Environmental Health,* **1,** 187-93.

WILLIAMSON, P. C. (1981). The TVA ammonia from coal project. In *Symposium Proceedings: Environmental Aspects of Fuel Conversion Technology, V.* EPA Report No. EPA 600/9-81-006.

WOLFF, R. K., SILBAUGH, S. A., BROWNSTEIN, D. G., CARPENTER, R. L. and MAUDERLY, J. L. (1979). Toxicity of 0.4 and 0.8 μg sulphuric acid aerosols in the guinea pig. *Journal of Toxicology and Environmental Health,* **5,** 1037-47.

ÖZKAYNAK, H., SPENGLER, J. D., THURSTON, G. D., GARSD, A., TOSTESON, T. D., COLOME, S. D., SMITH, C. M., BURBANK, B., EVANS, J. S. and SKORNIK, W. (1985). *Analysis of Health Effects Resulting from Population Exposures to Ambient Particulate Matter.* HERAP, US Department of Energy, Washington D.C.

11

The Effect of Coal Utilization Emissions on Natural and Man-managed Terrestrial and Freshwater Ecosystems

1. Introduction 282
2. Atmospheric Transport and Diffusion 284
3. Effects on Forest Ecosystems 289
    3.1. Effects of SO_2 and NO_x 289
        3.1.1. Effects on community composition 289
        3.1.2. Effects on tree species 290
        3.1.3. Interactions between air pollutants and other stress factors 292
        3.1.4. Combined effects of two or more pollutants 293
        3.1.5. Effects on components of forest ecosystems other than trees 293
    3.2. Effects of Acid Precipitation 294
        3.2.1. Effects on tree growth 294
        3.2.2. Effects on nutrient cycling 295
4. Effects on Grassland Ecosystems 297
    4.1. Effects on Community Composition 298
    4.2. Effects on Individual Grass Species 300
    4.3. Interactions between SO_2 and NO_x and Other Factors 301
5. Effects on Other Natural and Semi-natural Ecosystems 302
6. Effects on Agricultural Crops 303
    6.1. Effects of SO_2 and NO_x 303
    6.2. Effects of O_3 303
7. Effects of Air Pollutants from Coal Conversion Processes 304
8. Contamination of Ground and Surface Water by Coal Conversion Solid Waste 305
9. Bioaccumulation of Toxic Trace Elements from Coal Conversion Solid Wastes 306
10. Some Beneficial Effects of SO_2, NO_x and their Derivatives 306
11. Effects of Acid Precipitation on Freshwater Ecosystems 307
    11.1. Effects on Fish 308
    11.2. Effects on other Freshwater Organisms 309
12. Conclusions 310
13. References 311

1. Introduction

The environmental impacts of increased coal utilization are primarily the result of the emission of air pollutants arising from combustion processes, or coal gasification or liquefaction plants. These pollutants may be distributed over a relatively large area, whose extent depends upon the design and mode of operation of the combustion or conversion process. Liquid and solid wastes arising from coal utilization generally represent more localized problems. These are discussed along with the effects of coal mining, treatment or transport in other chapters.

The pollutants traditionally associated with coal combustion are sulphur dioxide (SO_2) and smoke (particulates). Modern coal combustion systems are designed to avoid the continuous discharge of particulates or else, in the case of large plants, to eliminate them from flue gases almost entirely by electrostatic precipitation, cyclonic devices or bag filters. In consequence, increased coal utilization should not result in serious smoke pollution problems. However, the control of SO_2 emissions has proved less feasible in technological or economic terms, and thus SO_2 must be considered a potential environmental hazard, particularly in view of the concerns arising from transboundary dispersion. Recently attention has become focused on nitrogen oxides (NO_x), produced during coal combustion (Chapter 5). 95 % of the NO_x emitted from coal-fired power stations is in the form of nitric oxide, which becomes oxidized rapidly in the air to nitrogen dioxide (NO_2) (CENE, 1981). Small amounts of nitrous oxide (N_2O) arise from combustion processes, although most atmospheric N_2O is of natural biological origin. N_2O emissions constitute a potential global problem in view of their possible role in stratospheric ozone (O_3) depletion and in their contribution to the "greenhouse effect". With distance from the source, both SO_2 and NO_2 become increasingly oxidized in the atmosphere. Both dry and wet deposition occur eventually on the Earth's surface. Wet deposition, mainly in aerosol form, occurs by in-cloud (rainout) and below-cloud (washout) scavenging.

Other emissions of significance from coal utilization include carbon dioxide (CO_2), trace metals (including radionuclides), discussed in Chapter 8, and organic compounds resulting from incomplete combustion, discussed in Chapter 9. CO_2 emissions have implications that are globally significant and potentially serious. CO_2 is the most important of a number of gases, present in low concentrations in the atmosphere, that are transparent to incoming solar radiation but absorb infrared radiation that is emitted by the Earth's surface. These so-called "greenhouse" gases will affect global temperatures as man-induced concentration changes occur in the future, thus giving rise to climatic changes. CO_2 is particularly subject to change due to the combustion of fossil fuels by man. Current global emissions of carbon are approximately 5 Gt, in the form of CO_2 (Marland and Rothy, 1984; Rothy and Masters, 1984). The assessment of the scale of future CO_2 emissions depends on how future energy use patterns develop (Ausubel and Nordhaus, 1983; Edmonds and Reilly, 1983a, 1983b; Marland, 1982; Mintzer and Miller, 1984; Nordhaus and Yohe, 1983; Perry, 1982; Reister, 1984; Rothy and Masters, 1984).

As well as uncertainties about the scale of future CO_2 emissions, there are also features concerning the buffering capacities of various environmental compartments that need to be resolved (Edmonds *et al.*, 1984; Siegenthaler, 1983). These uncertainties combine to produce a wide range of predictions concerning CO_2-induced warming from the various climate models that have been employed to assess possible effects (Schlesinger, 1983; Seidel and Keyes, 1983). Smagorinsky (1982) provides a synoptic account of the contemporary consensus. Fuller discussions are provided by Bach (1984) and Jäger (1983).

Coal is being substituted for oil in a range of industrial activities (EFN, 1984) and has been projected to increase its share of OECD primary energy requirements from 21 % to 31 % by the end of the century (IEA, 1982). The various coal conversion processes which are being considered for future large-scale operations are not only likely to produce SO_2 and NO_x, but also a number of other gaseous sulphur and nitrogen compounds, particularly carbonyl sulphide (COS), methyl mercaptan (CH_3SH), carbon disulphide (CS_2), hydrogen sulphide (H_2S), and ammonia (NH_3) (Gehrs *et al.*, 1981; Cleland, 1981). The environmental effects of these are largely unknown, but it is expected that under steady state operation raw product gas can be treated to allow sulphur and nitrogen compounds to be removed (Gas Processing Handbook, 1979).

Spectacular impacts on ecosystems of major sources of SO_2 are well documented, with massive destruction of vegetation, severe acute injury to foliage and obvious changes in community composition. However, most of these are the result of emissions arising from the smelting of sulphide ores. The effect of emissions from increased coal utilization are likely to be of a more subtle type, which may not be immediately detectable, but have the potential to cause chronic injury to organisms and substantial ecosystem modification in the long term.

It is now clear that the greatest threat from SO_2, NO_x and their derivates is to natural and semi-natural ecosystems, particularly grasslands, forests and freshwater habitats. In more intensively managed systems, modification of agricultural practices and the breeding of tolerant crop cultivars offer the potential for ameliorating some of the adverse impacts of air pollution. Treshow (1968) pointed out that, while much was known at that time about the sensitivity of agronomic and major forest species to air pollutants, far less was understood concerning the response of non-crop species and plant communities as a whole. Today this situation is little changed. Furthermore, secondary impacts of air pollutants on plant communities, as mediated by effects on other components of the ecosystem, such as plant diseases, herbivorous insects and decomposer organisms, have scarcely been studied. This review, while covering the impacts on individual species and groups of species, also attempts to interpret these, so far as is possible, within an ecosystem context.

2. Atmospheric Transport and Diffusion

The main motion of pollutants in the atmosphere is defined primarily by wind speed and direction. Turbulent variations of wind velocity cause pollutants emitted at a single point to spread out diffusively in a way not easily predicted. In order to determine the relationships between sources and receptors, mean wind velocities and atmospheric turbulence require adequate characterization. This can be complicated by several problems. For instance, much finer spatial and temporal resolution is needed to determine maximum impacts near a point source than to calculate monthly average impacts from sources at a distance of 100 km. Spatial and temporal scales also determine the distinction between mean and turbulent flows. Thus, in considering seasonal-average long-range transport, atmospheric eddies comparable in size to high and low pressure synoptic features may be considered as turbulent variations about a mean uni-directional flow. On a daily basis or for shorter distances, motion about these synoptic features should be considered as the deterministic part of the flow with the occurrence of turbulence on a smaller scale.

Commonly available meteorological data include upper air winds, temperature and humidity as a function of altitude and surface-based measurements. In the USA, upper air data are obtained twice a day at stations approximately 400 km apart. Surface-based data are available hourly with about 100–150 km spatial resolution. Micrometeorological parameters related to turbulence and diffusion are not generally directly measured and must be inferred empirically from quantities such as wind speed and cloud cover. Although it is widely believed that the structure of the meteorological networks imposes fundamental constraints upon the accuracy with which source-receptor relations can be calculated, such limitations have not been fully quantified. The way in which available meteorological data are used in air quality transport models introduces another level of approximation.

The scales of transport for pollutants are conventionally divided into categories: short-range, mesoscale and long-range (regional). These scales correspond approximately to 0–50 km, 20–200 km and 1200–2000 km respectively. Most research on advection and dispersion of pollutants on these scales has been confined to the planetary boundary layer (PBL) and, in particular, to fair weather conditions. These data are, therefore, most relevant to the understanding and modelling of dry deposition (fair weather conditions).

Short-range transport of pollutants has been of concern for several decades. Previous work has centred on the prediction of maximum concentrations using local meteorological data.

In the case of mesoscale transport, the variability of winds over the relevant time period and distance requires consideration. Problems in describing flow on this scale arise, since the features of interest are not within resolution of the rawinsonde (radio tracking) network. Often an accurate meteorological description of the PBL is required. This is especially so for complex flow conditions such as mountainous terrain and the presence of large urban areas or coastlines. In some circumstances, especially in the mesoscale problems mentioned above, the limiting factor in describing transport is a physically realistic theoretical framework rather than the adequacy of meteorological data.

Often at night a momentum boundary layer (at approximately 100 m in height) forms which acts to decouple the air flows above and below. Ground-based measurements are then inadequate for the description of the speed and direction at which materials emitted from taller stacks are transported. This complicates the determination of source-receptor relations. In addition, pollutants released at ground level are potentially subject to dry deposition, while those at higher altitudes cannot mix down to the ground level in less than a 24-hour period. The preferential removal of pollutants at night from low altitude emission sources is thought to be the primary reason for variations in deposition with stack height. Improvements in the description of nocturnal transport, especially those that lead to boundary-layer climatology, are needed to evaluate the effects of stack height and also to evaluate enhanced transport due to nocturnal jets.

Descriptions of the transport and diffusion of plumes over several hundred kilometres in fair weather and for flat terrain have been derived from the results of well-constructed field experiments. The extent to which these results are representative of other meteorological conditions and other locations requires further study. A limited number of studies have been completed in which tracers have been followed in the atmosphere for long distances. Results from these experiments showed horizontal errors of approximately 25% of the distance which the tracer travelled compared with model calculations. It is unclear whether these discrepancies result from the approximations of the models or are fundamentally related to the spatial and temporal sparseness of data used as inputs to these models.

Recent developments in the detection of perfluorocarbons at very low concentrations allow a new generation of long-range tracer experiments. In 1982 feasibility measurements in the USA on a Cross Appalachian Tracer Experiment (CAPTEX) demonstrated that tracers released in Ohio could be detected in New York State. Further experiments are planned.

The transport and diffusion of pollutants under cloudy and precipitating conditions is complex and has therefore received less attention to date than fair weather phenomena. The increased difficulties arise for several reasons:

the motion of air through clouds and over fronts is three dimensional necessitating specification of a vertical wind velocity;

a large fraction of precipitation (even in frontal storms) results from convective activity. The dimensions of convective cells and rain bands are too small for rawinsonde network resolution;

the passage of fronts is attended by wind shifts not spatially or temporally resolved by the rawinsonde network;

the motion and diffusion of air in and through storms is not adequately characterized in terms of the quantities necessary for a description of aqueous phase chemical reactions and deposition.

A practical consequence of these difficulties in describing the complex flow in storm conditions is that source-receptor relations, as deduced from commonly available meteorological information, are likely to be identified less accurately than those for fair weather. The calculation of three-dimensional wind fields at present requires the use of computationally intensive meteorological models. These have not yet been shown practical in air pollution applications.

Beside the description of transport in individual storm events there is a need for general information on the air motions associated with convective and frontal storms in order to determine source-receptor relations. Time and distance data for the emission of pollutants to cloud level are needed in order to determine the impacts of local sources in comparison with long-range transport. The in-cloud residence times of pollutants must be considered in evaluating the extent of chemical reactions within clouds. This, in turn, may have a significant bearing on the linearity of chemical reactions. Ventilation of boundary-layer air through fair weather cumulus clouds deserves further attention because of aqueous phase reactions which remove SO_2 and produce H_2SO_4.

The distance over which pollutants are transported depends largely upon rate of removal by deposition. Deposition can occur by dry and wet mechanisms. Dry deposition of gases and small aerosol particles can occur when turbulent air motions bring pollutants within a few millimetres of the surface. Diffusion, impaction and/or phoretic effects can then take the material to the surface. Uptake of pollutants at the surface depends on surface characteristics as well as the chemical and physical behaviour of the pollutant. In wet deposition material reaches the ground through various forms of precipitation. Both the dry and wet deposition processes are significant.

Dry deposition is conventionally expressed in terms of a deposition velocity; the ratio of the flux of material deposited to the airborne concentration. Deposition velocities have been determined for a variety of pollutants, atmospheric conditions and surfaces. A characteristic of these measurements is their variability in both time and place in response to differing atmospheric and surface characteristics. The variability attributed to atmospheric turbulence can be understood in terms of micro-meteorological theory (at least for stationary flows and homogenous terrain). Variability due to surface characteristics and pollutant type is at best only qualitatively understood.

Experiments to determine deposition velocities are intrinsically difficult and require careful site selection and instrumentation with rapid response times and high precision. SO_2 has been the most studied of those pollutants related to acid deposition and in Europe a comprehensive monitoring programme is in existence (EMEP, 1981). Agreement between measurements made under similar conditions is in the "factor of 2" range. There are important uncertainties in the deposition velocity of aerosol sulphate with measured values ranging from near zero to values comparable to that found for SO_2. Deposition velocities for many pollutants such as NO, NO_2, PAN, organic nitrates, NH_3 and HNO_3 have not been measured or have only recently received attention.

In assessing the adequacy of experimental and theoretical data on dry deposition velocities, two end uses of the velocities should be noted. Firstly, the dry deposition velocity when multiplied by an air concentration of the corresponding substance gives the flux of material to the ground—a quality needed for assessment of environmental impacts. Secondly, dry deposition is a major removal mechanism of pollutants from the atmosphere and as such is a controlling factor in the transport of acidic pollutants. A detailed description of the variability of deposition velocities with surface property is needed for the first application. For the second application, regional average quantities may suffice. It is unclear whether the data needed for even the second application are available. For the second application it may be important to distinguish between average deposition velocities during night and day. Night-time deposition could deplete material from the shallow lowest stable layer of the atmosphere. This depletion would occur preferentially for emissions from low level sources and might cause significant stack-height effects.

The focus of atmospheric research on wet deposition has shifted from characterization to questions relating to mechanistic interpretation of the incorporation of pollutants in precipitation. Such work is now progressing based on earlier work (relating to the scavenging of radionuclides and to cloud physics), but there remain many uncertainties concerning the qualitative features of precipitation scavenging.

Among the atmospheric processes thought to be important for the incorporation of pollutants in precipitation are:

formation of cloud droplets around pre-existing aerosol particles;
incorporation of aerosol particles into existing precipitation elements;
diffusion of gases into and out of hydrometeors with possible aqueous phase chemical reactions.

The latter two processes can take place in clouds or in falling precipitation below-cloud. These processes are "reversible" in that evaporation of hydrometeors puts pollutants back into the gas phase, possibly in a chemically altered form. Incorporation of aerosol particles into hydrometeors is closely linked with the various processes responsible for the creation of clouds and precipitation and hence has been a subject of extensive research.

3. Effects on Forest Ecosystems

Smith (1974) has categorized the relationship between air pollution and forest ecosystems into three classes: *Class I*, where vegetation and soils, although acting as an important sink for low levels of pollutants, suffer no harm or may even experience a beneficial effect through improved nutrition; *Class II*, where individual trees are adversely but subtly affected by moderate levels of pollution, mediated via such impacts as reductions in photo-synthesis or reproductive rate, or increased sensitivity to disease and insect infestation, the net effect being manifested as reduced productivity and species composition; *Class III*, where severe pollution results in acute injury or death of individual trees, leading eventually to a major simplification of the structure of the ecosystem, with disturbances that may include secondary effects on hydrology, erosion rates and local climate.

This section will concentrate on Class II relationships, which are probably the most important with respect to increased coal utilization, while Class I relationships will be discussed in Section 10. Class III relationships, of the type observed in the vicinity of large smelters, are not particularly relevant here in view of the adequacy of modern control techniques for the prevention of severe pollution problems close to a source.

3.1. Effects of SO_2 and NO_x

3.1.1. Effects on community composition

Four localized studies have been made recently into the impact of chronic levels of SO_2 (in two cases accompanied by NO_x) on forest composition in North America. McClenahen (1978) measured changes in species composition of a mixed mesophytic forest, in the Ohio River Valley, subjected to SO_2 and NO_x from coal-burning power plants and other industrial sources. A general decline was demonstrated in the density of canopy and herb layer species with increasing pollutant exposure, accompanied by a rise in sub-canopy and shrub species density. It was postulated that, because the canopy trees experienced the greatest exposure to pollution, they suffered increased mortality, thereby permitting an increase in the under-storey vegetation, while species diversity was depressed in the herb layer. Thus changes in the forest ecosystem were attributed to both the primary impact of air pollution and secondary effects arising from changes in competition. Rosenberg, Hutnik and Davis (1979) carried out a similar type of study in a mixed oak forest in central Pennsylvania, subjected for 25 years to an isolated source of SO_2 (and presumably NO_x) in the form of a coal-burning power plant. It was shown that the importance of *Pinus strobus* and *Betula lenta* increased with distance from the emission source, while that of *Quercus alba* and *Acer rubrum* decreased, these effects being more pronounced downwind than upwind of the power plant. In contrast to the study of McClenahen (1978), the herb, shrub and sub-canopy layers appeared more sensitive than the canopy trees.

The other two studies were performed around isolated natural gas refineries in Alberta, where SO_2 is the only pollutant of any importance. Winner and Bewley (1978) found a fall in the number of *Picea glauca* seedlings close to the source, suggesting an inhibitory effect of SO_2 on reproduction. Cover of both bryophyte and under-storey vascular plants decreased as SO_2 stress increased. There was some evidence that grasses increased in abundance in areas of high SO_2 pollution, where they invaded stands of SO_2-sensitive species. Legge (1980) demonstrated that the basal area of *Pinus contorta* × *banksiana* fell as a 14-year old SO_2 source was approached, accompanied by alterations in nutrient cycling within the forest, but was unable to detect any serious irreversible degradation of the ecosystem.

These studies indicate the possibility of substantial, but not immediately obvious, changes being induced in forest ecosystems by gaseous pollutants derived from coal combustion. It is difficult to make any generalized predictions from this limited number of investigations, in view of an absence of reliable air pollution monitoring in the study areas, the presence of other pollutants (e.g. fluorides and chlorine) in the case of McClenahen (1978), and a lack of consistency in the changes observed. Little is known about the time-course for these shifts in forest composition, but, as Kozlowski (1980) has pointed out, the subtle and gradual effects of a relatively low level of pollution may cause an eventual critical imbalance in the structure of an ecosystem, resulting in a subsequent, rapid deterioration.

An attempt has been made to predict changes in forest composition over 500 years of succession, induced by chronic SO_2 stress, using a model based on the sensitivity of 32 tree-species native to eastern North America (West, McLaughlin and Shugart, 1980). This suggests that the response of trees to air pollution may be very different under the competitive conditions prevailing in a forest stand, from that predicted on the basis of fumigations of individuals or single species: species of high or intermediate tolerance to SO_2 may experience growth stimulation in polluted forests, as a result of the weakening of competition from more susceptible species. Although the general conclusions of West, McLaughlin and Shugart (1980) concerning the nature of shifts in forest composition may be valid, the changes predicted for individual species must be viewed with suspicion: the model utilizes data on the sensitivity of individual species to acute injury produced by high concentrations of SO_2, whereas it has recently been demonstrated that this bears little relationship to chronic injury of tree species (Garsed and Rutter, 1982a, 1982b).

3.1.2. Effects on tree species

Many studies have been conducted into the response of individual tree species to SO_2 either on the basis of fumigation experiments or field surveys.

Knabe (1970, 1976) and Farrar, Relton and Rutter (1977) surveyed the occurrence of *Pinus sylvestris* in the Ruhr region of Germany and the Southern Pennine hills in England, respectively. This species was absent in those areas of the Pennines where the Winter mean SO_2 concentration exceeded 200 $\mu g\ m^{-3}$, and of the Ruhr where the annual mean exceeded the same value, with a threshold for injury of the foliage of 80 $\mu g\ m^{-3}$ in the latter region. It is difficult to ascribe causality in species presence-absence studies, but it is noteworthy that a recent decline in SO_2 levels in the southern Pennines has been accompanied by a marked improvement in local conifer trials which formerly showed very poor growth (Lines, 1979).

Attempts to determine a threshold concentration for effects of SO_2 on tree growth are fraught with problems arising from interactions with other environmental factors, the difficulty of defining pollution characteristics in terms of continuously fluctuating concentrations and the invariable presence of other phytotoxic pollutants. Materna, Jurgle and Kucera (1969) and Materna (1973) examined the performance of conifers in the Erzgebirge mountains of Czechoslovakia and found accelerated needle loss of isolated trees, but no overall growth reductions, where the annual mean SO_2 concentration was 25–50 $\mu g\ m^{-3}$, moderate growth reductions at 50–100 $\mu g\ m^{-3}$ and substantial depression of growth, accompanied by severe visible injury at $> 100\ \mu g\ m^{-3}$. These are greater effects than have been reported for similar SO_2 levels around urban areas, and it has been suggested (IERE, 1981) that this is explained by a greater frequency of peak concentrations $> 1000\ \mu g\ m^{-3}$ in the Erzgebirge. However, Garsed, Mueller and Rutter (1982) have shown that in a fumigation of *Pinus sylvestris* with a mean of 100 $\mu g\ m^{-3}$ SO_2, growth reduction was primarily dependent on the overall mean rather than on peaks within this of up to 750 $\mu g\ m^{-3}$ and work by Jones and Mansfield (1983) with *Phleum pratense* supports this conclusion.

Most fumigation experiments are of a relatively short duration, usually less than one year, while the impact of SO_2 on a tree may occur throughout its lifetime in the field. Thus there are dangers in extrapolating to the field from short-term fumigations. Keller (1977) has introduced the concept of "latent injury", where air pollution has an unfavourable impact on plant metabolism which may be manifested in growth reductions or other more obvious effects at a later date. This concept has subsequently been verified by a number of workers: Keller (1978a) fumigated *Fagus sylvatica* seedlings over Winter with 300 $\mu g\ m^{-3}$ SO_2 and found a reduction in the number of terminal buds which developed in the Spring, after the fumigation had terminated. Garsed, Mueller and Rutter (1982) found no effect of 100 $\mu g\ m^{-3}$ SO_2 on *Pinus sylvestris* over one year, but on continuing the fumigation for a second year, significant growth reductions took place. This latent effect has recently been confirmed in the field by Garsed and Rutter (1982b), who exposed five conifer species outdoors for two years in the centre of London and at an unpolluted rural site: over the first year all species grew better in London, reflecting higher temperatures in an urban area, but by the end of the second year growth was significantly reduced compared with the rural site. The mean SO_2 concentration in central London throughout the experiment was only 79 $\mu g\ m^{-3}$, although the effect of other pollutants cannot be discounted. $SO_2 : NO_2$ in central London is 1:0.7 suggesting a concurrent concentration of 55 $\mu g\ m^{-3}$ of NO_2.

IERE (1981) has recently concluded that the long-term threshold for SO_2 effects on yield lies between 130 and 150 $\mu g\ m^{-3}$. However, the field exposure and fumigation experiments of Garsed and Rutter (1982a, 1982b) suggests that 80–100 $\mu g\ m^{-3}$ over a 2-year period would be a more reasonable estimate. Furthermore, there are reports of injury to tree species by even lower concentrations: Houston (1974) recorded visible symptoms after fumigation of sensitive *Pinus strobus* clones with 70 $\mu g\ m^{-3}$ SO_2, while Keller (1981) has found reduced photosynthesis in *Picea excelsa* fumigated with 65 $\mu g\ m^{-3}$ over Winter. In general, deciduous trees are much less susceptible to SO_2 than conifers, as demonstrated by Garsed, Farrar and Rutter (1979) who fumigated *Betula pendula*, *B. pubescens*, *Acer pseudoplatanus* and *Quercus robur* with 150–170 $\mu g\ m^{-3}$ SO_2 for periods ranging from 14 to 71 weeks and found no deleterious effects except for a 12-day advance in leaf fall in the latter species. Currently the International Union of Forest Research Organizations (IUFRO) recommends that, for the maintenance of full forest productivity at most sites, annual and 24-hour mean SO_2 concentrations should not exceed 50 and 100 $\mu g\ m^{-3}$ respectively (Scholz, 1981; Wentzel, 1983).

3.1.3. Interactions between air pollutants and other stress factors

There is increasing evidence of marked interactions between low temperatures and injury by low SO_2 concentrations. Thus a fumigation of *Fagus sylvatica*, *Picea abies* and *Pinus nigra* under Winter conditions resulted in increased sensitivity to frost in the following Spring (Keller, 1978b), while 65 $\mu g\ m^{-3}$ SO_2 for 3 months in Winter enhanced frost injury on *Picea excelsa* (Keller, 1981). Spaleny (1979) has made similar observations in the field: in the Erzgebirge, SO_2 injury appears first on trees growing on mountain ridges where very severe climatic conditions are experienced in Winter; these trees proved to be significantly less frost-hardy than those growing in similar but unpolluted locations. Several workers (Huttunen, Havas and Laine, 1981; Huttunen, Karenlampi and Kolari, 1981; Soikkeli and Tuovinen, 1979) have also noted the close connection between pollution-induced injury on conifers and the effects of Winter stress factors, such as cold and drought, in central and northern Finland. Clearly the impact of air pollution arising from coal combustion is likely to be most serious in areas with a severe climate in Winter. This is recognized by IUFRO, who recommend a reduction in the maximum SO_2 concentrations to 25 and 50 $\mu g\ m^{-3}$ annual and 24 hour means, respectively, in order to maintain full forest productivity at such sites.

There is some evidence that clones of conifer species which are tolerant to drought also display tolerance to SO_2 injury (Braun, 1977; Klein, 1980). This raises the possibility that wild conifer populations growing in areas subject to regular water stress may have developed incidentally some degree of tolerance to SO_2 accompanying selection for drought resistance.

3.1.4. Combined effects of two or more pollutants

High temperature coal combustion always produces substantial quantities of NO_x as well as SO_2. Fowler and Cape (1982) have suggested that in rural areas of western Europe mean NO_2 concentrations exceed mean SO_2 levels by about 25 % (by mass). Thus, it is important to consider the combined effects of both NO_x and SO_2 when predicting the impact of coal combustion on ecosystems. Unfortunately, the impact of ambient concentrations of NO_x are only just beginning to be studied in detail. It is known, however, that SO_2 and NO_2 mixtures can produce either synergistic, antagonistic or additive effects on plants, depending upon the species and gas concentrations (Reinert, Heagle and Heck, 1975). Recent work has demonstrated synergistic effects with relatively low concentrations of the two pollutants ($166\,\mu g\,m^{-3}\,SO_2 + 118\,\mu g\,m^{-3}\,NO_2$) in reducing growth of *Alnus incana*, *Betula pendula*, *B. pubescens* and *Populus nigra* following a 150-day fumigation throughout the growing season (Whitmore and Freer-Smith, 1982).

Another important pollutant which may have interactions with SO_2 and NO_x on plant growth is ozone (O_3). This is generated photochemically from reactions between NO_x and hydrocarbons and occurs episodically at phytotoxic levels over wide areas of Europe and North America (Fowler and Cape, 1982; Bell, 1984). The possibility of interactions of O_3, SO_2 and NO_x with respect to phytotoxicity must be considered, but few investigations have been carried out. However, Mooi (personal communication—quoted in Bell, 1984) has recently demonstrated a remarkable impact on *Populus* × *interamericana* of adding 0.031 ppm (v/v) O_3, a concentration within the natural background, to a mixture of $60\,\mu g\,m^{-3}$ SO_2 and $43\,\mu g\,m^{-3}\,NO_2$: this resulted in a 44 % increase in leaf fall induced by the pollutant mixture over the controls.

3.1.5. Effects on components of forest ecosystems other than trees

Almost nothing tangible is known about the effects of SO_2 and NO_x on components of the forest ecosystem other than the trees. Knabe (1976) has reported that mycorrhizal fungi disappear or are reduced substantially in SO_2 polluted areas. Alstad, Edmunds and Weinstein (1982) suggested that trees are more sensitive to air pollutants than their associated insect herbivores and that pollution may increase the effects of such forest pests. In general, observations on changes in forest insect populations are of a circumstantial nature and have been made in very severely polluted areas, categorized as

Class III by Smith (1974). However, Hughes, Potter and Weinstein (1982) have recently demonstrated that *Glycine max* plants fumigated with $524\,\mu g\,m^{-3}$ SO_2 supported Mexican bean beetle larvae that grew faster and larger than on control plants. In addition, Dohmen, McNeill and Bell (1984) have shown in fumigation experiments dramatic stimulatory effects of SO_2 ($34–145\,\mu g\,m^{-3}$) and NO_2 ($20–100\,\mu g\,m^{-3}$) on the performance of *Aphis fabae* on *Vicia faba*. Changes in performance of the insect population were mediated through effects of the gases on the host plant. It is plausible that insect herbivores in forest ecosystems may be affected similarly by moderate levels of SO_2. Fungal pathogens of forest trees may either be stimulated or inhibited by SO_2 depending on circumstances. Observations were made by Grzywacz and Wazny (1973) on the abundance of a range of pathogenic fungi, including *Armillaria mellea* and *Fomes annosus*, in a Polish forest subjected to a strong point source of SO_2: all species were greatly reduced close to the source but showed an increase with distance up to a maximum in the presence of moderate levels of SO_2 ($100–450\,\mu g\,m^{-3}$) and then declining to some extent as air quality improved further. In contrast, other fungal pathogens are inhibited at moderate SO_2 concentrations: for example, *Rhytisma acerinum* on *Acer pseudoplatanus* were absent at sites with mean levels $> 90\,\mu g\,m^{-3}$ (Bevan and Greenhalgh, 1976).

3.2. Effects of Acid Precipitation

Outside urban and industrial areas, dry and wet deposition of SO_2 and its derivatives are approximately equal in magnitude except in areas of very high precipitation (IERE, 1981). Thus in more remote areas much of the impact of increased air pollution from coal combustion will be caused by acid precipitation. Natural rainwater has usually been assumed to have a pH of 5.6 and acidification to below this level is generally considered to be the result of man-made emissions, although recent evidence has suggested that unpolluted rain may reach pH 4.5 on occasions due to fluctuations in the natural sulphur cycle (Charlson and Rodhe, 1982).

3.2.1. Effects on tree growth

The possibility that increasing concentrations of sulphuric and nitric acids in precipitation are reducing conifer growth was first raised by Jonsson and Sundberg (1972) on the basis of a dendrochronological study in Sweden. A subsequent similar exercise by Abrahamsen, Horntvedt and Tveite (1977) in different areas of southern Norway did not demonstrate any relationship between tree growth and the pH of precipitation. Cogbill (1977) was also unable to find any consistent decreases in tree growth in the eastern USA which could be attributed to acid deposition. Clear indications of a long-term decrease in growth accompanying increasing acidity in precipitation were reported by Johnson *et al.* (1981). Tree ring analyses showed an abnormal reduction in growth of *Pinus rigida, P. echinata* and *P. taeda* over the last 25 years on upland soils in New Jersey which was correlated with a fall in the pH of stream water in the area. Johnson and Siccama (1983) and Tomlinson (1983) review other evidence of growth reduction in the north-east USA, particularly in red spruce (*Picea rubens*). It is worth noting that oxidant concentrations in the north-east USA will almost certainly have increased over the period of these studies, thus confounding the impacts (if any) of acid precipitation.

Many experiments have been conducted in which trees and other plant species are subjected to various doses of acidity in artificial rain. Usually these have employed inputs of acid which are well above those experienced in the field. Both stimulatory and inhibitory action of acid precipitation have been observed with low pH rain. Tamm and Winklander (1980) showed a decrease in the growth of *Pinus sylvestris* on plots fertilized with nitrogen, potassium and phosphorus but a stimulation on unfertilized plots; however, the acid applications were given over a 5-year period as single annual applications at pH 1.9 (IERE, 1981), which is clearly unrealistic for the field situation. A recent review by IERE (1981) concludes that "both stimulations and depressions of growth appear to be limited to rather extreme acid applications, certainly below pH values of 3.0, and there is no substantive evidence of a growth effect at the acid concentrations presently found in rain" (i.e. a median pH of about 4.2 in the most severely affected areas of North America and Scandinavia). Despite the lack of clear evidence of growth reductions on trees caused by ambient levels of acidity in rain, such effects have recently been demonstrated in experiments on agricultural crops (see Section 6).

3.2.2. *Effects on nutrient cycling*

The net impact of acid precipitation on forests will eventually only be determined by whole ecosystem studies in view of the many components of the system which are potentially susceptible. Thus, while low pH rain may have direct effects on foliage, changes in rain chemistry due to passage through the canopy and soil litter layer have to be considered, as well as possible effects on the soil which may modify ionic uptake by roots, soil biological processes (such as decomposition and nitrification) and the rate of mineral weathering (Tamm, 1976; Lee and Weber, 1982).

Acid rain has been shown consistently by many workers to increase the rate of leaching of cations from foliage (IERE, 1981): a decrease in pH from 6 to 4 results in a doubling of the loss of Ca and Mg from leaves (Horntvedt, Dollard and Joranger, 1980). Indirect impacts on foliage are also possible via effects on fungal pathogens: little is known about such effects, but Shriner (1977, 1978) has shown both stimulation and inhibition by artificial rain of pH 3.2 of a range of fungal pathogens, depending on the time of application in the parasite's life cycle.

Soil acidification reduces microbial growth and activity, but different types of soil differ considerably in their response to low pH rain (Evans *et al.*, 1981). Application of pH 2.7–3.1 rainfall increases the decomposition rate of the needle litter of *Pinus* spp. (Abrahamsen, Hovland and Hagvar, 1980; Roberts *et al.*, 1980), but effects of ambient levels of acidity remain uncertain. In contrast, IERE (1981) notes that humus decomposition rates tend to decrease with increased acidity within the range pH 3.5–4.5. Nitrification in soils is particularly sensitive to a reduction in pH as has been demonstrated by Hovland and Ishac (1975); lowering soil pH, however, would reduce N losses by leaching and increase its availability to roots in the form of NH_4^+ (Evans *et al.*, 1981). It has been concluded by Evans *et al.* (1981) that a change in the pH of the already acidic soils of coniferous forests in response to acid precipitation is likely to be so slow that it cannot readily be detected in the field. Nevertheless, long-term effects on microbial processes within the soil may ultimately influence nutrient cycling within the forest ecosystem with possible consequent changes in tree productivity.

Much attention has been paid to the possible impacts of acid precipitation on the availability of nutrient and toxic cations in the soil. Increased leaching of Ca, Mg and Al can be demonstrated when water of pH 3.5 or less is applied to soils (IERE, 1981). It has been suggested that in most coniferous forest soils, impacts of this nature will be minimal because further acidification is only likely if their pH is higher than that of the received rain (Bache, 1980). However, two recent publications provide evidence that changes in the exchangeable base content of forest soils are occurring in response to acid precipitation. Extensive field surveys in central Sweden have demonstrated a decline in exchangeable Ca^{2+}, Mg^{2+} and K^+ in forest soils between 1963 and 1973 (Troedsson, 1980). Particular attention has become focused on work in the Solling Highlands, FRG, an area subject to large inputs of H^+ from the atmosphere: over a period of 13 years there has been a fall in soil pH of 0.3 units accompanied by a substantial increase in soluble Al. Evidence has been produced demonstrating reduced Ca : Al molar ratios in seepage water at 90 cm depth in a forest soil. Other work emphasizes that under certain conditions strong acids in precipitation may be largely neutralized prior to the rain water's infiltration (into mineral soil by the forest canopy and litter layers of the soil (Richter, Johnson and Todd, 1983).

Seedlings of *Picea abies* have been shown to exhibit reduced root lengths at low Ca : Al ratios (Ulrich, 1982; Rost, 1982). Ulrich, Mayer and Khanna (1980) have suggested that mature *Fagus sylvatica* stands in the Solling area are also being damaged as a result of toxic effects of Al on the roots, which is exacerbated by increased release of this metal in hot dry summers when excess H^+ is produced by stimulation of nitrification processes. Gehrmann and Ulrich (1982) recently reported the results of an investigation into the regeneration of *F. sylvatica* seedlings in polluted areas: symptoms of Al toxicity were observed, with death of fine roots, but this could be alleviated by the addition of lime. Currently this is the subject of an intensive investigation and it is claimed that acid inputs to German forests may soon result in Al damage to tree roots over up to 2×10^6 ha (Pearce, 1982). Such claims raise the alarming possibility of very serious impacts on forest ecosystems, on a scale larger than hitherto considered.

The possibility of such serious impacts has led to concerted efforts to obtain accurate estimations of forest damage in certain countries. Foremost in undertaking such surveys has been the Federal Republic of Germany where considerable effort has been put into standardizing visual estimates of damage. Based on a 4 km grid, it was estimated that 3.7 m ha of forest were subject to visible damage in 1984. This represented 50 % of all forests, compared with 35 % showing damage in 1983 and 8 % in 1982. Fir (*Abies*) showed the highest incidence of damage (87 %) with spruce (*Picea*) at 51 % and pine (*Pinus*) at 59 %. It is emphasized that not only does the area suffering damage appear to be increasing, but so does the severity of damage.

A survey of needle loss by Spruce in 1984 in Sweden suggested 29 % of stands (by area) showed light damage (21–60 % needle loss) and 1 % was classed as badly damaged (61–80 % needle loss). In the UK a recent forest survey failed to detect significant injury (Binns *et al.*, 1985).

Such surveys do not give proof of the cause of such injury. The ozone-sulphur dioxide hypothesis has been given support by Bosch *et al.* (1983). It was suggested that O_3 acting in conjunction with SO_2 damages leaf cell membranes (and causes decreased frost resistance). As demonstrated by Prinz, Krause and Stratmann (1982) this results in accelerated leaching of Mg from spruce needles. The needle chlorosis followed by necrosis, so common now in certain forests in Central Europe, is a common symptom of Ca and Mg deficiency.

4. Effects on Grassland Ecosystems

Many permanent grasslands, particularly in upland areas, are subjected to minimal agricultural practice and thus represent a second major semi-natural man-managed ecosystem which is liable to damage by air pollutants arising from increased coal combustion. Unlike forests, grasslands can change in structure very rapidly as a result of intra- and inter-specific competition, thereby possessing the ability to adapt more readily in response to environmental stress. However, as in the case of forests, most species in a grassland are perennial and will be exposed to air pollution over many years, thus hindering meaningful extrapolations to the field from short-term fumigation experiments. Overall, grasslands appear to be less susceptible than forests which they invariably replace under the very severe SO_2 stress of Class III situations (Smith, 1974).

4.1. Effects on Community Composition

Bleasdale (1957) stated that "nothing is known of the effects of (air) pollution on the species composition of grasslands", but suggested that plants with storage organs would be at an advantage compared with those which relied on current assimilation for survival over the Winter, when SO_2 concentrations are usually at a maximum. Since then, little progress has been made in this field, at least with respect to chronic injury, although a considerable body of information has built up concerning the effects of SO_2 and, to a lesser extent, NO_2 on individual grass species.

Guderian (1966) described the results of an intensive study on the effects of SO_2 on competition and productivity within simple communities of forage plants. He fumigated plots of mixtures of two to four species with acute doses of SO_2 and measured changes in species composition at an unspecified later date (Table 11.1). In many cases substantial changes took place, with *Trifolium* species proving particularly sensitive and often being effectively eliminated from the sward. Reductions in the final total dry weight of the plots were not always, however, accompanied by changes in the relative proportions of the species concerned. This study indicates that interspecific variation in tolerance to SO_2 could lead to major shifts in the composition of grasslands in polluted areas primarily through changes in competition rather than direct elimination of species. The SO_2 doses administered (Guderian, 1966) were, however, very severe and unrepresentative of conditions likely to result from increased coal combustion. It is difficult to make realistic predictions on the basis of this work for community changes in response to chronic injury as Horsman *et al.* (1979) and Ayazloo and Bell (1981) have shown a marked lack of correlation between the response of grass species to acute and chronic SO_2 fumigations. Recently, however, it has been shown that *Trifolium* species are generally more sensitive than grasses to chronic SO_2 injury (Awang, 1979;

TABLE 11.1. *Changes in composition of forage species mixtures following acute SO_2 fumigations (examples from Guderian, 1966)*

Species	SO_2 ($\mu g\ m^{-3}$)	Duration of fumigation (h)	% cover		% reduction in total dry weight of plot
			Before fumigation	After fumigation	
Lolium multiflorum	1200	48	37	55	28
Vicia villosa			41	44	
Trifolium incarnatum			22	1	
Lolium multiflorum	5000	10	82	18	45
Trifolium pratense			99	1	
Vicia faba	1000	48	40	39	34
Lupinus luteus			17	20	
Pisum arvense			28	29	
Vicia sativa			15	12	

Bell, 1980); thus, it can be postulated that clovers may be reduced in abundance in SO_2 polluted areas, with a consequent deterioration in grassland quality.

Taylor and Leininger (1978) reported the results of an experiment in which a natural grassland in Montana was subjected over three growing seasons to SO_2 levels ranging between 60 and 210 $\mu g\ m^{-3}$, administered via an open-air fumigation system. Changes were recorded in plant community structure but they are very difficult to interpret: at one plot increasing SO_2 appeared to result in a decline in the importance of grasses compared with other herbaceous species, while at another similar plot the opposite situation occurred.

The Montana study is the only example of a controlled fumigation with SO_2 under field conditions where the response of all components of the ecosystem have been examined. A reduction in myccorhizal fungi, associated with grass rhizomes, occurred in response to SO_2, although the significance of this for grass productivity was unclear (Rice *et al.*, 1978). Litter decomposition rates in a mixed-grass prairie were reduced by 32–44 % in Spring and early Summer by atmospheric SO_2 levels of 200 $\mu g\ m^{-3}$ (Dodd and Lavenroth, 1981). Incorporation of sulphur into the substrate had no detectable effect.

Most invertebrate groups show no response to SO_2 with the exception of ground beetles of the genus *Canthon* which were reduced in numbers even at a mean of 60 $\mu g\ m^{-3}$ (Leetham, McNary and Dodd, 1978). Grasshopper species showed a significantly reduced density late in the growing season each year in the presence of all SO_2 concentrations (McNary *et al.*, 1981). This could influence the flow of carbon into the grassland litter layer (Leetham, McNary and Dodd, 1978). No very clear trends were detected in subterranean animals: no effects were seen on mites, with variable responses in nematodes (Leetham, McNary and Dodd, 1978). A mark and recapture exercise indicated a decrease in both mice and voles in all SO_2 treatments (Chilgren, 1978). Neither the mechanisms of these changes nor their significance for the ecosystem as a whole are understood. However, this work demonstrates the numerous subtle but complex interacting impacts that relatively low SO_2 levels can exert upon an ecosystem and highlights the need for further such integrated studies on a range of terrestrial habitats.

Currently very little is known about the amount of genetic variation in resistance to air pollutants which exists within natural populations before exposure to pollutants (Roose, Bradshaw and Roberts, 1982). However, recent studies have shown that several grass species have the ability to select for tolerance in response to both chronic and acute SO_2 stress in both mixed populations and monocultures. Horsman, Roberts and Bradshaw (1979) demonstrated that *Lolium perenne* populations in polluted areas of Liverpool were more tolerant to SO_2 than populations from clean areas. Ayazloo and Bell (1981) discovered the development of SO_2 tolerance in *L. perenne*, *Holcus lanatus*, *Phleum bertolonii*, *Festuca rubra* and *Dactylis glomerata* populations at one or more of three polluted sites in northern England where mean SO_2 levels were between 100 and 200 $\mu g\ m^{-3}$. Furthermore, Bell, Ayazloo and Wilson (1982) have shown that such tolerance can develop in 4–5 years' exposure to a mean of about 120 $\mu g\ m^{-3}$ SO_2. This phenomenon has several important implications. Firstly, it is clear that SO_2 must be exerting a deleterious effect at these relatively low levels in order for selection to occur. Secondly, that given sufficient time, the adverse impact of SO_2 on a grassland will be ameliorated to some extent by the development of an increasing level of tolerance; however, it should be noted that often, but not always, SO_2-tolerant plants show poorer growth in clean air than sensitive individuals. Finally, it seems probable that inherent interspecific differences in the ability to select for tolerance may contribute to shifts in species composition of grasslands in polluted areas.

4.2. Effects on Individual Grass Species

Considerable difficulty has been experienced in establishing a minimum long-term threshold concentration of SO_2 for the reduction of growth in grass species (Bell, Rutter and Relton, 1979; Mansfield and Freer-Smith, 1981; Bell, 1982). In particular, controversy has arisen over the interpretation of experiments in which plants have been grown in moderately polluted areas in chambers ventilated with either ambient or charcoal-filtered clean air. Table 11.2 gives the results of a number of experiments of this type carried out in a suburb of Sheffield by Crittenden and Read (1978, 1979) and Awang (1979). Substantial reductions in growth occurred in the ambient air chamber

TABLE 11.2. *Effects of ambient air on shoot dry weight of grass species in chamber experiments in Sheffield, UK*

Species	Mean SO_2 concentration ($\mu g\ m^{-3}$)	Duration (days)	% reduction in ambient cf. clean air	$p \leqslant$	Reference
Lolium perenne	70	56	36	0.001	Crittenden and Read (1978)
	59	131	20	0.01	
	69	86	25	0.001	
	63	116	26	0.001	
Lolium multiflorum	67	56	36	0.001	Crittenden and Read (1979)
Dactylis glomerata	45	72	42	0.001	
Lolium perenne	44	28	14	0.05	Awang (1979)[a]
	38	28	25	0.05	
Lolium multiflorum	44	28	14	0.05	
	38	28	23	0.05	
Dactylis glomerata	44	28	39	0.01	

[a] Total dry weight.

where mean SO_2 levels were between 38 and 70 $\mu g\ m^{-3}$. These concentrations are generally lower than have been shown to cause yield reductions in the same species in fumigations with SO_2 (Mansfield and Freer-Smith, 1981; Bell, 1982). A probable explanation of this discrepancy is the continuous presence of NO_x and intermittent occurrence (in Summer) of O_3 in the ambient air of Sheffield which may have contributed to the observed suppression of growth. This is supported by reports of fumigations with SO_2 and NO_2 mixtures within the range 166–194 $\mu g\ m^{-3}$ and 118–139 $\mu g\ m^{-3}$ respectively producing either additive or synergistic effects in reducing the growth of several grass species (Ashenden and Mansfield, 1978; Whitmore and Freer-Smith, 1982). Another factor which may have enhanced the effects of SO_2 in the work at Sheffield is that the experimental conditions in some cases resulted in exceptionally slow growth, a circumstance which appears to increase greatly the susceptibility of grasses to chronic SO_2 injury (Bell, Rutter and Relton, 1979; Davies, 1980; Jones and Mansfield, 1982). A third factor which needs to be taken into account is the change in sensitivity to dose with time. Whitmore and Freer-Smith (1982) subjected *Poa pratensis* to 177 $\mu g\ m^{-3}$ SO_2 in outdoor chambers for 11 months. Destructive sampling from October onwards showed that early resistance to the prevailing SO_2 concentration had given way by January to considerable reductions in dry weight, reaching 64% in March. The adverse effect of SO_2 then declined again into the Summer and in September a 17% stimulation of dry weight was recorded. Colvill *et al.* (1983) demonstrated a similar effect for *Lolium perenne*.

Estimations of a threshold dose for chronic SO_2 injury to grass species are tentatively put at between 70 and 180 $\mu g\ m^{-3}$ respectively by Mansfield and Freer-Smith (1981) and IERE (1981). This demonstrates the uncertainties in this field.

4.3. *Interactions between SO_2 and NO_x and Other Factors*

Recent work has highlighted a number of important interactions between SO_2 and various environmental factors on the performance of grass species. It is now clear that SO_2 injury is greatly enhanced by low light intensities and low temperatures (Davies, 1980; Jones and Mansfield, 1982) and thus grasslands in which plants overwinter in a green state are particularly at risk. Furthermore, as in the case of tree species (see Section 2.1.3), there is now good evidence that SO_2 increases the sensitivity of Gramineae to frost damage: Davison and Bailey (1982) showed that a fumigation with 250 $\mu g\ m^{-3}$ SO_2 for 5 weeks reduced both the survival rate and subsequent growth of *Lolium perenne* plants which were later subjected to an artificial freezing regime. Baker, Unsworth and Greenwood (1982) provide support for this on the basis of field observations by showing that wheat plants grown in an open-air fumigation system in the presence of 250–1500 $\mu g\ m^{-3}$ SO_2 showed substantially more leaf injury (attributed to cold-stress) after a period of severe Winter weather than control plants growing in ambient air (*c.* 50 μg m^{-3} SO_2).

Many investigations on the effects of SO_2 on grasses have failed to examine the roots after fumigation. This is unfortunate, because it is becoming apparent that chronic levels of SO_2 often reduce the growth of roots to a much greater extent than the shoots (Bell, 1982; Jones and Mansfield, 1982). Under field conditions this could have a serious effect on grassland productivity by increasing susceptibility to drought or reducing the potential for regrowth following defoliation by grazing or mowing or damage by trampling.

IERE (1981) has pointed out that little information is available on the influence of wet foliage on SO_2 injury which might be important in many temperate regions where moist conditions prevail for much of the time. In fact, Elkiey and Ormrod (1981) have demonstrated a large increase in visible injury produced by SO_2, NO_2 or their mixture on *Poa pratensis* plants subjected to a brief daily misting regime.

Clearly, predictions of the impact of increased coal utilization must take account of not only the effect of environmental factors on pollution sensitivity but also the impact of pollutants on other environmentally induced stresses on vegetation.

5. Effects on Other Natural and Semi-natural Ecosystems

Little is known about the impact of SO_2 and NO_x derived from coal combustion on natural or semi-natural terrestrial ecosystems other than forests and grasslands. Hill *et al.* (1974) and Thompson, Kats and Lennox (1980) fumigated a range of species common in deserts of the western USA with SO_2 and NO_2. Even in the presence of an abundant water supply permitting maximum stomatal opening there was generally a very high level of tolerance to both pollutants. Thus, increased coal utilization in such areas is likely to be much less harmful to natural vegetation than in temperate regions.

One major temperate ecosystem for which there is good evidence of changes resulting from coal combustion is the blanket bogs of the southern Pennine hills in northern England (see also effects on trees in this area—Section 2.1.2). These plant communities are located in close proximity to the industrial cities of Manchester and Sheffield and have been subjected to coal-smoke pollution for the last 200 years. This pollution was accompanied by major changes in the species composition of the blanket bogs, notably the near disappearance of the formerly extensive cover of *Sphagnum* species. Although various anthropogenic factors, such as burning and increased stocking with sheep, have been suggested as contributing to this change, there is now good evidence that air pollution is strongly implicated. Ferguson, Lee and Bell (1978) and Ferguson and Lee (1979) demonstrated respectively that a fumigation with 131 μg m^{-3} SO_2, or the addition of solutions of bisulphite at concentrations which occur in southern Pennine rain, reduced the growth of six *Sphagnum* species;

however, *S. recurvum*, the only species still present to any extent in the area, proved more resistant than the other species tested. Subsequently these findings have been confirmed by Ferguson and Lee (1980) in a field trial where bisulphite solutions were applied to natural populations of *Sphagnum* species at an unpolluted site. This work indicates that ecosystems in which *Sphagnum* species are a major component may be particularly at risk from increased coal combustion.

6. Effects on Agricultural Crops

6.1. Effects of SO_2 and NO_x

Recently there has been a serious attempt to quantify the reduction of agricultural crop yields by acidic depositions (Irving, 1983; Loucks and Armentano, 1982). Crop response to increasing dose is generally non-linear and not always easy to interpret (Irving, 1983). Evans, Conway and Lewin (1980) showed that small reductions in seed weight of *Glycine max* occurred at pH 4.0 and growth inhibition of *Beta vulgaris* was noted at the same pH along with the appearance of leaf lesions (Evans *et al.*, 1982). Further experiments on *Glycine max*, using simulated acidic rain, showed decreased seed yields at pH 4.1, 3.3 and 2.7 compared to pH 5.6. These reductions were due to a decrease in the number of pods per plant (Evans *et al.*, 1983). These experiments were conducted outdoors and an interaction between ambient pollutants may have occurred. Combined impacts of acid precipitation and pollutants in other forms have not been intensively studied, but should be taken into account when trying to interpret field studies and make predictions. Irving and Miller (1981) subjected *Glycine max* to a mean of 570 $\mu g\ m^{-3}$ SO_2 and/or artificial rain adjusted to pH 3.1 with nitric and sulphuric acids: the acid rain increased seed dry weight, possibly due to a nutritional effect of extra nitrogen or sulphur, while SO_2 reduced seed yield; there was no significant effect of the mixture, the two effects appearing to balance out. Norby and Luxmoore (1983) exposed *Glycine max* to simulated acid rain (pH 2.6 – 5.6) and SO_2 and O_3. Growth inhibition resulted only from rain of pH 2.6, due to physical injury to the leaves, and there was no significant pH × air pollution interactions.

6.2. Effects of O_3

In the USA the National Crop Loss Assessment Network (NCLAN) has carried out an extensive assessment of crop dose-response data of O_3 exposure. Again (see Section 6.1), yield-dose relationships were not found to be linear, but reductions due to 0.04–0.06 ppm seasonal 7 h day^{-1} mean exposures (compared to 0.025 ppm) were found for a number of crops. *Glycine max* and *Arachis hypogea* appeared to suffer the greatest yield reductions, closely followed by *Gossypium*, with certain cereals showing considerably smaller reductions (Heck *et al.*, 1983).

Troiano *et al.* (1983), using *Glycine max*, found that the presence of O_3, when simulated acid rain was administered, gave increased reductions in vegetative yield and also reduced the reproductive mass, compared to simulated acid rain treatments in the absence of O_3.

7. Effects of Air Pollutants from Coal Conversion Processes

There is still much to be learned about the impacts on ecosystems of nitrogen and sulphur gases such as H_2S, COS, CS_2, CH_3SH and NH_3. These may be emitted, under certain conditions of start-up, shut-down or accidents, from large-scale coal conversion plants (Shriner, McLaughlin and Taylor, 1982; Taylor *et al.*, 1983).

Taylor *et al.* (1983) have studied the flux of H_2S, COS, CH_3SH and CS_2 to *Phaseolus vulgaris*, *Glycine max* and *Lycopersicon esculentum*. Deposition velocities of the different gases varied considerably between the three species, but in all cases were lower than those recorded for SO_2, overall being ranked as $SO_2 > H_2S > COS > CH_3SH > CS_2$. Kluczewski *et al.* (1983) have shown that COS at a very low concentration (6.5 $\mu g\ m^{-3}$) is taken up by *Lolium perenne* at a rate approximately three times greater in the light than in the dark, and that the sulphur component can subsequently be translocated to the roots. Thus COS enters pathways within the plant and so has the potential to cause metabolic injury. The phytotoxicity of COS has been put at a threshold value of 800 $\mu g\ m^{-3}$ for chronic injury (Kotcon, 1976), but more recent work (Taylor and Selvidge, 1984) suggests a value of only half this.

Some information is available on H_2S effects on plants. Thompson and Kats (1978) have demonstrated reductions in growth of *Vitis vitifera*, *Lactuca sativa* and *Medicago sativa* following prolonged fumigation with 417 $\mu g\ m^{-3}$ H_2S, where 139 $\mu g\ m^{-3}$ either stimulated growth or had no effect. The consensus of opinion is that on a volume/volume basis H_2S is generally less toxic than SO_2 (Thornton and Setterstrom, 1940; Thompson, Kats and Lennox, 1979). Gehrs *et al.* (1981) note that stack concentrations of H_2S are not expected to exceed 13,900 $\mu g\ m^{-3}$ and that under *normal* operation sufficient dilution should occur to prevent phytotoxic effects on plants. However, De Kok, Thompson and Mudd (1982) have reported inhibition of various crop plants at 43–430 $\mu g\ m^{-3}$ H_2S, and work by Taylor and Selvidge (1984) on *Phaseolus vulgaris* confirms this. If adverse effects are produced at the lower end of this range, it represents a ten-fold reduction in the previous suggestion for the threshold for injury and raises the possibility that H_2S emissions from coal conversion processes might damage local vegetation. There are two reports of the effects of adding O_3 to a fumigation with H_2S. Coyne and Bingham (1978) showed an increased reduction in net photosynthesis of *Phaseolus vulgaris* when 0.07 ppm O_3 was added to either 4518 or

6992 $\mu g\ m^{-3}$ H_2S. Bennett, Barnes and Shinn (1980) fumigated the same species with various combinations of O_3 (0.05–0.13 ppm) and H_2S (417–9730 $\mu g\ m^{-3}$) and found a greater reduction in yield in the presence of O_3 compared with H_2S alone.

A recent assessment of the environmental impacts of coal conversion pollutants by Gehrs *et al.* (1981) considers that NH_3 emissions are unlikely to be harmful to vegetation but that fugitive H_2S releases might cause damage. Nihlgård (1985), however, has suggested that forest dieback in Europe might be explained by high wet and dry deposition of ammonia and ammonium ions. Taylor and Selvidge (1984) have concluded that effects on vegetation of H_2S, SO_2 and COS are strongly dose related. An accurate assessment of effects in the field close to a commercial scale plant will eventually need to be made.

8. Contamination of Ground and Surface Water by Coal Conversion Solid Wastes

As well as residual organic components, solid wastes contain a variety of macro and trace elements, many of which are significantly enriched in comparison with coal feedstocks. Additionally, the mineralogic residence times of these elements and their susceptibility to leaching depends highly on temperature and atmospheric regimes attained during processing or combustion.

Generally, coal conversion wastes may be grouped into three mineralogic categories which qualitatively represent a temperature sequence: (1) low temperature pyrrhotite-like residues; (2) partially clinkered, medium temperature residues containing crystalline aluminium silicates and iron oxides; and (3) fused, high temperature, amorphic ferrous silicate residues. Of these wastes, the medium and high temperature residues are most comparable to conventional coal combustion wastes (fly ash and water quencher slag). The ultimate environmental impacts of coal conversion solid waste will depend on the relative distribution of these wastes. Leachate composition has been found to be largely determined by the chemical composition of the feedstocks used. Concentrations of Al, B, Ca, total Fe, Na, SO_4^{-2} and Zn were reduced as leachates passed through soil. Although the degree of reduction varied between soil types, those with the highest pH (7.1) and cation exchange capacity were most efficient in removing metals from solution (Griffin *et al.*, 1980). Tamura and Boegly (1980) observed that soil from a proposed coal gasification site attenuated Ni transport from gasifier ash (Tamura and Boegly, 1980). On the other hand, it has been found that soil from a proposed waste disposal site was not effective in lowering the concentration of sulphate in the leachate from gasifier ash (Francis *et al.*, 1981). This particular gasifier ash generated an acidic (pH 3.5–4.0) leachate containing very high concentrations of sulphate.

More emphasis has been given to inorganic components of solid wastes than organic components. A variety of polyaromatic and aliphatic compounds have been isolated from coal-derived solid wastes. In the case of SRC-I mineral ash residue, it has been shown that a significant portion of the organic components are readily leached by aqueous solutions (Browman and Maskarinec, 1981).

9. Bioaccumulation of Toxic Trace Elements from Coal Conversion Solid Wastes

Bioaccumulation in aquatic foodchains may result from surface and groundwater contamination following the disposal of solid wastes in landfills or ashponds. Bioaccumulation in terrestrial foodchains may result from revegetation of disposal sites (Bern, Neufeld and Shapiro, 1980). Terrestrial plants can accumulate elevated levels of B, Be, Ni, Cd, Zn and Co if grown on mixtures of coal gasification ash and soil. Yield data are not significantly different from those obtained from soil alone, indicating that, firstly, high concentrations of Zn, Ni and Cd are not phytotoxic and, secondly, significant quantities of these toxic trace elements could be transported to man through a grass-to-man foodchain. Such considerations are discussed in Chapter 8.

10. Some Beneficial Effects of SO_2, NO_x and Their Derivatives on Terrestrial Ecosystems

In any assessment of the net impact of increased coal utilization on terrestrial ecosystems it is necessary to take into account possible beneficial effects. Both sulphur and nitrogen are essential constituents of proteins and it has been shown that low levels of SO_2, H_2S, NO_2 and NH_3 can act as sources of these nutrients, particularly under conditions of deficiency in the soil, the gases being metabolized along normal pathways (Cowling and Koziol, 1982). In addition, the deposition of SO_4^{2-} and NO_3^- in acid precipitation may prove beneficial to sulphur and nitrogen deficient ecosystems (Evans *et al.*, 1981). Inputs of atmospheric nitrogen may be of significance for the productivity of natural and man-managed ecosystems which are not subjected to fertilizer additions. Evans *et al.* (1981) suggest that nitrogen in polluted rain may be particularly important in the nutrition of forests. Furthermore, acids in precipitation will increase the rate of weathering of soil minerals, thus releasing plant nutrients into solution, while in alkaline soils they may reduce the pH to an optimum for plant growth (Overrein, Seip and Tollan, 1980). Nutritional effects probably explain many of the observed cases of stimulation of plant growth in experiments with simulated acid precipitation, particularly when nitric acid is utilized.

Although sulphur deficiencies have been recorded in forest soils in North America remote from pollution inputs, in general, relatively small amounts of sulphur from the atmosphere will prove sufficient for the nutritional requirements of trees (Evans *et al.*, 1981). Sulphur inputs in fertilizers to agricultural land in Europe have been falling over the last 30 years, accompanied by a substantial and continuing increase in the application of nitrogen fertilizers (Bache and Scott, 1979). This trend has the potential to cause sulphur deficiency in vegetation which appears as the sulphur : nitrogen ratio falls below critical values (Murphy, 1978). There is now good evidence that in parts of Europe where the wet + dry deposition of sulphur from the atmosphere is below 15 kg ha^{-1} yr^{-1}, sulphur deficiency is present in some grasslands (Bell, 1984). Estimates based on field trials suggest that about 30 % of the area of productive grassland in the Republic of Ireland, a country with low atmospheric inputs, may experience some degree of sulphur deficiency (Murphy, 1979).

The net impact of the beneficial and deleterious effects of atmospheric N and S pollutants on terrestrial ecosystems is not understood at present, but must be quantified in order that the full implications of air pollution arising from increased coal combustion are appreciated.

11. Effects of Acid Precipitation on Freshwater Ecosystems

Concern about the adverse impacts of acid precipitation on natural ecosystems originated from reports of declining pH in southern Scandinavian lakes and a serious deterioration of fish stocks coincident with a period of increasing acidity of precipitation. This, and similar observations in the north-eastern USA and adjacent parts of Canada, has led to intensive investigations into the influence of acidic precipitation on freshwater ecosystems. A causal relationship between the acidity of precipitation and of lakes in areas with granitic or gneissic geology is now firmly established, although the possibility remains that changes in land-use, in particular replacement of farmland by coniferous forest, may also have contributed to acidification of freshwaters in some areas (Rosenqvist, 1978; Harriman and Morrison, 1980). There is now good evidence that lakes and streams have become acidified in both Europe and North America in areas which have the common features of precipitation pH < 4.6 and geology consisting of slow-weathering bedrock and thin soils deficient in buffering materials (Evans *et al.*, 1981). Although acid precipitation may, to some extent, influence the pH of lakes by direct deposition, in most cases its impact will be modified substantially by changes in the solution chemistry as a result of passage through the catchment area. This is illustrated by the Integrated Lake Watershed Study in the Adirondack Mountains in New York State: three lakes with catchments of similar geological formation and receiving similar direct inputs of atmospheric pollutants, but with different levels of acidity in the water (pH 4.5, 5.5–7.0 and 7.0), are being studied to determine the relationships between their water chemistry and acid precipitation. It has been concluded that the differences in lake acidity are accounted for primarily by the ability of the catchments to neutralize H^+ deposited from the atmosphere, this being low where the topography is steep and the soil shallow and high where there is a larger watershed and deeper soil (Johannes and Altwicker, 1980; Galloway *et al.*, 1980). At present, there is a considerable degree of uncertainty as to the relative importance of dry deposition of gaseous SO_2 and NO_x in contributing to lake acidity because of the difficulty in measuring this process (Overrein, Seip and Tollan, 1980; Evans *et al.*, 1981). However, an understanding of this is vital if meaningful predictions are to be made concerning the impact of coal combustion pollutants at different distances from the emission source.

Softwater lakes, unaffected by acid precipitation, are usually buffered by HCO_3^- and their pH is generally < 5.0. As the input of H^+ is increased this buffer becomes increasingly exhausted with very large but, to some extent, temporary falls in pH during periods of high runoff from the catchment as when snow-melt occurs in Spring. As the buffer is completely consumed, the pH falls below 5.0 and the levels of aluminium and some other metals rise rapidly (Wright and Henricksen, 1979). The effects of acid precipitation on lake chemistry in areas where the nature of the geological formation permits a higher degree of buffering capacity are unknown (Evans *et al.*, 1981), but the possibility must be considered that, given sufficient time, similar trends may be observed to those outlined above.

11.1. Effects on Fish

Fish population numbers are severely reduced, or they are eliminated altogether, from freshwater habitats when the pH falls below 4.8, but the sensitivity of different species varies, with the salmonids being particularly susceptible. Overrein, Seip and Tollan (1980) quote the following ranking of sensitivity on the basis of controlled experiments and field observations in Norway: *Salmo gairdneri* (rainbow trout), *S. salar* (salmon), *S. trutta* (sea trout and brown trout), *Perca fluviatilis* (perch), *Salvelinus alpinus* (char), *S. fontinalis* (brook trout), *Esox lucius* (pike) and *Anguilla anguilla* (eel). Although acidification affects lower trophic levels in freshwater ecosystems (see Section 7.2), elimination of fish stocks does not appear to result from changes in their food supply. Reproductive failure due to high mortality of eggs and fry is generally regarded as the prime cause of fish population losses (Rosseland *et al.*, 1980), although episodes occur when large numbers of more mature fish are killed, usually during the Spring when snow-melt results in a sudden inflow of low pH water.

High acidity impairs the osmotic balance of fish by interfering with the uptake of salts through the gills. However, toxicity is not controlled solely by pH but is also markedly dependent on a range of modifying factors, including water hardness, type of acid and activity of the fish (Graham and Wood, 1981). In areas where acidification has eliminated fish, salt concentrations, particularly calcium, in the water are generally low, and it is apparent that this is an important factor in causing physiological impairment and reductions in fecundity (Johansson, Kihlström and Wahlberg, 1973; Beamish *et al.*, 1975). Howells, Brown and Sadler (1983) conclude that at pH–4.5 lakes will usually be fishless where Ca concentrations are below 0.8 mg l^{-1}. For a given pH, fish mortality can be reduced by raising the calcium concentration in water (Leivestad, Muniz and Rosseland, 1980), and liming of natural waters has been employed on occasions to protect fisheries. Elevated aluminium levels, released during passage of acidic waters through the catchment, now appear to be a major factor in fish mortality. Aluminium causes a further disturbance of the osmotic balance of fish apparently by interfering with ion exchange at the gills as a result of stimulating excessive mucus production, thereby resulting in physical clogging (Overrein, Seip and Tollan, 1980). However, the chemical speciation of Al, which is strongly dependent on pH, is an important factor in its toxicity to fish. Maximum toxicity of Al occurs at pH 5, with little effect at pH 4.0. It is now considered that this explains the disappearance of fish populations from lakes with moderate levels of acidity which would not normally be expected to cause physiological stress (Leivestad, Muniz and Rosseland, 1980). Although complete elimination of fish has taken place in many areas subject to acid precipitation, screening experiments have demonstrated a high degree of genetic variability in response to low pH (Edwards and Gjedrem, 1979), which offers some prospect for breeding acid-tolerant populations. Whether or not a sufficient level of such tolerance can be achieved for the restocking of acidified lakes remains to be determined.

11.2. Effects on Other Freshwater Organisms

Acidification may cause a multitude of changes in freshwater ecosystems in addition to the impact on fish. The species composition of phytoplankton is often changed, with an overall decline in diversity as the more acid-sensitive species are eliminated (Renberg and Hellberg, 1982), but evidence concerning effects on total biomass is not consistent (Evans *et al.*, 1981). In many acid lakes and rivers an abundant growth of filamentous algae and mosses develops (Overrein, Seip and Tollan, 1980). In particular, dense mats of *Sphagnum* species may occur which can contribute further to the deterioration of the water quality as a result of their high capacity to exchange H^+ for basic cations in the lake water (Hendrey and Vertucci, 1980). Acid conditions result in a shift from bacterial to fungal decomposers which reduces nutrient cycling within the ecosystem, a situation which may be aggravated further by the *Sphagnum* mat physically restricting exchange between sediments and the overlying water (IERE, 1981). Both zooplankton and benthic invertebrates are simplified in species composition with increasing acidification, although total biomass does not always decrease (Evans *et al.*, 1981). The significance of such changes for the fish population is not certain. However, the sensitivity of both crustaceans and snails to declining pH may be of significance in moderately acidic lakes where direct toxic effects on fish do not take place: Økland and Økland (1980) have predicted that a fall in pH to 6.0 would effectively eliminate most of these organisms which represent 45% of the calorific intake of trout in Norwegian lakes, resulting in a 10–30% reduction in the trout population. Effects on other organisms (*Rana temporaria* and *Bufo bufo*), including egg shell thinning in some bird species, are gradually being documented (Hultberg, 1983).

On the other hand, a few species appear to be favoured by conditions in acidified waters, the Corixidae in particular increasing in abundance and importance in the diet of the surviving fish (Rosseland *et al.*, 1980).

12. Conclusions

The major impacts of increased coal utilization on ecosystems are likely to be in the form of injury by SO_2 and NO_x in the environs of the emission sources and damage by acid precipitation in more remote locations. In addition, a range of little studied pollutants, emitted by coal conversion processes, may have localized effects.

The main problems will arise from chronic impacts on ecosystems which may not be immediately obvious but could lead to more marked effects in the longer term. Currently very little is understood about the prolonged effects of moderate concentrations of SO_2, NO_x and their derivatives on the functioning of terrestrial ecosystems. Both forests and grasslands may be adversely affected over a prolonged period of SO_2 concentrations within the range 50–100 $\mu g\,m^{-3}$, particularly when accompanied by NO_x pollution. Gaseous pollutants derived from coal combustion are likely to represent the greatest hazard to ecosystems which are located in areas subject to severe Winter conditions: this may reduce the threshold concentrations for SO_2 injury by as much as 50%. Coniferous forests and *Sphagnum*-dominated communities are those terrestrial ecosystems which are probably at greatest risk although few studies have been made on the effects of pollutants on most other types of ecosystem. In grasslands there is evidence that the adverse effects of SO_2 and NO_x may be ameliorated by rapid selection for tolerance in grass species. Effects on ecosystem function via impacts on components other than higher plants are only now being elucidated, but reduction in decomposition rates and the surge in certain herbivorous populations are of considerable importance, even at relatively low levels of pollution.

The impact of air pollution on forests is a matter of grave concern, whatever its exact mechanism. Earlier predictions of reductions of conifer growth are only now being tested by extensive field surveys or experiments. There is now increasing concern that precipitation of pH 4.0–4.5 may be causing changes in soil chemistry which are approaching a threshold for severe and widespread damage to forests in parts of Europe. There now seems little doubt that increased coal combustion can be one cause of a fall in the pH of freshwaters in areas with sensitive geological formations with a multitude of adverse impacts including the widespread elimination of fish stocks.

13. References

ABRAHAMSEN, G., HORNTVEDT, R. and TVEITE, B. (1977). Impacts of acid precipitation on coniferous forest ecosystems. *Water, Air and Soil Pollution*, **8**, 57–73.

ABRAHAMSEN, G., HOVLAND, J. and HAGVAR, S. (1980). Effects of artificial acid rain and liming on soil organisms and the decomposition of organic matter. *Effects of Acid Precipitation on Terrestrial Ecosystems* (ed. by T. C. Hutchinson and M. Havas), pp. 341–62. Plenum Press, New York.

ALSTAD, D. N., EDMUNDS, G. F. and WEINSTEIN, L. H. (1982). Effects of air pollutants on insect populations. *Annual Review of Entomology*, **27**, 369–84.

ASHENDEN, T. W. and MANSFIELD, T. A. (1978). Extreme pollution sensitivity of grasses when SO_2 and NO_2 are present in the atmosphere together. *Nature*, **273**, 142–3.

AUSUBEL, J. and NORDHAUS, W. D. (1983). A review of estimates of future carbon dioxide emissions. *Changing Climate*, pp. 153–82, National Academy Press, Washington, D.C.

AWANG, M. B. (1979). The effects of sulphur dioxide pollution on plant growth with special reference to *Trifolium repens*. Ph.D. thesis, University of Sheffield.

AYAZLOO, M. and BELL, J. N. B. (1981). Studies on tolerance to sulphur dioxide of grass populations in polluted areas I. Identification of tolerant populations. *New Phytologist*, **88**, 203–22.

BACHE, B. W. (1980). The acidification of soils. *Effects of Acid Precipitation on Terrestrial Ecosystems* (ed. by T. C. Hutchinson and M. Havas), pp. 183–202. Plenum Press, New York.

BACHE, B. W. and SCOTT, N. M. (1979). Sulphur emissions in relation to sulphur in soils and crops. *International Symposium on Sulphur Emissions in the Environment* (ed. by F. F. Ross), pp. 242–54. Society for Chemical Industry, London.

BACH, W. (1984). *Our Threatened Climate: Ways of Averting the CO_2 Problem Through Rational Energy Use.* Reidel, Dordrecht.

BAKER, C. K., UNSWORTH, M. H. and GREENWOOD, P. (1982). Leaf injury on wheat plants exposed in the field in Winter to SO_2. *Nature*, **299**, 149–51.

BEAMISH, R. J., LOCKHART, W. L., VAN LOON, J. L. and HARVEY, H. H. (1975). Long term acidification of a lake and resulting effects on fishes. *Ambio*, **4**, 49–102.

BELL, J. N. B. (1982). Sulphur dioxide and the growth of grasses. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 225–46. Butterworth, London.

BELL, J. N. B. (1984). Air pollution problems in Western Europe. *Gaseous Air Pollutants and Plant Metabolism* (ed. by M. J. Koziol and F. R. Whatley). Butterworth, London.

BELL, J. N. B., AYAZLOO, M. and WILSON, G. B. (1982). Selection for sulphur dioxide tolerance in grass populations in polluted areas. *Proceedings of the Second European Ecological Symposium, Berlin* (ed. by R. Bornkann, J. A. Lee and M. R. D. Seaward), pp. 171–80. Blackwell, Oxford.

BELL, J. N. B., RUTTER, A. J. and RELTON, J. (1979). Studies on the effects of low levels of sulphur dioxide on the growth of *Lolium perenne* L. *New Phytologist*, **83**, 627–43.

BELL, R. M. (1980). Plant growth and gaseous pollutants in urban areas. Ph.D. thesis, University of Liverpool.

BENNETT, J. P., BARNES, K. and SHINN, J. H. (1980). Interactive effects of H_2S and O_3 on the yield of snap beans (*Phaseolus vulgaris* L.). *Environmental and Experimental Botany*, **20**, 107–14.

BERN, J., NEUFELD, R. D. and SHAPIRO, M. A. (1980). Solid waste management of coal conversion residuals form a commercial-size facility: environmental engineering aspects. Report No. DOE/ET/20023-5. US Department of Energy, Washington, D.C.

BEVAN, R. J. and GREENHALGH, G. H. (1976). *Rhytisma acerinum* as a biological indicator of pollution. *Environmental Pollution*, **10**, 271–85.

BINNS, W. O., REDFERN, D. B., REYNOLDS, K. and BETTS, A. J. A. (1985). *Forest Health and Air Pollution: 1984 Survey*. Forestry Commission Research and Development Paper 142, Forestry Commission, Edinburgh.

BLEASDALE, J. K. A. (1957). Smoke pollution and the growth of plants. *Herbage Abstracts*, **27**, 161–4.

BOSCH, C., PFANNKUCH, E., BAUM, U. and REHFUESS, K. E. (1983). Über die erkrankung der fichte (*Picea abies* Karst.) in den hochlagen des Bayerischen waldes. *Forstwissenschaftliches Centralblatt*, **102**,

BRAUN, G. (1977). Über Ursachen der Immissionsresistenz bei Fichte und Folgerungen fur die Resistenzzuchtüng. *Forstwissenschaftliches Centralblatt*, **96**, 62–7.

BROWMAN, M. G. and MASKARINEC, M. P. (1981). Environmental aspects of organics in selected coal conversion solid wastes. *Environmental Science and Technology*.

CENE (1981). *Coal and the Environment*. Commission of Energy and the Environment. HMSO, London.

CHARLSON, R. J. and RODHE, H. (1982). Factors controlling the acidity of natural rainwater. *Nature*, **295**, 683–5.

CHILGREN, J. D. (1978). Small mammal investigations at ZAPS: demographic studies and responses to gradient levels of SO_2. *Bioenvironmental Impact of a Coal-Fired Power Plant* (ed. by E. M. Preston and T. L. Gullett), pp. 764–89. 4th Interim Report, EPA-600/3/79–044.

CLELAND, J. G. (1981). Environmental hazard ranking of pollutant generated in coal gasification processes. Report No. EPA-600/S7-81-101. Industrial Environmental Research Laboratory, Research Triangle Park.

COGBILL, C. V. (1977). The effect of acid precipitation on tree growth in eastern North America. *Water, Air and Soil Pollution*, **8**, 89–93.

COLVILL, K. E., BELL, R. M., ROBERTS, T. M. and BRADSHAW, A. D. (1983). The use of open-top chambers to study the effects of air pollutants, in particular sulphur dioxide on the growth of *Lolium perenne* L. Part II. The long-term effect of filtering polluted air or adding SO_2 to rural air. *Journal of Environmental Pollution (Series A)*, **31**, 35–55.

COWLING, D. W. and KOZIOL, M. J. (1982). Mineral nutrition and plant response to air pollutants. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 349–75. Butterworth, London.

COYNE, P. I. and BINGHAM, G. E. (1978). Photosynthesis and stomatal light responses in snap beans exposed to hydrogen sulphide and ozone. *Journal of the Air Pollution Control Association*, **28**, 1119–23.

CRITTENDEN, P. D. and READ, D. J. (1978). The effects of air pollution on plant growth, with special reference to sulphur dioxide II. Growth studies with *Lolium perenne* L. *New Phytologist*, **80**, 49–65.

CRITTENDEN, P. D. and READ, D. J. (1979). The effects of air pollution on plant growth with special reference to sulphur dioxide III. Growth studies with *Lolium multiflorum* Lam. and *Dactylis glomerata* L. *New Phytologist*, **83**, 645–51.

DAVIES, T. (1980). Grasses more sensitive to SO_2 pollution in conditions of low irradiance and short days. *Nature*, **284**, 483–5.

DAVISON, A. and BAILEY, I. F. (1982). SO_2 pollution reduces the freezing resistance of ryegrass. *Nature*, **297**, 450–2.

DE KOK, L. J., THOMPSON, C. R. and MUDD, J. B. (1982). Effects of H_2S on water-extractable sulphydryl content of crop plants. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 501–23. Butterworth, London.

DODD, J. L. and LAUENROTH, W. K. (1981). Effects of low-level SO_2 fumigation on decomposition of western wheatgrass litter in a mixed grass prairie. *Water, Air and Soil Pollution*, **15**, 275–61.

DOHMEN, G. P., MCNEILL, S. and BELL, J. N. B. (1984). Increases in *Aphis fabae* pest potential caused by air pollution. *Nature*, **307**, 52–3.

EDMONDS, J. and REILLY, J. (1983a). A long-term global energy-economic model of carbon dioxide release from fossil fuel use. *Energy Economics*, **5**, 74–88.

EDMONDS, J. and REILLY, J. (1983b). Global energy and CO_2 to the year 2050. *The Energy Journal*, **4**, 21–47.

EDMONDS, J. A., REILLY, J., TRABALKA, J. R. and REICHLE, D. E. (1984). *An Analysis of Possible Future Atmospheric Retention of Fossil Fuel Co_2*. Report No. DOE/OR/21400-1, Department of Energy, Washington.

Edwards, D. and Gjedrem, D. (1979). Genetic variation in survival of brown trout eggs, fry and fingerlings in acidic water. Report FR16/79, SNSF, Oslo.

EFN (1984). *Förbränningsteknik.* Report No. 7, Energiforskningsnämden, Stockholm.

Elkiey, T. and Ormrod, D. P. (1981). Sulphur and nitrogen nutrition and misting effects on the response of bluegrass to ozone, sulphur dioxide, nitrogen dioxide or their mixtures. *Water, Soil and Air Pollution,* **16,** 177–86.

Emep (1981). *The Cooperative Programme for Monitoring and Evaluation of Long-range Transmission of Air Pollutants in Europe.* ECE, United Nations.

Evans, L. S., Conway, C. A. and Lewin, K. F. (1980). Yield responses of field-grown soybeans exposed to simulated acid rain. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 162–3. SNSF, Oslo.

Evans, L. S., Hendrey, G. R., Stensland, G. J., Johnson, D. W. and Francis, A. J. (1981). Acidic precipitation: considerations for an air quality standard. *Water, Air and Soil Pollution,* **16,** 469–509.

Evans, L. S., Lewin, K. F., Cunningham, E. A. and Patti, M. J. (1982). Effects of simulated acid rain on yields of field-grown crops. *New Phytologist,* **91,** 429–41.

Evans, L. S., Lewin, K. F., Patti, M. J. and Cunningham, E. A. (1983). Productivity of field-grown soybeans exposed to simulated acidic rain. *New Phytologist,* **93,** 377–88.

Farrar, J. F., Relton, J. and Rutter, A. J. (1977). Sulphur dioxide and scarcity of *Pinus sylvestris* in the industrial Pennines. *Environmental Pollution,* **14,** 63–8.

Ferguson, P. and Lee, J. A. (1979). The effects of bisulphite and sulphate upon photosynthesis in *Sphagnum. New Phytologist,* **82,** 703–12.

Ferguson, P. and Lee, J. A. (1980). Some effects of bisulphite and sulphate on the growth of *Sphagnum* species in the field. *Environmental pollution (Series A),* **21,** 59–71.

Ferguson, P., Lee, J. A. and Bell, J. N. B. (1978). The effects of sulphur pollutants on the growth of *Sphagnum* species. *Environmental Pollution,* **16,** 151–61.

Fowler, D. and Cape, J. N. (1982). Air pollutants in agriculture and horticulture. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 1–26. Butterworth, London.

Francis, C. W., Boegly, W. J. Jr., Turner, R. R. and Davis, E. C. (1981). Coal conversion solid waste disposal. Presented at the American Society of Civil Engineers Conference, Energy in the Man-Built Environment, August 3–15, 1981, Vail, CO.

Galloway, J. N., Schofield, C. L., Hendrey, G. R., Altwicker, E. R. and Troutman, D. E. (1980). An analysis of lake acidification using annual budgets. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 254–5. SNSF, Oslo.

Garsed, S. G., Farrar, J. F. and Rutter, A. J. (1979). The effects of low concentrations of sulphur dioxide on broad-leaved British tree species. *Journal of Applied Ecology,* **16,** 217–26.

Garsed, S. G., Mueller, P. W. and Rutter, A. J. (1982). An experimental design for studying the effects of fluctuating concentrations of SO_2 on plants. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 455–7. Butterworth, London.

Garsed, S. G. and Rutter, A. J. (1982a). The relative sensitivities of conifer populations to SO_2 in screening tests with different concentrations of sulphur dioxide. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 474–5. Butterworth, London.

Garsed, S. G. and Rutter, A. J. (1982b). Relative performance of conifer populations in various tests for sensitivity to SO_2, and the implications for selecting trees for planting in polluted areas. *New Phytologist,* **92,** 349–67.

Gas Processing Handbook (1979). *Hydrocarbon Processing, April 1979,* pp. 99–107.

Gehrmann, J. and Ulrich, B. (1982). Der Einfluss des Sauren Niederschlages auf die Naturverjüngung der Buche. *Immissions belastungen von Waldökosystemen,* pp. 32–42. Landesanstalt für Ökologie Landschaftsentwicklung und Forstplanung Nordrhein-Westfalen, Recklinghausen.

Gehrs, C. W., Shriner, D. S., Herbes, S. E., Salmon, E. J. and Perry, H. (1981). Environmental, health and safety implications of increased coal utilization. *Chemistry of Coal Utilization,* Second Supplementary Volume (ed. by M. A. Elliot). Wiley, New York.

Graham, M. S. and Wood, C. M. (1981). Toxicity of environmental acid to the rainbow trout: indications of water hardness, acid type and exercise. *Canadian Journal of Zoology,* **59,** 1518–26.

GRIFFIN, R. A., SCHULLER, R. M., SULOWAY, J. J., SHIMP, N. F. CHILDERS, W. F. and SHILEY, R. H. (1980). Chemical and biological characterization of leachates from coal solid wastes. *Environmental Geology Notes*, Illinois State Geological Survey, 89–99.

GRZYWACZ, A. and WAZNY, J. (1973). The impact of industrial air pollutants on several important pathogenic fungi. *European Journal of Forest Pathology*, **3**, 129–41.

GUDERIAN, R. (1966). Reaktionen vol Pflanzengemeinschaften des Feldfutterbaues auf Schwefeldioxideinwirkingen. Schriftenr. d. Landesanstalt für Immissions und Bodennutzungsschutz des Landes NW, No. 4, pp. 80–100. Verlag W. Girardet, Essen.

HARRIMAN, R. and MORRISON, B. (1980). Ecology of acid streams draining forested and non-forested catchments in Scotland. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 312–3. SNSF, Oslo.

HECK, W. W., ADAMS, R. M., CURE, W. W., HEAGLE, A. S., HEGGESTAD, H. E., KOHUT, R. J., KRESS, L. W., RAWLINGS, J. O. and TAYLOR, O. C. (1983). A reassessment of crop loss from ozone. *Environmental Science and Technology*, **17**, 573–81.

HENDREY, G. R. and VERTUCCI, F. A. (1980). Benthic plant communities in acidic Lake Colden, New York: *Sphagnum* and the algal mat. *Ecological Impact of Acidic Precipitation* (ed., by D. Drabløs and A. Tollan), pp. 314–5. SNSF, Oslo.

HILL, A. C., HILL, S., LAMB, C. and BARRETT, T. W. (1974). Sensitivity of native desert vegetation to SO_2 and SO_2 and NO_2 combined. *Journal of the Air Pollution Control Association*, **24**, 153–7.

HORNTVEDT, R., DOLLARD, G. J. and JORANGER, E. (1980). Effects of acid precipitation on soil and forest, 2. Atmosphere-vegetation interactions. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 192–3. SNSF, Oslo.

HORSMAN, D. C., ROBERTS, T. M. and BRADSHAW, A. D. (1979). Studies on the effect of sulphur dioxide on perennial ryegrass (*Lolium perenne* L.), II. Evolution of sulphur dioxide tolerance. *Journal of Experimental Botany*, **30**, 495–501.

HORSMAN, D. C., ROBERTS, T. M., LAMBERT, M. and BRADSHAW, A. D. (1979). Studies on the effect of sulphur dioxide on perennial ryegrass (*Lolium perenne* L.) I. Characteristics of fumigation system and preliminary experiments. *Journal of Experimental Botany*, **30**, 485–93.

HOUSTON, D. B. (1974). Response of selected *Pinus strobus* L. clones to fumigations with sulphur dioxide and ozone. *Canadian Journal of Forest Research*, **4**, 65–8.

HOVLAND, J. and ISHAC, Y. Z. (1975). Effects of simulated acid precipitation and liming on nitrification in forest soil. Report FR14/75. SNSF, Oslo.

HOWELLS, G. D., BROWN, D. J. A. and SADLER, K. (1983). Effects of acidity, calcium and aluminium on fish survival and productivity—a review. *Journal of the Science of Food and Agriculture*, **34**, 559–70.

HUGHES, P. R., POTTER, J. E. and WEINSTEIN, L. H. (1982). Effects of air pollution on plant-insect interactions: increased susceptibility of greenhouse-grown soybeans to the Mexican bean beetle after plant exposure to SO_2. *Environmental Entomology*, **11**, 173–6.

HULTBERG, H. (1983). Effects of acid depositions on aquatic ecosystems. *Symposium-Acid Deposition: a Challenge for Europe* (ed. by H. Ott and H. Stangl), pp. 167–85. Commission of the European Communities, Brussels.

HUTTENEN, S., HAVAS, P. and LAINE, K. (1981a). Effects of air pollutants on the wintertime water economy of the Scots pine (*Pinus sylvestris*). *Holarctic Ecology*, **4**, 94–101.

HUTTUNEN, S., KARENLAMPI, L. and KOLARI, K. (1981b). Changes in osmotic potential and some related physiological variables in needles of polluted Norway spruce (*Picea abies*). *Ann. Bot. Fennici*, **18**, 63–71.

IEA (1982). *World Energy Outlook*. OECD, Paris.

IERE (1981). *Effects of SO_2 and its Derivatives on Health and Ecology, Volume 2: Natural Ecosystems, Agriculture and Fisheries*. Report of a Working Group, International Electric Research Exchange.

IRVING, P. M. (1983). Acidic precipitation effects on crops: a review and analysis of research. *Journal of Environmental Quality*, **12**, 442–453.

IRVING, P. M. and MILLER, J. E. (1981). Productivity of field-grown soybeans exposed to acid rain and sulphur dioxide alone and in combination. *Journal of Environmental Quality*, **10**, 473–8.

JÄGER, J. (1983). *Climate and Energy Systems: A Review of Their Interactions*. Wiley, Chichester.

JOHANNES, A. H. and ALTWICKER, E. R. (1980). Atmospheric inputs to three Adirondack lake watersheds. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 256–7. SNSF, Oslo.

JOHANSSON, N., KIHLSTRÖM, J. E. and WAHLBERG, A. (1973). Low pH values shown to affect developing fish eggs. *Ambio*, **2**, 42–3.

JOHNSON, A. H. and SICCAMA, T. G. (1983). Acid deposition and forest decline. *Environmental Science and Technology*, **17**, 294–305.

JOHNSON, A. H., SICCAMA, T. G., WANG, D., GURNER, R. S. and BARRINGER, T. H. (1981). Recent changes in patterns of tree growth rate in the New Jersey Pinelands: a possible effect of acid rain. *Journal of Environmental Quality*, **10**, 427–30.

JONES, T. and MANSFIELD, T. M. (1982). The effect of SO_2 on growth and development of seedlings of *Phleum pratense* under different light and temperature environments. *Environmental Pollution (Series A)*, **27**, 57–71.

JONES, T. and MANSFIELD, T. M. (1983). Studies on dry matter partitioning and distribution of ^{14}C-labelled assimilates in plants of *Phleum pratense* exposed to SO_2 pollution. *Environmental Pollution (Series A)*, **28**, 199–207.

JONSSON, B. and SUNDBERG, R. (1972). Has acidification by atmospheric pollution caused a growth reduction in Swedish forests? Research Notes No. 20, Royal College of Forestry, Stockholm.

KELLER, T. (1977). Begriff und Bedeutung der "latenden Immissionsschädigung". *AllgemeineForst-und Jagdzeitung*, **148**, 115–20.

KELLER, T. (1978a). Wintertime atmospheric pollutants—do they affect the performance of deciduous trees in the ensuing growing season? *Environmental Pollution*, **16**, 243–7.

KELLER, T. (1978b). Frostschäden als Folge einer "latenten" Immissionsschädigung. *Staub, Reinhaltung der Luft*, **38**, 24–6.

KELLER, T. (1981). Folgen einer winterlichen SO_2—Belastung für die Fichte. *Gartenbauwissenschaft*, **46**, 170–8.

KLEIN, B. (1980). Zusammenhänge zwischen Immissions und Trockenresistenz bei Fichte, *Picea abies* (L.) Karst. *European Journal of Forest Pathology*, **10**, 186–90.

KLUCZWESKI, S. M., BELL, J. N. B., BROWN, K. A. and MINSKI, M. J. (1983). The uptake of (^{35}S)-carbonyl sulphide by plants and soils. *Ecological Aspects of Radionuclide Releases* (ed. by P. Coughtrey). Blackwell, Oxford.

KNABE, W. (1970). Kiefernwaldverbreitung und Schwefeldioxid-Immissionen im Ruhrgebiet. *Staub, Reinhaltung der Luft*, **39**, 32–5.

KNABE, W. (1976). Effects of sulphur dioxide on terrestrial vegetation. *Ambio*, **5**, 213–8.

KOTCON, J. B. (1976). Assessment of effects of gas effluents. *Coal Technology Programme Quarterly Progress Report for Period ending December 31, 1976*. ORNL Report No. 5252.

KOZLOWSKI, T. T. (1980). Impacts of air pollution on forest ecosystems. *Bioscience, February 1980*, 88–93.

LEE, J. J. and WEBER, D. E. (1982). The effect of sulphuric acid rain on major cation and sulphate concentrations of water percolating through two model hardwood forests. *Journal of Environmental Quality*, **11**, 57–64.

LEETHAM, J. W., MCNARY, T. J. and DODD, J. L. (1978). Effects of controlled levels of SO_2 on invertebrate consumers. *Bioenvironmental Impact of a Coal-fired Power Plant* (ed. by E. M. Preston and T. L. Gullett), pp. 723–89. EPA-600/3-79-044, US Environmental Protection Agency, Corvallis.

LEGGE, A. H. (1980). Primary productivity, sulphur dioxide and the forest ecosystem: an overview of a case study. *Effects of Air Pollutants on Mediterranean and Temperate Forest Ecosystems*, pp. 51–62. USDA, Pacific Southwest Forest and Range Experiment Station, Berkeley.

LEIVESTAD, H., MUNIZ, I. P. and ROSSELAND, B. O. (1980). Acid stress in trout from a dilute mountain stream. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 318–9. SNSF, Oslo.

LINES, R. (1979). Airborne pollutant damage to vegetation—observed damage. *International Symposium on Sulphur Emissions and the Environment* (ed. by F. F. Ross), pp. 234–41. Society for Chemical Industry, London.

LOUCKS, O. L. and ARMENTANO, T. V. (1982). Estimating crop yield effects from ambient air pollutants in the Ohio River valley, *Journal of the Air Pollution Control Association*, **32**, 146–64.

McClenahen, J. R. (1978). Community changes in a deciduous forest exposed to air pollution. *Canadian Journal of Forest Research*, **8**, 432–8.

McNary, T. J., Milchunas, D. G., Leetham, J. W., Lauenroth, W. K. and Dodd, J. L. (1981). Effect of controlled levels of SO_2 on grasshopper densities on a northern mixed-grass prairie. *Journal of Economic Entomology*, **74**, 91–3.

Mansfield, T. M. and Freer-Smith, P. H. (1981). Effects of urban air pollution on plant growth. *Biological Reviews*, **56**, 343–68.

Marland, G. (1982). The impact of synthetic fuels on global carbon dioxide emissions. *Carbon Dioxide Review: 1982* (ed. by W. C. Clark), pp. 406–10. Clarendon Press, Oxford.

Marland, G. and Rothy, R. M. (1984). Carbon dioxide emissions from fossil fuels: a procedure for estimation and results for 1950–82. *Tellus*, **36B**, 232–61.

Materna, J. (1973). Relationship between SO_2 concentration and damage of forest trees in the Slavkov forest region. *Prace Vulhm*, **43**, 169–77.

Materna, J., Jurgle, J. and Kucera, J. (1969). Vysledky mereni koncentraci kyslicniku sirichitého v lesich Krusnych hor. *Ochrana ovzdusi*, **6**, 84–92.

Mintzer, I. and Miller, A. (1984). *Impacts of Energy Conservation Measures and Renewable Energy Technologies on Global Emissions of Carbon Dioxide*. World Resources Institute, Washington, D.C.

Murphy, M. D. (1978). Responses to sulphur in Irish grassland. *Sulphur in Forages* (ed. by J. C. Brogan), pp. 95–109. An Foras Taluntais, Dublin.

Murphy, M. D. (1979). Much Irish grassland is deficient in sulphur. *Farm and Food Research*, **10**, 190–2.

Nihlgård, B. (1985). The ammonium hypothesis: an additional explanation to the forest dieback in Europe. *Ambio*, **14**, 2–8.

Norby, R. J. and Luxmoore, R. J. (1983). Growth analysis of soybean exposed to simulated acid rain and gaseous air pollutants. *New Phytologist*, **95**, 277–87.

Nordhaus, W. D. and Yohe, G. (1983). Future paths of energy and carbon dioxide emissions. *Changing Climate*, pp. 87–152, National Academy Press, Washington, D.C.

Overrein, L. N., Seip, H. M. and Tollan, A. (1980). *Acid Precipitation—Effects on Forest Fish*. Final Report of the SNSF Project 1972–1980. SNSF, Oslo.

Pearce, F. (1982). The menace of acid rain. *New Scientist, 12 August*, 419–24.

Perry, A. M. (1982). Carbon dioxide production scenarios. *Carbon Dioxide Review: 1982* (ed. by W. C. Clark), pp. 337–63. Clarendon Press, Oxford.

Prinz, B., Krause, G. H. M. and Stratmann, H. (1982). *Waldschaden in der Bundesrepublik Deutschland*. LIS, Berichter No. 28.

Reinert, R. A., Heagle, A. S. and Heck, W. W. (1975). Plant responses to pollutant combinations. *Responses of Plants to Air Pollution* (ed. by J. B. Mudd and T. T. Kozlowski), pp. 159–77. Academic Press, New York.

Reister, D. B. (1984). *An Assessment of the Contribution to Gas to the Global Emissions of Carbon Dioxide*. Final Report GR1-84/003, Gas Research Institute, Chicago.

Renberg, I. and Hellberg, T. (1982). The pH history of lakes in southwestern Sweden, as calculated from the subfossil diatom flora of the sediments. *Ambio*, **11**, 30–3.

Rice, P. M., Pye, L. H., Boldi, R., O'Loughlin, J., Tourangeau, P. C. and Gordon, C. C. (1978). The effects of "low level SO_2" exposure on sulphur accumulation and various life responses of some major grassland species on the ZAPS sites. *Bioenvironmental Impact of a Coal-fired Power Plant* (ed. by E. M. Preston and T. L. Gullett), pp. 494–591. EPA-600/3-79-044, U.S. Environmental Protection Agency, Corvallis.

Richter, D. D., Johnson, D. W. and Todd, D. E. (1983). Atmospheric sulfur deposition neutralization, and ion leaching in two deciduous forest ecosystems. *Journal of Environmental Quality*, **12**, 263–71.

Roberts, T. M., Clarke, T. A., Ineson, P. and Gray, T. R. G. (1980). Effects of sulphur deposition on litter decomposition and nutrient leaching in coniferous forest soils. *Effects of Acid Precipitation on Terrestrial Ecosystems* (ed. by T. C. Hutchinson and M. Havas), pp. 381–93. Plenum Press, New York.

Roose, M. L., Bradshaw, A. D. and Roberts, T. M. (1982). Evolution of resistance to gaseous air pollutants. *Effects of Gaseous Air Pollution in Agriculture and Horticulture* (ed. by M. H. Unsworth and D. P. Ormrod), pp. 379–409. Butterworth, London.

Rosenberg, C. R., Hutnik, R. J. and Davis, D. D. (1979). Forest composition at varying distances from a coal-burning power plant. *Environmental Pollution*, **19**, 307–17.

Rosenqvist, I. T. (1978). Acid precipitation and other possible sources for acidification of rivers and lakes. *Science of the Total Environment*, **10**, 39–49.

Rosseland, B. O., Sevaldrud, I., Svalastog, D. and Muniz, I. P. (1980). Studies on freshwater fish populations—effects of acidification on reproduction, population structure, growth and food selection. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 336–7.

Rost, K. (1982). Toxicität von Aluminium bei Keimpflanzen. Dissertation, University of Göttingen.

Rothy, R. M. and Masters, C. (1984). *Past and Future Releases from Fossil Fuel Combustion*. Institute for Energy Analysis, Oak Ridge, Tennessee.

Schlesinger, M. E. (1983). A review of climate models and their simulation of CO_2-induced warming. *International Journal of Environmental Studies*, **20**, 103–14.

Scholz, F. (1981). Geneological aspects of air pollution effects on northern forests. *Silva Fennica*, **15**, 384–91.

Seidel, S. and Keyes, D. (1983). *Can We Delay Greenhouse Warming*? Environmental Protection Agency, Washington, D.C.

Shriner, D. S. (1977). Effects of simulated rain acidified with sulphuric acid on host-parasite interactions. *Water, Air and Soil Pollution*, **8**, 9–14.

Shriner, D. S. (1978). Effects of simulated acidic rain on host–parasite interactions in plant diseases. *Phytopathology*, **68**, 213–8.

Shriner, D. S., McLaughlin, S. B. and Taylor, G. E. (1982). Ecological implications of air pollutants from synthetic fuels processing. *Beyond the Energy Crisis, Vol. 2* (ed. by R. A. Fazzolare and C. B. Smith), pp. 669–76. Pergamon Press, Oxford.

Siegenthaler, U. (1983). Uptake of excess CO_2 by an outcrop-diffusion model of the ocean. *Journal of Geophysical Research*, **88**, 35987–608.

Smagorinsky, J. (1982). *Carbon Dioxide and Climate—A Second Assessment*. National Academy Press, Washington, D.C.

Smith, W. H. (1974). Air pollution—effects on the structure and function of the temperate forest ecosystem. *Environmental Pollution*, **6**, 111–29.

Soikkeli, S. and Tuovinen, T. (1979). Damage in mesophyll ultrastructure of needles of Norway spruce in two industrial environments in central Finland. *Ann. Bot. Fennici*, **16**, 50–64.

Spaleny, J. (1979). Influence of atmospheric SO_2 on the frost-hardiness of Norway spruce. *International Symposium on Sulphur Emissions and the Environment* (ed. by F. F. Ross), pp. 272–3. Society for Chemical Industry, London.

Tamm, C. O. (1976). Acid precipitation: biological effects in soil and on forest vegetation. *Ambio*, **5**, 235–8.

Tamm, C. O. and Wiklander, G. (1980). Effects of artificial acidification with sulphuric acid on tree growth in Scots pine forest. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 188–9. SNSF, Oslo.

Tamura, T. and Boegly, W. J. Jr. (1980). Leaching studies of coal gasification solid wastes to meet RCRA requirements for land disposal. *Proceedings of Second DOE Environmental Control Symposium*. CONF 800334/1, Washington D.C., 397–414.

Taylor, G. E., McLaughlin, S. B., Shriner, D. S. and Selvidge, W. J. (1983). The flux of sulphur-containing gases to vegetation. *Atmospheric Environment*, **16**,

Taylor Jr. G. E. and Selvidge, W. J. (1984). The phytotoxicity in *Phaseolus vulgaris* of five sulfur-containing gases released from advanced fossil energy technologies. *Journal of Environmental Quality*, **13**, 224–30.

Taylor, J. E. and Leininger, W. C. (1978). Plant community changes due to low level SO_2 exposure. *Bioenvironmental Impact of a Coal-fired Power Plant* (ed. by E. M. Preston and T. L. Gullett), pp. 610–31. EPA-600/3-79-044, US Environmental Protection Agency, Corvallis.

Thompson, C. R. and Kats, G. (1978). Effects of continuous H_2S fumigation on crop and forest plants. *Environmental Science and Technology*, **12**, 550–4.

Thompson, C. R., Kats, G. and Lennox, R. W. (1979). Effects of fumigating crops with hydrogen sulfide or sulfur dioxide. *California Agriculture*, **32**, 9–10.

Thompson, C. R., Kats, G. and Lennox, R. W. (1980). Effects of SO_2 and/or NO_2 on native plants of the Mojave Desert and Eastern Mojave—Colorado Desert. *Journal of the Air Pollution Control Association*, **30**, 1304–9.

Thornton, N. C. and Setterstrom, C. (1940). Toxicity of ammonia chlorine, hydrogen cyanide, hydrogen sulphide and sulphur dioxide gases III. Green plants. *Contributions from Boyce Thompson Institution*, **11**, 3143–56.

TOMLINSON, G. H. II (1983). Air pollutants and forest decline. *Environmental Science and Technology*, **17**, 246–56.

TRESHOW, M. (1968). Impact of air pollutants on plant populations. *Phytopathology*, **58**, 1108–13.

TROEDSSON, T. (1980). Ten years' acidification of Swedish forest soils. *Ecological Impact of Acid Precipitation* (ed. by D. Drabløs and A. Tollan). SNSF, Oslo.

TROIANO, J., COLAVITO, L., HELLER, L., MCCUNE, D. C. and JACOBSON, J. S. (1983). Effects of acidity of simulated rain and its joint action with ambient ozone on measures of biomass and yield in soybean. *Environmental and Experimental Botany*, **23**, 113–9.

ULRICH, B. (1982). An ecosystem orientated hypothesis on the effect of air pollution on forest ecosystems. Paper for the 1982 Stockholm Conference on *Acidification of the Environment*.

ULRICH, B., MAYER, R. and KHANNA, P. K. (1980). Chemical changes due to acid precipitation in a loess-derived soil in central Europe. *Soil Science*, **130**, 193–9.

WENTZEL, K. F. (1983). IUFRO-studies on maximal SO_2 emissions standards to protect forests. *Effects of Accumulation of Air Pollutants in Forest Ecosystems* (ed. by B. Ulrich and J. Pankrath), pp. 295–302. Reidel, Dordrecht.

WEST, D. C., MCLAUGHLIN, S. B. and SHUGART, H. H. (1980). Simulated forest response to chronic air pollution stress. *Journal of Environmental Quality*, **9**, 43–9.

WHITMORE, M. E. and FREER-SMITH, (1982). Growth effects of SO_2 and/or NO_2 on woody plants and grasses observed during the Spring and Summer. *Nature*, **300**, 55–7.

WINNER, W. E. and BEWLEY, J. D. (1978). Contrasts between bryophyte and vascular plant synecological responses in an SO_2-stressed white spruce association in Central Alberta. *Oecologia (Berlin)*, **33**, 311–25.

WRIGHT, R. F. and HENRICKSEN, A. (1979). Sulphur: acidification of freshwaters. *International Symposium on Sulphur Emissions and the Emissions and the Environment* (ed. by F. F. Ross). Society for Chemical Industry, London.

ØKLAND, J. and ØKLAND, K. A. (1980). pH level and food organisms for fish: studies of 1,000 lakes in Norway. *Ecological Impact of Acidic Precipitation* (ed. by D. Drabløs and A. Tollan), pp. 326–7. SNSF, Oslo.

Index

Acid mine drainage 41
Acid precipitation
  and agriculture 303
  and freshwater ecosystems 307-11
  and grassland 297–302
  and invertebrates 299
  and nutrient cycling 295–7, 306
  and soil 289, 295–6, 306
  and tree growth 294–7
  and water chemistry 295, 307–8
  dry deposition 283, 285–8
  nutritional effects 306–7
  wet deposition 283, 287–8
Acidity
  in colliery spoil 41, 44
Africa
  coal reserves 9
  coal resources 5, 8
  oil production 2
Agriculture
  and coal transport 73
  effect of SO_2 and NO_x on 284, 303
  effect of ozone on 303–4
  on reclaimed land 39–43
Alcohol
  in CLM combustion 85
Aluminium (Al)
  in coal waste 41, 44, 305
  in fly ash 179, 181, 184
  in freshwater 309
  in gasification processes 146, 187
  in liquefaction processes 128
  in soil 192, 296–7
  production 16
Ames test 256
Ammonia (NH_3) 250, 284
  and vegetation 304–6
  deposition 288
  in FGD 103
  in gasification processes 263–4, 267–8, 270, 274
  in liquefaction processes 139
  removal of 100
  synthesis 105, 108, 115, 126, 130, 132, 135–8
Antimony (Sb) 200
  in coal 173–8
  in fly ash 179, 181–2, 184, 200
  in gasification processes 187–90, 211, 271–4
  in liquefaction processes 211
  in urban aerosols 195, 197
  in vegetation 204
Aquatic ecosystems
  acidification of 41, 284, 307–10
  BaP in 233–5
  fish 308–9
  fly ash leaching 205–6, 209
  insects 309–10
  trace elements in 193, 305–6
  vegetation 309
Arsenic (As) 172, 206, 221
  global emissions 197–8
  in coal 173–8, 186
  in fly ash 179, 181–5, 194–5, 198, 200–1, 203
  in gasification processes 187–90, 211, 271–3
  in liquefaction processes 211
  in soil 205
  in urban aerosols 195–7, 208
  in vegetation 203–4
Ash ponds 202–3, 207
  trace element mobilization 205–6, 209
Asia
  coal resources 5
Australia 19, 176, 182
  coal production in 20, 27
  coal reserves in 7, 9
  coal resources in 6, 176
  coal trade 24, 26
  steam coal in 13
  AVCO ARC coal process 148

Barium (Ba)
  in fly ash 179
  in gasification processes 187, 271–4
Belgium 246
  coal trade 25
Benzo (*a*) pyrene (*see also* Polycyclic aromatic hydrocarbons)
  as PAH indicator 225
  atmospheric 227–9, 249
  carcinogenicity 222, 225–6
  emission of 160, 219, 225

  in freshwater 233–4
  in gasification processes 260–1, 268
  in jellyfish 235
  in Man 239–40
  in plants 231–2, 235
  in seas 234–5
  in soil 225, 230–2, 236–7
  in USA 219
  in USSR 224
Bergius-Pier process 117
Beryllium (Be) 172
  in coal 173–8
  in gasification processes 187, 271–4
  in vegetation 306
Birds 310
Boron (B) 91, 172, 200, 206, 305
  in coal 173–8
  in gasification processes 187–90, 271–4
  in soil 194
  in vegetation 203–4, 306
Botswana
  coal production in 22
  coal reserves in 7
  coal resources in 6
Bromine (Br)
  in fly ash 179
  in gasification processes 187, 270
  in urban aerosols 195
Brown coal 8, 34, 36, 41
  characteristics 125
  hydrogenation of 128
  world reserves of 9
BTX aromatics 108, 115, 148

Cadmium (Cd) 195
  global emissions 197–8
  in coal 173–8
  in fly ash 179, 181, 192, 194, 200
  in gasification processes 187–90, 271–4
  in liquefaction processes 191
  in soil 192, 202–3, 205
  in vegetation 306
Caesium (Cs)
  in fly ash 179, 200
Calcium (Ca)
  in coal 92
  in colliery spoil 44
  in fly ash 179, 184
  in freshwater 305, 309
  in gasification processes 187
  in soil 295–6
Canada 4
  acid deposition in 307
  coal and oil prices in 13
  coal production in 22
  coal trade 24
  coal reserves in 7, 9
  coal resources in 6
  energy consumption 10
Carbon (C) 112, 115, 139, 184, 272, 283
Carbon dioxide (CO_2) 132, 141, 143, 147
  emissions 260, 283–4
  future concentration 283
  greenhouse effect 245, 283–4
  impact on climate 283–4
  in gasification processes 263–4, 267, 269, 274, 277
  in liquefaction processes 139
Carbon disulphide (CS_2) 277, 284, 304
Carbon monoxide (CO) 91, 115
  emissions 76, 160–1, 246
  in gasification processes 132, 147, 189, 263–4, 267, 274, 277
  in liquefaction processes 139
Carbonyl sulphide (COS) 284, 304–5
Carcinogens
  coal tar 220–2, 225, 227
  dose-response relationship 158
  environmental sources 160, 227–9
  from gasification processes 190, 251, 278
  from liquefaction processes 248
  in fly ash 200–1
  polycyclic aromatic hydrocarbons 160, 218–40
Catalysts 109, 210
  disposal of 192, 207
  in FGD 101
  in gasification processes 131–2, 142–6, 186, 189–92
  in H-coal processes 123, 126
  in liquefaction processes 115, 119, 121, 191
Cement 12, 16
Centrally Planned Economies 8, 11, 26
  energy consumption 10
  coal production in 22
  coal trade 26–7
Cerium (Ce)
  in fly ash 179
Chalcophiles
  in fly ash 180, 185
China 19
  coal gasification in 138
  coal production in 22
  coal reserves in 7, 9
  coal resources in 5–6
  coal trade 24–5
  land reclamation 36
Chromium (Cr) 172, 206–7
  in coal 173–8
  in fly ash 179, 181–3, 192, 201, 208
  in gasification processes 187–90, 271–4
  in liquefaction processes 18, 191
  in soil 192
Clay minerals 44

Climate
  greenhouse effect 283–4
CMEA countries 2, 138
Coal
  as a chemical feedstock 17, 105
  chemical composition 88, 97–8
  chemical structure of 112–17, 258, 305
  conversion technologies 3, 17, 105–49
  cost of electricity generation 15
  displacement of other fuels 12, 14, 25, 284
  environmental effects 14, 29
  extraction of 3–4
  for heating 14
  in cement production 16
  price 12–14, 19, 25
  prospects 2–3, 25–7
  strategy 3–4
  thermal treatment of 124–5
  trace elements in 89, 171–211
  world trade 2, 23–7
Coal ash 42
  emission control 103
  in gasification processes 142
  in USSR coal 19
  removal of 82, 98
  sulphur in 91–5
  trace elements in 206
Coal combustion 25, 81–104
  CO_2 emissions 160, 283–4
  domestic 14, 184–5, 208
  effect on biosphere 289–311
  effluents from 89–97, 160–1, 282
  health risks of 246–8
  methods of 82–9
  modification of 99–100, 228, 246
  PAH pollution 89, 99, 225, 237–40
  radiological impact 160, 165, 198–9, 201–2, 208
  solid wastes from 160, 202–7, 209, 305–6
  trace element emissions 178–81, 189, 192–207, 210
Coal consumption
  annual global 198
  for electricity production 12
  for the iron and steel industry 12, 16
  predictions of 2, 15–16, 25–6
Coal conversion processes 3, 16–18, 26, 185–92
  air pollution from 304–5
  economic viability of 16–17, 107
  emissions from 149, 238–9, 283
  flow diagrams of 109–10, 141, 262, 265, 269
  gasification 129–47, 250–6, 260–79
  health effects 167, 248–51, 269, 278
  hydrogenation 114–29, 239, 259
  liquefaction 17, 112–29
  plant size 18
  products 106, 108, 276
  prospects 17–18
  technologies 17, 50, 105–49, 209–11, 220
  trace elements in 185–92, 305
  waste 206–7, 305–6
Coal demand 2, 11–18
  effect of industrial change 2–3
  in OECD countries 12
  predictions 2–3, 26, 50–1
Coal extraction
  economics of 3
  impacts of 47–8
Coal-fired power plants
  atmospheric pollution from 160–1, 192–202, 237–8, 283, 289
  development of 25
  effluents from 160–3, 208
  health impacts 75, 160, 166
  in Australia 14
  in Poland 14
  in USSR 19
  solid waste from 201–2
  trace element enrichment 179–85, 189, 192–203, 208
  trace element fallout 192–5
  trace element mobilization 178–9, 186
  trace element partitioning 179–80, 188, 209–10
Coal fuel cycle
  estimated effects of 80
  health risks 245
Coal gas 17, 131
  fuel cell 18
Coal geography
  in USA 54–5
  in USSR 52–4
Coal industry
  future 50
  in USSR 54
Coal mining
  environmental characteristics 49
  impacts of 47–8
  mine characteristics 49
  types of 51
Coal oil 17, 109–10, 114–5, 120, 136, 141
  characterization of 117
Coal processing
  cleaning 51, 98–9
  enrichment 51
  water requirement model 60, 66
Coal production 1–3, 12, 27, 178, 201
  and land reclamation 32–3, 37
  and water needs 50–2, 57–71
  annual growth in 2, 56
  coal producing countries 19–22, 26
  costs 21, 32
  developing country potentials 25

  effect of transition to oil 2
  future technologies 50–2
  in USSR 53
  resource demands of 47–71
  trends 19
Coal rail transport 51
  air pollution by 74–5
  cost of 23
  environmental effects of 74–6
  human injury in USA 75, 79
  in USSR 20
Coal reserves
  classification of 3, 5
  in USSR 53
  ratio to production 5, 56–7
  underground mines 49
  world estimates of 7, 9
Coal resource requirements 48–50
  water 50–2, 57–71
Coal resources 3–5, 20
  distribution of 5, 20, 52–5
  ratio to production 8
  world estimates of 5–6, 8
Coal tar 132, 141
  carcinogenicity of 220–2, 225–7
  chemical composition of 221
  PAH in 221–2
Coal trace elements 98, 171–217
  concentrations of 172–7
  enrichment of 179–80, 186–7, 189, 196–7
  in different coal ranks 173–4
  occurrence of 171
  origin of 171–2
  partitioning during combustion 179–80, 188
Coal transport 12, 19, 23, 25, 27, 39
  by conveyor 51
  by rail 51, 74–6
  by truck 51, 74, 76–7, 79
  by water 23, 51, 74, 77, 79
  cost of 4, 23
  environmental impact of 73–80
  health hazards of 74, 77, 79
  in Australia 23
  in Europe 23
  in Japan 23
  in USA 23, 74, 78
  in USSR 19, 74
  road damage 77
Coal waste
  carcinogenic properties of 227
  chemical properties of 41
  physical properties of 41
  storage 39
Coal waste disposal 39
  colliery spoil 39–40
  environmental impacts 47
  from coal cleaning 99
Coal water resources 67–71
  in USA 67–70
  in USSR 69–71
Coalification 112
  stages of 171–2
Cobalt (Co) 207
  in fly ash 181, 192
  in gasification processes 187–90, 271–4
  in liquefaction processes 128, 192
  in soil 192
  in vegetation 204, 306
Coking coal 19–20, 23, 51–2
  coking plants 42
  in USSR 56
  trade 24–5
Combined cycles (CC) 86
Combined power and process methods 86–9
  combination options 88–9
  in gasification 131
  in USSR 131
Copper (Cu) 172, 198
  in coal 173–8
  in fly ash 179
  in gasification processes 187, 271–4
  in liquefaction processes 191
  in soil 192, 205
  in urban aerosols 195
  in vegetation 204
Crop plants
  SO_2 impacts on 303
  O_3 impacts on 303–4
Cyanide (CN) 95, 250
  in gasification processes 268–70, 272, 274
Czechoslovakia 193, 201, 230, 291
  coal production in 22
  gasification processes 277
  land reclamation 34
  strip mining 37

Deposition
  dry 283–7, 294, 305, 307
  velocities 285–8, 304
  wet 283, 287–8, 294, 305, 307
Developing regions 9
  coal cost 26
  coal production in 19, 22
  energy consumption 10
  land reclamation in 40
  predicted production 25
  prospects 27
Desulphurization (*see also* Flue gas desulphurization) 190
  in Synthoil process 191
Diesel fuel 110–1
Diffusion combustion 82
District heating schemes 14

  in FRG 14
  in Sweden 14
  in USA 14
Domsdorf method 42
Donor solvent process 107, 114, 119, 126, 191
  EDS process 121–2
Dust control 58, 100
  in gasification plants 86

East European countries
  coal production in 19, 20
Economic recession 2, 3, 190
  decline in iron and steel production 10
  in Western countries 2, 16
Economy of coal development 3
EDS process 121–2
EEC 176–7
  coal production 21
  coal trade 24–5
Electricity 14–15
  future generation of 14, 52
  generation costs 15
Electricity consumption 14
  in OECD countries 14
  increase in 14
Electricity production from coal 12, 17, 23, 52, 131
  in CPE countries 14, 19, 56
  in EEC 14, 21
  in other countries 14
  in South Africa 14, 20
  in USA 14, 20
  in USSR 53
  risk to public health 160–1, 168
  SO_2 emissions from 161
Emissions (*see also* Liquid and Fugitive emissions)
  aircraft 229
  atmospheric trace elements 192–203
  atmospheric transport 284–8
  automobile 74, 76, 228–9
  benzo (*a*) pyrene 89, 160, 237, 260
  CO_2 283–4
  control technologies 97–103, 246
  exposure commitment model 156–69
  gaseous emissions 91, 149, 160, 252–3, 257, 269, 282–305
  global trace elements 197–9
  health hazards 160–1, 199–200, 207
  long-range transport 246, 285–7
  NO_x 83–5, 99–100, 160, 161, 283–4, 289–94
  PAH 89, 218
  particulate 76, 103, 160–1, 194, 197, 199–200, 246, 260, 283
  radionuclide 162, 163–5, 192, 201–2
  SO_2 85, 100, 160–1, 283–4, 289–84
  solid emissions 90, 202–7, 250, 254–5, 257, 305–7
Energy
  intensive industries 16
  price 4, 9–11
Energy balance 2
Energy conservation 2
Energy consumption 2
  and domestic policy 25
  and economic growth 8–11
  and GDP 8
  and GWP 8–9
  and population growth 9
  and reclamation 45
  change in 8–11
  effect of industrial change 9–11
  effect of price 9–10, 26
  household 10
  in Eastern bloc 14
  of OECD countries 10
  share of coal in 2, 11–12, 284
Energy crisis 1–2
Energy imports 2
  to USA 2
  to Western Europe 2
Energy requirements
  world 2
Energy studies
  AGIP 2
  ICR 14
  IEA 2, 5, 8, 13–14, 17, 21, 24
  IIASA 56
  OTA 56
  USBM/USGS 4
  WEC 4–8
  WOCOL 56
Energy substitution 2
Energy supply 1–2
  cost of 5, 11–14
  from liquid hydrocarbons 2, 12–13
  from nuclear power 12
  from solid fuels 2, 11–14
  fuel substitution 1–2, 11–14
  models of 3
Entrained Bed Gasification 133, 143–4
Epigenic mineralization 172
Europe 250, 293, 297
  acid deposition in 305, 307
  coal trade 25
  economic growth in 25
Exposure commitment model
  applicability of 158–9
  application of 160–9
  concepts 156–7
  definitions 157
  use in environmental pollutant

  assessment 157, 269
EXXON catalytic gasification process 142, 145–6

Federal Republic of Germany 19, 165, 173–4, 230
  coal and oil prices 12–13
  coal gasification in 142–3, 149
  coal liquefaction 110, 121, 149, 191
  coal production 21
  coal reserves 5, 7
  coal resources 5–6, 48–9
  coal trade 24–5
  emissions from power plants 161–2
  energy consumption 11
  land reclamation in 36, 41
  strip mining in 37, 41
Fischer-Tropsch synthesis 108, 111, 132, 138
Fish 193, 206
  benzo (*a*) pyrene 235
  water acidification 307–9
Fixed bed coal combustion 82–3
  sulphur compounds in 91
Flue gas desulphurization 15, 100–3
  Bellman Lord process 102
  methods 101
  water requirement model 60–1, 66
Flue gas nitrogen removal 101–3
Fluidized bed combustion (FBC) 18, 84, 103–4, 237
  fly ash 200–1
  sulphur removal 93–4
  types 103
Fluidized bed gasification 132–3, 142–5
Fluorine (F) 206
  in coal 173–8
  in gasification processes 187–90, 270–4
  in soil 194
  in vegetation 203
Fly ash 91, 162, 178–85, 192, 198, 305
  and plant growth benzo (*a*) pyrene in 225
  chemical composition of 208
  disposal of 202, 209
  fractionation of 178–9, 182–3, 197
  health hazards 199–201
  leaching of 204–6
  reclamation of 43
  removal 89, 127, 160
  trace element enrichment 179–84, 194–5, 199–201, 210
  trace elements in 202–7
Food chain
  PAH in 229–36
Forest ecosystems
  community composition 289–90
  effects of pollutants 284, 289–97
  insects in 293–4
  nutrient cycling 296, 306–7
  stress factors in 292–3
Forestry (*see also* Forest ecosystems)
  and coal transport 73
  on reclaimed land 39–45
France 1, 230
  coal and oil prices in 13
  coal production 21
  coal trade 21, 24–5
  energy consumption 10
Fugitive emissions (*see also* Emissions)
  in coal transport 74–7
  in gasification processes 253
Fumigation expriments 291–304
FUNCAT COGAS Symposium 144
Fungal pathogens 293–4, 296

Gallium (Ga)
  in fly ash 200
  in gasification processes 187
Gasification 17–8, 99–103, 105, 109, 129–47, 282
  basic principles 106, 131–2
  catalysts in 131, 142, 142–6
  commercial processes 132–8
  cost of 17
  development trends 144–7
  emission sources 149
  EXXON process 142, 145–6
  Fischer-Tropsch synthesis 108, 111, 132, 138
  for electricity generation 131
  gas composition 139–40, 260–7, 276–7
  in situ 18, 108
  Koppers-Totzek process 108, 133, 188–90, 207, 260, 262, 277
  Lurgi pressure process 107–8, 132–4, 138–42, 188–90, 256, 260–7, 277
  PAH from 238
  pollution control 252–6
  pyrolysis 132–3, 147–9
  Ruhrchemie AG/Ruhrkohle AG 143
  special processes 138
  Texaco process 143, 260, 270–5
  trace elements in 186–90, 206–7
  Wellman-Galusha process 190, 277
  Winkler process 108, 129, 132–3
Gasoline 108, 110–11, 118, 129
  chemical composition of reformed 130
German Democratic Republic (GDR)
  energy consumption 10
  land reclamation in 33–4, 38, 41
  strip mining in 37–8, 41
Germanium (Gr)
  in gasification processes 187
Germany 1

  coal production in 22
  energy consumption 10
  hydrogenation in 115
  steam coal 13
Grassland ecosystems
  community composition 298–300
  effects of pollutants on 284, 297–302
  nutrient status 307
  SO_2 tolerance 299–301
  stress factors in 301–2
Grate firing *see* Fixed bed coal combustion
Great Britain 1, 19, 173–4, 184, 223, 228, 246, 291, 299–300, 302
  coal and oil price in 13
  coal gasification in 107, 142
  coal liquefaction in 107
  coal production in 21, 22, 48–9
  coal reserves 7, 176
  coal resources 6, 21
  coal trade 24–5
  energy consumption 10–11
  fly ash disposal in 202
  industrial revolution in 1
  land reclamation in 33, 36, 42
  steam coal 13
Greenhouse effect 246
  temperature change 283–4
Gross domestic product (GDP) 8
Gross world product (GWP) 8–11

H-coal process 17, 107, 122–3, 191
  catalyst consumption in 126
Hafnium (Hf)
  in gasification processes 187
Hard coal 5, 8, 174
  world reserves of 9
Harm commitment method 159
Health hazards 199–201
  accidents 75, 78–80
  benzo (*a*) pyrene 225, 239
  coal tar 220–1
  evaluation 257–62
  exposure commitment model 158
  from coal conversion 267, 276, 278
  from coal mining 167
  from coal processing 167, 207–8, 245
  from coal transport 74, 77–9, 167
  from electricity generation 160, 167
  from nuclear fuel cycle 162–5
  from waste management 167
  in coal liquefaction processes 250
  medical monitoring of 251, 257, 278
  PAH 220, 225, 230, 234
  sulphate exposure 247
Heating 52
Heavy metals (*see also* Trace elements) 259
  health hazards 160
  in aquatic ecosystems 193, 305–6
  in coal waste 41, 44
  in soil 205
Helium (He) 144
High Gradient Magnetic Separation 98
Hydrocarbons 2, 91, 293
  emissions 76, 160–1
  in gasification processes 132, 256, 259–60, 263–4, 267, 270, 273–7
  in liquefaction processes 112–6, 120, 128, 131
  in Lurgi oil 276
  in reformed gasoline 130
  role on energy scene 2
Hydrogen (H)
  in gasification 132, 143, 147, 189, 263–4, 267, 272, 274, 276–7
  in liquefaction 112, 115, 120, 122, 126, 139
Hydrogen cyanide (HCN)
  in gasification processes 262–4, 267
Hydrogen sulphide (H_2S) 92, 115, 126, 284
  effect on vegetation 304–6
  in gasification processes 262–4, 267, 269, 277
  in liquefaction processes 140
Hydrogenation 17, 105, 111, 114–29, 186, 239
  health hazards of 248–9, 269, 276–9
  of coal distillate 130–1
  of slurry 119–20, 191
  product chemical composition 112, 114–6, 276
  products of 115–6, 128, 259, 276, 278
  reactor 120
  water requirements 52

IIASA energy study 56
  high growth scenario 57
  low growth scenario 57
India 19, 25, 186
  coal production in 22
  coal reserves in 7
  coal resources in 6
Industrial revolution 1
Iodine (I)
  in fly ash 179, 181
  in gasification processes 187
  in urban aerosols 195
Ireland 307
Iron (Fe) 207, 305
  in colliery spoil 44
  in fly ash 179, 181, 184, 192
  in gasification processes 146, 187, 190
  in liquefaction processes 126
  in urban aerosols 195
Iron and steel industry 10, 12, 147

  coal demand in 16
  crisis of 12
  in OECD 16
  in the UK 16
Iron pyrites (FeS_2) 41, 98–9
  in coal waste 41–2
  in colliery spoil 43
  pyritic oxidation 41
Italy
  coal and oil price 13
  coal trade 24
  energy consumption 10–11
  steam coal 13

Japan
  coal and oil price 13
  coal gasification in 147
  coal liquefaction in 107
  coal production 22
  coal trade 24–6
  energy consumption 10
  FGD in 103
  steel production in 24

Kinetic combustion 82
Koppers-Totzek process 108, 262
  commercial installations 135–7
  entrained bed process 133, 260
  gas composition 277
  trace elements in 188–90, 207
Korea 25
  coal production in 22
  potential coal production 25
Koyne method 42

Land disturbance 29–31, 40, 44
  classification of 30, 41
  land ratios 39
  subsidence 30, 40
Latin America 2
  coal production in 2
  coal reserves in 9
  coal resources in 5, 8
  coal trade 24–5
  oil production in 2
Leaching
  during transit 76
  of BaP 230
  of cations 44, 295–6
  of spoil 44, 205–6, 210
  of trace elements 305
Lead (Pb) 172
  global emissions 197–8
  in coal 173–8
  in fly ash 179, 181–4, 192, 194, 200
  in gasification processes 187–90, 211, 271–3, 275
  in liquefaction processes 191
  in soil 192, 203, 205
  in urban aerosols 195, 197
Lethal Concentration (LC) 256
Lethal Dose (LD) 256
Lignite 18, 115, 138, 142
  hydrogenation of 118, 120–1
  in FRG 49
  in USA 174, 176
  in USSR 19, 53
Limestone
  in coal combustion 84–5, 94, 99
  in FBC 104
Linear no-threshold model (LNTM) 158, 247
Liquefaction 50, 58, 105, 260, 283
  Bergius-Pier process 117
  cost of 17
  definition 106
  direct 110, 112–29
  donor solvent process 107, 114, 119, 191
  emission sources 149
  flow diagram of 119, 124
  H-coal process 17, 107, 122–3, 191
  health hazards 248, 250
  indirect 109, 138–41
  liquid emissions 253–4
  PAH from 238–9
  Saarbergwerke AG process 120
  solvent-refined coal processes 191
  supercritical process 112
  Synthoil process 191–2
  technology 117–29
  trace elements in 190–2, 206–7
  VEBA AG/Ruhrkohle AG process 120, 191
  water requirement model 64–6
  yields 121–3
Liquid emissions 91, 158, 160
  from coal conversion 253–4, 257
Liquid hydrocarbons 2
  in reformed gasoline 130
Liquid mixture combustion 84–5
Lithium (Li)
  in gasification processes 144
Lithophiles 180, 195
Loess 38–9, 42
  in GDR 42
  in USSR 43
Lurgi pressure gasification 107–8
  aqueous steam composition 268
  commercial installations 134
  emissions from 149, 256, 260
  flow diagrams 262, 265
  gas composition 139, 260–7, 276–7
  health hazards 249

  indirect liquefaction process 138–41
  moving bed reactor 132–3, 138–42, 260
  oil composition 276
  particulate composition 261, 266–7
  trace elements in 188–90

Magnesium (Mg) 297
  in colliery spoil 44
  in fly ash 179, 184
  in gasification processes 187
  in soil 295–6
Magnetism
  use in coal cleaning 98
Magnetohydrodynamic (MHD) 18
  technology 86–8
Manganese (Mn) 206
  in colliery spoil 44
  in fly ash 179
  in gasification processes 271–3, 275
  in liquefaction processes 191
  in urban aerosols 195
Mayer method 99
Mercaptans 262, 284
Mercury (Hg) 172, 200
  global emissions 197–8, 209
  in coal 173–8
  in fly ash 179, 184
  in gasification processes 187–90, 211, 267–8, 271–3, 275
  in liquefaction processes 211
  in soil 192–3, 205
  in urban aerosols 195, 197
  in vegetation 204
Metallurgical coal 12, 16
  trade in 23
Methane (CH_4) 107, 126, 132, 141, 145–7
  in gasification processes 263–4, 267, 274, 277
  in liquefaction processes 140
Methanol synthesis 100, 111, 138, 142
Middle East 2
Mineral resources
  classification of 3, 5
  estimates of 5–9
  exhaustion of 3
  extraction of 3–4
Mines (underground) (*see also* Coal mining)
  environmental characteristics 49
  in Australia 20–1
  in other countries 21
  in South Africa 20–1
  in USSR 19, 54
  land disturbance from 29–30, 44, 47
  land reclamation 38
  spoil characteristics 39, 44
  spoil reclamation 39–40, 44
  types of 51
  water requirement model 59–60
Mining 29
  cost 21
  methods 29
  profitability of 4
Mining technology
  in USSR 19
Mining waste reclamation
  back stowage 40
  for agriculture 39
  for forestry 39
  from surface mining 41
  from underground mining 44–5
  in Ruhr 39
Molybdenum (Mo) 206–7
  in fly ash 200
  in gasification processes 187–90
  in liquefaction processes 126–8, 192, 271–3, 275
  in vegetation 204
Moving bed gasification 132–3, 138–42
Mutagens 251

Naphtha 17
Natural gas 3, 14, 219
  displacement by other fuels 16–17
  future usage 50
  in combined cycles 86
  synthetic 18
Nerosine 231
Netherlands
  coal gasification in 143, 146
  PAH in 219
Nickel (Ni) 305
  global emissions 197–8
  in coal 173–8
  in fly ash 179, 182–3, 201
  in gasification processes 186–90, 271–3, 275
  in liquefaction processes 191
  in soil 192, 202, 205
  in urban aerosols 195
  in vegetation 204, 306
Nitrogen (N) 112, 128, 271, 306
  coal combustion 99
  coal emissions 94
  deficiency in colliery spoil 41–5
  in gasification processes 263–4, 267, 272, 274, 277
  in liquefaction processes 139
  soil 42, 295
Nitrogen oxides (NO_x) 94–7, 230, 247
  and acid precipitation 289–311
  chemical transformation of 246
  deposition of 288
  effects on biosphere 97, 245, 289–311
  emission control 97, 99, 100–4

  emissions of 76, 83–5, 89–91, 160–1, 246, 283–4
Nuclear energy 12, 14
  exposure assessment 162, 165
  health effects of 163
  generation costs 15
  in OECD 14
  in UK 21
Nuclear reactors
  and gasification 131, 146

OECD countries 2, 9, 12, 16, 22
  coal demand 12
  coal trade 27
  electricity consumption 14
  energy consumption 10
Oil 1, 5, 12, 14, 16, 219
  appearance on energy market 1–2
  coal derived *see* Coal oil
  displacement by other fuels 2, 16, 284
  electricity generation 15, 17
  price 2, 9, 12–13, 16
Oil production
  in Africa 2
  in developing countries 27
  in Latin America 2
  in the Middle East 2
  in the USA 2
Opencast mines *see* Surface mines
Overburden 30–1, 42
Oxosynthesis process 143
Ozone (O_3) 234, 246, 283
  effect on agriculture 303–5
  effect on grass species 301
  effect on trees 297
  interaction with other pollutants 293, 304

Peat 43, 171
Perfluorocarbons 286
Petrol 12, 126–7, 259
  carcinogenicity 220, 225
  lead in 197–8, 228
Phenols 108, 113, 115, 132, 148, 239, 250
  in gasification processes 263–4, 267–8, 270
Phosphate
  deficiency 41, 43
  in colliery spoil 44
  soil 295
Plant uptake
  of NO_x 289–305
  of PAH 231–2, 236–7
  of trace elements 193, 202–6
Plasma pyrolysis 148–9
Poland 19, 192–3, 294
  coal production 20, 22
  coal reserves in 7
  coal resources in 6, 20
  coal trade 12, 20, 24
  coal waste disposal 40
  colliery spoil salinity 44
  land reclamation in 34, 41
  steam coal 13, 20
Pollution
  acid precipitation 294–7
  air 40, 74, 76–7, 158, 160, 219, 228, 247, 282–305
  atmospheric transport 284–8
  control of 149, 158, 251–5
  deserts 302
  forests 289–97
  freshwater 41, 233–4, 305–10
  from coal conversion processes 268–79, 304–5
  from power plants 160–1, 192–207
  grassland 297–302
  greenhouse effect 283
  noise 74, 76–7
  photochemical 293
  seas and oceans 234–5
  source receptor relationships 285–8
  types of 159, 161, 287
Pollution control 58, 66, 228, 246
  and exposure commitment model 156, 158
  in gasification processes 250–1, 267–9, 278–9
  of sulphur emissions 283
  of trace elements 208
  technologies 246, 252–5, 267, 279
Polycyclic aromatic hydrocarbons (*see also* Benzo (*a*) pyrene) 89, 99, 218–40
  atmospheric 229–30
  background levels of 239
  benzo (*a*) pyrene 160, 219, 224–7, 231, 260–1, 268
  carcinogenicity of 221–2
  degradation of 230, 236
  from automobile emissions 218, 225, 228–9
  from heating 218, 227–8
  from industry 218, 228
  from volcanoes 236–7
  in aviation exhausts 218, 229
  in food 229, 232, 239
  in freshwater 233–4, 239
  in Man 239–40
  in plants 231–2, 236–7
  in soils 225, 229–31, 237
  in solid waste 306
  marine 234–5
  metabolism of 223, 236
  micro-organism synthesis of 236
  plant synthesis of 236

structure of 222–4
Polycyclic organic matter (POM) 227, 237, 246
Potassium (K)
  in colliery spoil 44
  in fly ash 43, 179
  in gasification processes 144–5, 187
  in MHD 88
  radioactive 199
  soil 41, 295–6
Power stations *see* Coal-fired power stations
Process heat
  from nuclear reactors 132, 144, 146
Pulverized fuel combustion 83, 202
  fuel ash (PFA) 83
  sulphur compounds in 91–2
Pyrolysis reactions 87, 219, 258
  AVCO ARC coal process 148
  in gasification processes 132, 147–9, 260
  in liquefaction processes 106
  kinetics of 143
  plasmapyrolysis 105, 148–9

Radioactivity
  emissions 89, 162, 164, 192, 198, 201–2, 257
  exposure commitment model 156, 160, 162–7
Radionuclides 162–5, 192, 201–2, 283, 288
Radium (Ra)
  in air 192
Reclamation 29, 31–45
  economics of 32, 41, 45
  in semi-arid regions 40
  land use efficiency 39
  legislation 32–6
  objectives 31–2
  recent developments 36, 40–3
  site engineering technology 37–8
  soil treatment 38–9
  surface levelling 37–8, 43
  terrace forming levelling 38
  uses of reclaimed land 32, 40
Reclamation strategies 33, 38–40
  agriculture 39–40
  Domsdorf method 42
  fertilization 42–5
  forestry 39–40, 43
  hydraumatic seeding 43
  legumes 42–3, 45
  liming 309
  Koyne method 42
  revegetation 37
  top soil application 42–3
  water recreation and amenity areas 40
Regional coal scenarios 55–7
Rubidium (Rb)
  in gasification processes 144, 187

Saarbergwerke AG process 120
Salinity
  in colliery spoil 44
SASOL liquefaction plant 138–41, 250
  flow diagram 141
Scandinavia 294, 296–7, 307–9
Scandium (Sc)
  in urban aerosols 195
Selenium (Se) 172, 200, 206
  in coal 173–8
  in fly ash 179, 181–5, 194–5
  in gasification processes 187–90, 271–3, 275
  in soil 194, 205
  in urban aerosols 195–7, 208
  in vegetation 203–5
Silicon (Si)
  in gasification processes 187
Sludge ponds
  reclamation 43
Slurry 58, 101
  hydrogenation of 123, 191
  in CLM 85
  in gasification processes 143
  in liquefaction processes 119–20, 123, 129
Slurry pipelines 51, 74
  environmental impacts of 77
  health hazards of 79
  in the USA 52
  water requirement model 60, 66
Sodium (Na) 305
  in colliery spoil 44
  in fly ash 184
  in gasification processes 144, 187
Soil 42
  acidification of 289, 295–6, 310
  bacteria 296
  benzo (*a*) pyrene degradation in 225
  benzo (*a*) pyrene in 225, 229–31, 236–7
  buffering capacity of 296, 307–8
  nitrogen status 295–6, 306
  polycyclic aromatic hydro-carbons (PAH) 229–32, 236–7
  trace elements in 193–5, 202–5, 305
Solid fuels
  share in energy supply 2
Solvent-refined coal (SRC) processes 191
South Africa 250
  coal gasification 138, 277
  coal liquefaction 107–9, 185
  coal production in 20
  coal reserves in 7

  coal resources in 6, 8
  coal trade 23–6
  steam coal 13, 20
South Korea
  coal trade 25
Spain
  coal trade 24–5
Steam coal 12–13, 20, 123
  price of 13
  trade 23–5
Steel industry (*see also* Iron and steel industry)
  in Japan 24
  in USSR 19
  production 12, 25
Strip mining *see* Surface mines
Strontium (Sr)
  in gasification processes 187
Substitute natural gas (SNG)
  from gasification processes 131–2, 138, 141, 145, 188, 190
Sulphur (S) 21–2, 128, 206, 221, 306
  emissions 83, 100
  in coal 98–9, 112, 184
  in coal combustion 92–3, 99
  in fly ash 184
  in gasification processes 190, 256, 259, 262, 268, 272, 274
  in liquefaction processes 139
Sulphur dioxide (SO_2) 85, 88, 91–2, 94, 101, 104
  acid precipitation 289–311
  annual emissions of 91, 161
  deposition 284, 287
  effect on biosphere 289–311
  emissions 785, 160, 246, 283–4
  in gasification processes 256
  interactions with other pollutants 293
  plant tolerance 298–303
  species sensitivity 290–1
  threshold concentration 291–2, 301
Sulphur oxides 90–7, 230
  effects on biosphere 245, 287–311
  emission control 100
Supercritical process 112
Surface mining
  and coal trade 25
  in Australia 20–21, 25
  in Canada 21, 25
  in Czechoslovakia 37
  in France 21
  in Germany 21
  in Latin America 22
  in Poland 21
  in South Africa 20–21
  in UK 21, 42
  in USA 20–21, 25, 54
  in USSR 19, 43
  land disturbance by 29–30, 41
  land reclamation of 38, 41–3
  mining techniques 37–8
  water requirement model 58–9
Synergistic effects 158, 246, 293, 301
Synthesis gas 17, 107–8, 132–3, 138, 142–3, 189
Synthetic fuels 16–18, 58, 123–4, 127, 186, 190, 206
Synthoil process 138–9, 191–2
  catalyst in 191
  trace elements in 191
  water requirements 52

Taiwan
  coal trade 24–5
  steam coal in 13, 24
Terrestrial ecosystems and acid rain 289–305
  beneficial effects of SO_2 and NO_x 306–7
  trace elements in 202, 296, 306
Texaco gasification process 143, 260
  emissions from 149
  flow diagram 269
  health hazards 249
  output stream composition 270–5
Thermal coal 3, 12
Thiocyanides 268
Third World countries *see* Developing regions
Thorium (Th) 199
  atmospheric 192
  in coal 176–8
  in fly ash 179
  in gasification processes 187–90
  in soil 192–3
  in urban aerosols 195
Threshold concentration 157–8, 246
  of COS 304
  of SO_2 291–2, 301
Tin (Sn)
  in gasification processes 187
Titanium (Ti)
  in gasification processes 187
Trace elements
  bioaccumulation of 306
  crustal abundance of 177–8, 181, 187, 196
  emissions of 89, 160, 189, 192–207, 283
  enrichment factors 177, 181–2, 185–9, 194–7, 208
  in animals 203–4, 206
  in coal ash 206
  in effluent streams 186–7, 189, 192, 197, 207
  in fly ash 43, 179–85, 194, 198–207
  in gasification processes 186–90, 206,

    209, 270–1
  in liquefaction processes 191, 206, 209
  in soil 41–2, 192–5, 202–5, 296
  in solid waste 305
  in urban aerosols 195–7
  mobilization 205–7, 209
  organic affinity of 172
Transport *see* Coal transport

U-gas process 107, 142
UN 3, 5
  and energy studies 5
UNSCEAR 156, 162–4
Uranium (U) 199
  atmospheric 192
  combustion characteristics of 180
  in coal 174–8, 202
  in fly ash 179–80, 200
  in gasification processes 187–90
  in soil 192–3, 205
USA 1, 5, 19, 26, 173–4, 181, 204–5, 208, 222, 246
  acid deposition in 289–90, 294, 299, 303, 306–7
  atmospheric pollutants in 76, 161, 192–5, 247–8, 285–6, 293
  benzo (*a*) pyrene in 222, 230, 237
  coal and oil price 13, 26
  coal ash disposal 202
  coal conversion in 17
  coal future development 50–2
  coal gasification in 107, 138, 142, 144, 186, 277
  coal geography 54–5
  coal liquefaction in 107, 186, 190, 248, 250
  coal production 20, 22, 27, 51, 55
  coal reserves 5, 7, 9, 55
  coal resources 5–6, 20, 48–9
  coal trade 20, 24, 26
  coal transport 51–2, 74, 77–8
  coal water requirements 63–6, 68–70
  energy consumption 10
  FBC in 104
  land disturbance in 21, 41
  land reclamation in 34–5, 38, 43
  location of coal 174–7
  nuclear utilization and health 167
  pollution control 251
  steam coal 13
  steel production in 12
  strip mining 20, 37–8, 43, 54
USSR 2, 4–5, 19, 88
  coal conversion processes 18–19, 276
  coal future development 50–2, 54
  coal geography 52–4
  coal liquefaction in 123, 126–9
  coal production in 14, 19, 20, 22, 49, 51
  coal reserves 5, 7, 9, 53
  coal resources 5–6, 19
  coal trade 24–5
  coal transport 52, 74
  coal utilization and health 168
  coal water requirements 63–6, 69–71
  economic growth 11
  energy consumption 11, 14, 52
  land disturbance in 30, 33, 41
  land reclamation in 33, 37, 43
  nuclear fuel utilization and health 168
  PAH in 222, 227–30, 233, 235
  strip mining in 19, 37, 43

Vanadium (V) 207
  in coal 173–8
  in fly ash 179, 181, 192, 200
  in gasification processes 187–90, 271–3, 275
VEBA AG/Ruhrkohle AG process 191
Vegetation
  acid rain effects on 284, 289–90, 294–307
  benzo (*a*) pyrene in 229, 231–2, 236–7
  conifers 290–3
  COS in 304
  deciduous trees 292
  forests *see* Forest ecosystems
  grassland 297–304
  mosses 302–3
  on fly ash 203–5
  trace element accumulation in 306
  trace element enrichment 193, 202–5

Water requirement model 58–65
  constants in 62–3, 65
Wellman-Galusha process 190, 277
WELMM 48–9
Western Europe 2, 26
  coal reserves 9, 12
  coal resources 5
  coal trade 21, 26
  steel production in 12
  steam coal 13
Western industrialized countries 8
  coal production costs 21
  coal production in 19
  economic growth in 26
  trade predictions 27
Winkler process 108, 138
  fluidized bed 84, 132–3
WOCOL study 56
World wars 1, 2

Zimbabwe
  coal production in 22
Zinc (Zn) 172, 305
  global emissions 197–8
  in coal 173–8
  in fly ash 179, 181, 183–5, 192, 194, 200
  in gasification processes 187–90, 271–3, 275
  in soil 192, 202–3, 205
  in urban aerosols 195
  in vegetation 204, 306
Zirconium (Zr)
  in gasification processes 187