ATMOSPHERIC POLLUTION
Its History, Origins and Prevention

Fourth Revised Edition (in SI Units)

A R MEETHAM

in collaboration with

D W BOTTOM A HENDERSON-SELLERS

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Pergamon Press

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BY

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Preface to Fourth Edition

"WHEN the London smog of December 1952 was stirring the public conscience in Britain, American cities such as Pittsburg and Los Angeles had already begun to deal with contamination of town air from sources such as domestic backyard incinerators, industrial chimneys and exhaust gases from motor-cars. Now, after further smog incidents in 1956, 1957 and 1962, and a drastic Act of Parliament, there is good evidence that Britain is following suit. Indeed the realization is growing that the British, with their humid maritime climate and dense industrial population, should be following no one, but leading the world in the technique of ridding the atmosphere of its man-made pollution."

THIS is how the Preface to the Third Edition began. Now, over 20 years after the Clean Air Act of 1956, and indeed 27 years after the senior author produced the first edition, we feel the time is right to present a new edition of "Atmospheric Pollution: its origins and prevention". The three previous editions have, fortuitously, spanned the fascinating history of air pollution control in Britain. The text, initially written to serve as an introduction to air pollution and air pollution control, now stands in its own right as the only authoritative description of our nation's concern with understanding and control of air pollution. Time has produced this metamorphosis from introductory guide to historical review and it is in this light that this new edition is offered and for this reason that the title of the text has now been modified to "Atmospheric Pollution: its history, origins and prevention".

The emphasis throughout the text is upon the evolution of our understanding of sources and controls of air pollution. Since the third edition (published in 1964) enormous strides have been made in the scientific investigation of air pollution; no attempt is made to synopsize these new vast fields of research although reference is made to major current works throughout. However, even a topic as thoroughly investigated as air pollution control does not stand still and the authors feel that in many senses the problems of awareness, monitoring and control of air pollutants have not changed fundamentally from the situation in 1952. Despite Britain's conspicuous success in tackling smoke pollution (described in the third edition) and, more recently, a similar but more limited triumph over sulphur dioxide (described in the National Survey of Air Pollution 1961–1971, Warren Spring Laboratory), there is now the suggestion that particulates (especially lead) and, on a global scale, carbon dioxide and chlorofluorocarbons, may present just as great or greater threats to our environment. Thus the record in this book of earlier approaches and successes may, we suggest, be applied to current problems. The techniques of making surveys of atmospheric pollution; in defining, analysing and explaining pollution levels within geographical areas cannot be divorced from our understanding of dispersion dynamics and meteorological controls of gaseous and particulate constituents.

The interest of the well-educated layman in air pollution and air pollution control has, we feel, also come full circle. The fuel crisis, underlined by the OPEC price increase of 1973, has combined with economic pressures to make well-informed consideration of individual households' fuel use and insulation a sensible, worthwhile and even financially beneficial pursuit.

In keeping with the current trend in scientific literature (and in accordance with the views of the authors), the units used in this new edition will be those of the S.I.—based largely on the metric system. However, since British legislation (especially in boiler practice) is at present still in imperial units these obsolescent units will, where appropriate, be also included in brackets.

Explicit references are gathered at the end of the book together with an extensive and up-to-date bibliography.

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CHAPTER 1

Introduction

THIS book is meant to be of use to all who are professionally interested in atmospheric pollution—environmental health officers, architects, engineers, meteorologists, legislators, city councillors, boiler operators and builders. It is also intended to encourage and help the many citizens who are aware of the degree of atmospheric pollution, and who by their writing, conversation, or example, are helping in the fight against it.

The term pollution is used to describe the admixture of any foreign substance which we dislike with something pleasant or desirable. Atmospheric pollution, therefore, is an undesirable substance mixed with the open air. Any objectionable gas in the air is atmospheric pollution, whether it is harmful or merely unpleasant, but this definition does not apply only to gases. The air frequently contains solid particles or tarry droplets, less than about 10 μm in diameter, which continue in suspension for a long time. Since any particulate matter is liable to cause trouble, these aerosols, as they are sometimes called, are all atmospheric pollution, irrespective of their chemical nature. Also, by general consent, the term atmospheric pollution is applied to larger particles, when these are lifted into the air by the wind or emitted from a chimney. Although the largest particles escape from the atmosphere relatively quickly by falling to the ground, they are capable of causing damage and intense irritation. Finally, there is the risk of radioactive materials in the air, in amounts sufficient to injure public health, whether directly or through selective absorption by food plants and animals.

Some atmospheric pollution, notably that of natural origin, and dust emission from quarries and, more recently, the reintroduction of open-cast mining is produced in the open air. More commonly, the constituents are produced indoors under conditions which are more or less controlled, and are then discharged into the open as the most convenient way to get rid of them. Usually the first thing we do when a room becomes stuffy is to open a door or window; fresh air enters and drives the bad air by some other exit into the open. If pollution is produced very rapidly within a room such natural ventilation may be inadequate, and we equip the room with ventilating hoods, fans, and flue pipes leading to the outside air. Many such installations are to be found in factories where fumes, dusts or gases are liable to be produced; in our own homes every boiler and fireplace has an exhaust flue through which both potential atmospheric pollutants and, perhaps more importantly from an economic point of view, waste heat escape to the atmosphere.

There would seem to be little danger in discharging a small quantity of pollution into the open, where it is so much diluted that it quickly becomes harmless. In 1 sq. km there are about 10,000 tonnes of air below roof level but, when it is realized that as little as one part per million, i.e. 0·01 tonne of pollution in 10,000 tonnes of air, is sufficient to make the air unpleasant to breathe, a simple calculation will show how quickly such a quantity of air may be seriously polluted. Before the Clean Air Acts, domestic combustion sources were commonly responsible for coal consumption at a rate of about 0·5 tonne per minute over every square kilometre of an urban area. On average each tonne of coal produces about 0·05 tonne of pollution, including solid particles and gases such as sulphur dioxide, (but not counting carbon dioxide). When all this pollution was emitted into the atmosphere, as it often was, it was enough to cause serious pollution in the air below roof level in 36 sec. Usually, of course, much of the pollution would spread above roof level and cleaner air would be continually brought in from outside the town and above it; but if this natural ventilation failed, the urban populace would have been gasping for breath in $\frac{1}{2}$ hr, and dead in 5–10 hr.

The success of the Clean Air Act and the switch to other forms of heating sources in preference to coal and smokeless solid fuels has resulted in the replacement of local combustion (domestic coal fires) by centralized combustion (e.g. fossil-fuelled power stations). The total pollution load to the atmosphere is thus not altered, yet the origin of the source is changed. In addition the height of the emission is elevated from roof level (pollution levels compounded by "trapping effects") to several tens or hundreds of metres (power station "tall chimneys").

In the 1950s it was estimated that over 8 million tonnes of atmospheric pollution were produced each year in Great Britain from the combustion of coal and its derived fuels. This caused far more damage than pollution from any other source, even from decaying vegetation, the evaporation of sea spray, or windblown dust. Although current damage due to combustion-derived pollutants is noticeably less it is still important economically and continues to be the most serious source of atmospheric pollution. Pollution from fuel is thus the main subject of this book.

To complete the definition of atmospheric pollution it might seem necessary to enumerate the chemical compounds in it, but this would be an academic exercise of little real value. There is no reason why pollution should not contain any of the naturally occurring chemical elements as well as the man-made or artificial elements (e.g. lawrencium). The various pollution materials which escape from chimneys can be more conveniently divided according to their properties into three groups: (1) the reactive substances, (2) the finest particles, which remain suspended in the air for a long time, ultimately being deposited as dirt on walls, ceilings and other surfaces, and (3) the relatively coarse particles which quickly fall to the ground.

Each group is dominated, as it happens, by one constituent. (1) The reactive substances include sulphur dioxide, sulphur trioxide, carbon monoxide, ammonia, hydrochloric acid, compounds of fluorine and radioactive materials, but sulphur dioxide is by far the most important in ordinary town air. (2) The finest particles are mostly the "smoke" which is produced when fuels are imperfectly burned. (3) The coarser particles are mostly mineral matter and grit from fuel; though smaller in size they are the same material as the ash which collects under a fire, together with particles of unburnt and partly burnt fuel. Much attention will be given to sulphur dioxide, smoke and ash, not only because they are most important but because they serve jointly as prototypes: any other form of pollution when released into the atmosphere will behave similarly to one or other of them.

Growth of pollution

The amount of atmospheric pollution which we endure today is partly the consequence of our living in communities where all kinds of fuel may be burnt, and partly due to our voracious demand for goods manufactured with the help of heat and power which come mostly from fossil fuels. In London, coal was first used on a serious scale in the thirteenth century, when local reserves of firewood were nearing exhaustion. Smoke from this coal soon invoked complaints, and in 1273 Parliament passed an act which prohibited the burning of coal in London. In 1306 an artificer was tried, condemned and executed for this offence. By Elizabeth's time the law had evidently been relaxed, for a deputation of women went to Westminster to see the Queen about "the filthy dangerous poisonous use of coal". Another fruitless effort to rid London of smoke was made by the philanthropist John Evelyn in the reign of Charles II. He proposed that the factories of brewers, dyers, lime-burners, salt and soap-boilers, and others of the same class, should be moved lower down the Thames; and that Central London, as we now know it, should be surrounded by a green belt thickly planted with trees and scented flowers.

In other countries also coal had a limited industrial use. The Italian craftsman Vannoccio Biringuccio wrote in 1540: "Besides trees, stones that occur in many places have the nature of true charcoal; with these the inhabitants of the district work iron and smelt other metals and prepare other stones for making lime for building. But now I do not wish to think of that far-away fuel, for we see that Nature provides things for our every need, and she always generates in abundance of trees".

In England as a whole, and not merely in London, the "abundance of trees" began to be inadequate in the eighteenth century. Before then, goods were manufactured, as the word implies, by hand, and the commonest source of power was the muscular effort of men and animals. There were also water wheels, windmills, and sailing ships but no satisfactory way to convert the energy stored in fuel into mechanical energy had yet been developed. Furnaces were used in a number of industrial processes and, in particular, the iron-workers were heavy consumers of fuel. They required higher temperatures than could be attained by burning raw wood, so they burnt wood charcoal, and though this was an excellent fuel for their purpose, it was obtained by a very wasteful process. The demands of the iron industry, in conjunction with equally wasteful domestic fires and ignorance of the arts of forestry, ultimately caused our national shortage of wood. Moreover, the demands of the iron industry were a major cause of the industrial revolution for men sought a substitute for charcoal, and they discovered how to convert coal into coke which was suitable for metallurgical processes. The coke was mechanically stronger than charcoal, permitting the invention of the blast furnace. This led to an increased production of all forms of iron, and encouraged the invention of engines and machinery made of iron.

In the nineteenth century came the completely unforeseen developments of the industrial revolution. In one manufacture after another, hand-work was replaced by machine-work, and the output of goods increased enormously. Great new industries sprang up in hitherto obscure districts where the requisite raw materials, of which the chief was coal, were found.

New towns arose in a haphazard manner, never keeping pace with the needs of the rapidly increasing population. Life in England was largely changed, ultimately perhaps for the better, since few of us would willingly give up all the advantages that machinery has brought. However that may be, in some ways the change was for the worse, and we are still suffering from the damage undergone by society as a result of those decades of blind economic readjustment. In particular, problems of atmospheric pollution increase very much as a town grows in population density or industrial activity, and for many years the air of most British towns must have rapidly deteriorated.

Our bad habit of polluting the air with waste products of combustion and other chemical processes was formed long ago, but since the beginning of the industrial revolution a minor irritation has become a great social evil. In towns and industrial districts rain water lost its purity; ash and other solids fell continuously to the ground; the air contained a suspension of fine particles which penetrated indoors, to be deposited on walls, ceilings, curtains and furniture; our clothing, our skins, and our lungs have been contaminated; metals corroded, buildings decayed, and textiles wore out; vegetation was stunted; sunlight was lost; our natural resistance to disease was lowered. In a hundred and one ways the miasma of atmospheric pollution lowered our vitality and our enjoyment of life. During the third quarter of the twentieth century public awareness and indeed public outcry against these evils has grown considerably. Citizens are nowadays unwilling to accept that being urban dwellers of necessity implies an increased risk of certain cancers, respiratory illnesses, increased blood lead levels and decreased life expectancy.

Control of pollution

Most progress has been made, so far, against industrial smoke. Smoke is a sure sign that fuel is being wasted, and the manufacturer saves money if he eliminates it by burning his fuel more efficiently. There have been fewer successes against the ash, sulphur dioxide, and other incombustible waste products in industrial flue gases, because there is usually no immediate economic justification for removing them. Since 1933 in America and 1946 in Britain, areas have been marked out in which there are heavy penalties for the emission of smoke, whether from industry or domestic chimneys. Much has already been done to alleviate the pollution in a number of the world's cities. Some advances have been made by individuals and private firms, some by local authorities, and some by central governments. Engineers in Britain, America, France, Germany and other West European countries, Russia and Japan have greatly improved the science of combustion in its application to raw coal, and more efficient combustion has always caused a reduction of smoke. Scientists have developed processes for transforming coal into various smokeless fuels; they have also designed numerous devices for removing dust and sulphur dioxide from flue gases. Manufacturers have tried to keep abreast of advances in engineering and science; their success may be judged by the differences between recent plants and those built 20–50 years ago. Local authorities have fought against pollution with such weapons as they possess, and they have many successes to their credit. The part played by central governments has been to prepare these weapons, by making laws under which local authorities can effectively act, and by sponsoring industrial research. In 1956 the Clean Air Act was introduced following the Beaver Report of 1954. (Some additions and amendments followed in the Act of 1968.) Smoke emissions, and to a lesser degree sulphurous emissions, decreased rapidly. Smoke-control areas were initiated where only smokeless fuels are permitted: London, Manchester and other urban areas were transformed into cities where sunlight amounts were inrceased, incidence of smogs fell and the air quality vastly improved.

Scope of the book

Although atmospheric pollution can be reduced or eliminated in many different ways, each way involves questions of economics, the time factor, availability of materials, priority over other urgent reforms and, to be frank, of individual and social psychology. To provide a basis for consideration of this variety of questions, it is not enough for a book to give information about atmospheric pollution, its measurement, distribution and effects. These matters are important, but there is need to go into the subjects of fuel, fuel-burning applicances, industrial processes, and domestic requirements, not, perhaps, with too technical an approach, but deeply enough to provide at least a foundation for more technical studies.

The next six chapters deal with fuels, furnaces, and fires; the treatment of such subjects has been kept as free as possible from technical jargon. It is necessarily brief, but the reader may find further help in the texts listed in the Bibliography at the end of the book. The eight following chapters are given to a study of the properties of atmospheric pollution. Remedial measures are then considered and the last chapter is an account of the law in Englnnd and in other countries in so far as it concerns atmospheric pollution.

CHAPTER 2

Origin of Fuel

Energy and the Origin of the Earth

ALMOST all the energy available on the Earth can be ultimately attributed to the Sun. The Earth's surface is exposed to solar radiation (1360 W m^{-2}) which heats both ground and atmosphere, drives the atmospheric circulation and, perhaps most importantly, permits photosynthesis. Hence all the fuels based upon plant and animal by-products as well as the "alternative" energy sources such as solar, wind and wave power depend upon the Sun as a radiant source. Only the nuclear industry, which utilizes spontaneous breakdown of naturally occurring radioactive isotopes (for instance uranium-235) has a power source which is older than the Solar System itself since these heavy nuclides were almost certainly derived from supernovae explosions prior to the condensation of the Sun and its planetary system 4·6 thousand million years ago.

Successful use of available fuel resources depends upon a number of fundamental qualities of the Earth and its environment. In particular, the release of heat by burning (i.e. oxidation) is a satisfactorily and easily implemented procedure since approximately 20 per cent of our atmosphere consists of free molecular oxygen.

The atmosphere of the Earth (and probably all the terrestrial planets) is of secondary origin: that is, not a remnant of the solar nebula. This is deduced from the marked depletion of the noble gases in the Earth's atmosphere relative to the solar abundance, which indicates that any primitive atmosphere must have been very rapidly dispersed after the formation of the planet. An internal source for the atmospheres of all the terrestrial planets has been the generally accepted theory since the comprehensive assessment of possible sources for the "excess volatiles" by Rubey in 1951. Outgassing from the planetary surface would have resulted in a predominantly carbon dioxide atmosphere (similar to the atmospheres of both Mars and Venus today). However, the Earth's atmosphere has been continuously reworked throughout the history of the planet due to the presence of large masses of surface liquid water and life. Indeed the free oxygen in the atmosphere is a direct product of the evolution of plant life on Earth, which, since its origin at least 3·4 thousand million years ago, has produced oxygen as a waste product from photosynthesis—the imbalance between photosynthetic combination of carbon into plant cell tissue and reoxidation and return of carbon dioxide to the atmosphere results from rapid burial of non-decayed organic matter.

Fossil Fuels

The solid framework of plants is made of cellulose, whose molecular formula is $(C_6H_{10}O_5)_n$ where n is an unknown number, probably between 1000 and 5000. Plants produce their entire substance out of carbon dioxide and water, with the help of traces of compounds from the soil which contain nitrogen, phosphorus, potassium and other elements.

It is important to observe that if six molecules of carbon dioxide ($6CO_2$) and five molecules of water ($5H_2O$) were simply joined together, the result would be $C_6H_{10}O_{17}$; thus, in forming cellulose, plants reject twelve out of every seventeen oxygen atoms. To do this they require energy, and they get the necessary energy from solar radiation. Conversely, cellulose may be burnt in air, when it takes up the twelve oxygen atoms, forming water and carbon dioxide, with the emission of heat. The other organic substances in plants are formed in a similar way to cellulose, and they too are combustible.

Plants are among the most fundamental form of life because they utilize carbon dioxide, which is a simple inorganic compound. Animals cannot directly use carbon dioxide; they assimilate their carbon from the plants or other animals which they eat. So even the fuels which come from animals are the indirect consequence of vegetable life, and all the organic fuels, being ultimately derived from plants, owe their potential heat-energy to the light and radiant heat of the sun.

Cellulose is a carbohydrate; that is, besides carbon, it contains atoms of hydrogen and oxygen in the proportion 2 : 1, the same as in water. When carbohydrates are burnt, they yield approximately the same quantity of heat as the carbon in them would produce by itself; their hydrogen is of little value as a fuel because it is already chemically bound. Evidently it is a disadvantage for a fuel to contain oxygen, and it is desirable to consider what other natural organic compounds exist which contain a lower proportion of oxygen than the carbohydrates.

The sugars and lignins found in plants and the proteins that constitute a large part of animal substance are little or no better than cellulose as fuels; but in the metabolism of both plants and animals, some carbohydrate is converted into fats, which are stored as reserves of food within the living organism. Fats contain relatively little oxygen, and when burnt they generate about twice as much heat as an equal weight of carbohydrates. Until comparatively recently, beef and mutton tallow were commonly used as fuels, as well as oils extracted from fish and from the seeds of various plants.

Other fuels are made by bacteria from the remains of dead plants and animals and from animal waste products. The richest natural fuel, marsh gas, is made in this way. It consists mainly of methane, CH_4, which has a double advantage as a fuel since it contains a high proportion of hydrogen but no oxygen.

Today the most important fuels are coal, mineral oil and natural gas and these have been produced by slow geological processes. If dead vegetation undergoes prolonged heating or compression it loses oxygen, and though it also loses hydrogen, it changes into a better fuel. In some measure these changes can be copied in the laboratory. By the action of heat only, wood is converted to charcoal which is nearly pure carbon; by the combined action of heat and pressure, wood or damp cellulose can be converted to a