Atmospheric Pollution

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ATMOSPHERIC POLLUTION

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Preface

Evidence shows that the first primitive living cells developed in an atmosphere very different from the existing one of today. The early atmosphere is believed to have consisted of some mixture of methane, carbon monoxide, carbon dioxide, water vapor, ammonia, and nitrogen, not at all conducive to present forms of life. Oxygen is thought to have been available in small amounts produced by the slow processes of photolysis and thermal dissociation of water, the effect of which would merely lead to a small amount of local oxidation at the earth's crust.

Analysis of fossil records indicates that the earth's atmosphere has undergone a sequence of qualitative changes. These prehistoric changes are reflected in adaptive changes in living matter to conform with environmental variations. The appearance of oxygen in the atmosphere, the result of photosynthesis, led to the evolution of cells that could survive its toxic effects and eventually to cells that could capitalize on the high energy levels of oxidation metabolism. There is also evidence that many species did not adapt and in some cases ceased to exist.

Although the atmosphere's composition has remained relatively constant over the most recent past there has been widespread evidence of localized variation in quality. Such variations are viewed as deviations from the atmospheric norm and are implicit in the use of the term "atmospheric pollution." Indeed, it has been the extreme atmospheric concentrations of pollutants resulting in dramatic disasters in the area of public health that have given impetus and importance to the study of atmospheric pollution.

The extent to which atmospheric composition will be affected in the long term depends upon the capacity of assimilative processes that provide an acceptable ecological equilibrium.

There is little information that suggests air pollution was an important problem up to the end of the thirteenth century. From the fourteenth century and onward when coal began to be used extensively in Europe for domestic heating and primitive industrial processes, coal smoke and gas was by far the most important air pollutant. Coal was not considered to be a "natural" fuel as was wood and the noxious sulphur compounds of combustion provided a constant reminder of this concern. An especially irritating combination of coal smoke and fog popularly known in the United Kingdom as "smog" appears to have caused substantial increases in urban area death rates recorded in the early 1900s and before. Much of the early information on these episodes has come from retrospective examination of vital records. The use of oil and gas as both domestic and industrial fuels has served to reduce the coal smoke problem. However, it is still an important contributor to air pollution in many industrialized and urban areas.

The introduction of petroleum and natural gas has had major impact on all sectors of energy use. Most noteworthy are the changes brought about in transportation and the provision of raw materials for a vast array of petrochemical products. These changes have had their own impacts upon the atmosphere. The potential hazards of internal combustion engine exhaust were pointed out as early as 1915. In 1954, A. J. Haagen-Smit of the California Institute of Technology was the first to identify the constituents and reactions involved in the production of photochemical smog. He showed that hydrocarbons, nitrogen oxides, and ozone interreact by photochemical processes to produce the Los Angeles type smog. The primary source of hydrocarbons and nitrogen oxides in the Los Angeles area is the motor vehicle, the density of which increased dramatically as a result of the wartime development in the 1940s. Many cities of the world approach having all the conditions necessary to create severe episodes of photochemical smog. The conditions include prolonged periods of air stagnation, abundant sunlight, and elevated emission of the primary pollutant inputs of hydrocarbons and nitrogen oxides. Most of these cities lack only the high motor vehicle densities. Tokyo already has all of the necessary conditions and the problem.

It was not until 1977 that photochemical smog was first identified as a problem in Europe, and the effects of photochemical oxidants on the environment are now rapidly coming into focus. Originally considered to be an urban problem, it has begun to take on a regional character. High ozone concentrations exceeding air quality standards have been observed over widespread rural areas located downwind of major metropolitan centers. Of recent concern is the role of photochemical oxidants in the formation of acidic substances as related to acid deposition.

The first sign of regional air pollution in northwestern Europe was that of acid precipitation. This resulted in the acidification of lakes and rivers in Scandinavia because the soils there were unable to neutralize the increased acidity due to precipitation. An OECD study on the acidification of precipitation and the long range transport of air pollution begun in 1972 showed that significant amounts of sulphur pollutants were causing serious contamination of the atmosphere over the region. Also, much of this air pollution was being transferred from sources in other industrialized and densely populated areas, sometimes over many hundreds of kilometers. More recently acid precipitation has been identified as a serious problem in North America resulting in similar effects on the environment due to the widespread distribution of sulphur pollutants.

As suggested in this brief historical perspective, the importance of atmospheric quality has been recognized only during the past few decades. During this time there have been many important developments in the understanding of the atmospheric pollution problem. Contributions to this understanding have come from the physical, biological, and social sciences. The study of atmospheric pollution is not only an interdisciplinary field, but as a science it spans the spectrum from basic research to operational use. There is also the need to identify new emerging problems in the field and to develop better methods of dealing with existing problems.

This brings into focus the purpose of this volume and the type of audience for which it has been prepared. The object is to package the more important aspects of current atmospheric pollution technology in an organized manner and present this mainly technical material to a group of specialists with various scientific backgrounds but with little, if any, experience in dealing with such problems. The majority of the material presented in these papers represents state-of-the-art methods of dealing with air quality problems. Most of the papers also contain basic background material and are extensively referenced allowing the reader to further consult the literature. Although the topics dealt with are wide ranging and attempt to capture the most current approaches, they are not intended to be complete in the sense that all aspects of the topic are covered.

The material contained in this volume was presented in Damascus, Syria, in August 1985 to a group of university and government research scientists from a number of Middle East countries. To provide motivation and topical discussion, an attempt was made to include material of special interest to these participants. Evidence of this is most obvious in the chapters dealing with sources of emissions, air pollution controls, and the short papers on applications.

The book is organized in nine parts, each containing one or more individual papers, along the following lines:

1. Atmospheric pollution and planning studies
2. Emission sources
3. Pollutant formation and transformation
4. Effects on man and vegetation
5. Air pollution control
6. Atmospheric dispersion and modelling
7. Sampling and analyses
8. Monitoring air quality
9. Applications

The purpose of the discussion that follows is to introduce each of the above topics. Readers with special interests must refer to the table of contents for relevant papers by topic.

Part 1: A general discussion of issues fundamental to the study of air quality problems and the planning of air pollution studies can be found in this part. There are many different factors which must be considered when planning such a study. These include the identification of sources and types of emissions, the behaviour of different pollutants in the atmosphere, the dynamic structure of the atmosphere and the effects on pollutant dispersion and deposition. In practice it is found that a number of simplifications must be made due to the complexity of the atmospheric processes involved. What should be included in the scope of a study, and the manner in which this should be done is highly dependent on the time and space resolution required. The measurement program must be planned accordingly. These factors are discussed in the context of local, urban, regional, and global scale problems.

Part 2: The sources that contribute to atmospheric pollution are both natural and man made and are the subject of Part 2. Significant natural sources of emissions include: windblown dust, volcanic emissions, combustion products from forest fires, vegetation, and sea-salt spray. The relative contribution of natural sources to ambient levels exhibits a marked variation depending upon the compound measured and the locations selected for measurement. As pointed out in this section, such sources, particularly wind blown dust, are considered to be significant in Middle East countries.

Major anthropogenic sources of atmospheric pollution in these countries include the petrochemical industry, power generation stations, cement and fertilizer plants, and the internal combustion engine. Fumes generated by households from heating and cooking in densely populated areas, in combination with emissions and dust from motor vehicles and industry, can form serious urban air pollution problems.

In the Middle East emissions tend to be concentrated in certain places with practically zero emissions between these places. On a regional scale these sources may be considered as large point sources or urban plumes.

For practical purposes, one may distinguish between pollutants which occur in such quantities that acute effects may be observed near the sources, and pollutants which appear in smaller amounts. The latter may give rise to long term effects due to accumulation in the environment or in the human body. Well known pollutants in the first group include the sulphur, nitrogen, and carbon oxides, and their hydrogen acids. There are also their components which appear in less specific forms as particles and hydrocarbons.

In the second group are found various trace elements such as zinc, cadmium, and lead, and some highly stable groups of organic components such as the polycyclic aromatic hydrocarbons (PAH) and a number of chlorinated hydrocarbons e.g., the polychlorinated biphenyls (PCBs) and many pesticides well known for their toxic effects.

The more important emissions in connection with regional contamination are sulphur dioxide, nitrogen oxides, and hydrocarbons. In climates that are relatively dry and with a large amount of sunlight, it is considered that photochemical oxidants could become a serious pollution problem.

The widespread use of pesticides in agriculture leads to their increased concentration in the atmosphere. They are disseminated into the air by crop-spraying and evaporation from soils and plant foliage. These are chemically stable substances and when entrained as dust have the potential of being transported over long distances by air masses.

Part 3: In spite of the unchanging nature of the atmosphere's major components, it is in reality a dynamic system with its gaseous constituents continuously being exchanged with oceans, vegetation, and biological organisms. Also present are a number of gases and particulate matter that occur in small but sometimes highly variable amounts. Among these are the common air pollutants, the formation and transformation of which is the subject of this section.

Gases are produced by chemical processes within the atmosphere itself, by biological activity, volcanic emissions, and the industrial activity of man. They are removed from the atmosphere by chemical reactions, biological activity, physical processes in the atmosphere such as aerosol formation, and by exchange with the soil and water. Fossil fuel combustion is the major source of inorganic gaseous air pollution. Sulphur and nitrogen oxides are formed by the oxidization of sulphur contained in the fuel and of the nitrogen from either the fuel or the atmosphere. Thermodynamic theory can be used to predict the extent of oxidation of these elements at elevated temperatures typical of combustion exhaust gases.

Sources of particulate matter or aerosols can be classified as primary or secondary. Primary aerosols are those emitted in particulate form directly from sources such as dust or smoke. Secondary aerosols refer to particles produced in the atmosphere. Secondary aerosol sources of natural origin are associated with gas phase chemical reactions which produce particulated phase material through condensation. Particulate matter can be distinguished on the basis of size and chemical composition. Once aerosols are in the atmosphere, their size, number, and chemical composition can be changed and removed by several mechanism. Many of these transformation and removal processes are discussed in these chapters.

Part 4: There is substantial evidence that air pollution affects the health of humans and animals, damages vegetation and materials, reduces visibility, contributes to safety hazards, and generally has a deleterious effect on the well being of the biota. Although certain of these effects are specific and measurable, such as damage to materials and reduced visibility, there is a significant degree of uncertainty in establishing firm relationships between air pollution and effects on the biota. Cause and effect relationships, frequently so firmly established in the physical sciences are not so easily obtained when biological organisms are involved.

Further complicating the determination of effects of air pollutants on the biota is the manner in which they interact to produce a synergistic effect in some combinations and an antagonistic effect in others.

There are, however, reasons for optimism. The development of newer techniques in toxicology, for example, is one of the elements which has led to an increased awareness of the potential effects of air pollution. Today pollutants can be measured in terms of extremely small amounts both in the atmosphere and within the organism.

Knowledge of health effects have been enhanced through three types of investigations: (1) retrospective statistical studies of past morbidity and mortality which correlate geographic location, meteorological data, and other physical and demographic factors with air pollution, (2) epidemiological studies of relations of morbidity, mortality, and respiratory functions to variations in air pollution, (3) laboratory studies which document animal and human responses associated with air pollution.

Investigations of the foregoing types provide associative evidence only and do not establish a firm cause-and-effect relationship.

The study of air pollution effects on vegetation is important because of the economic losses incurred and because vegetation injury provides an indication of how air pollutants may eventually affect man. Monitoring of sensitive plant species can enable the determination of specific air pollutant presence and the relative concentration that caused the plant injury. However, caution must be used in diagnosing apparent pollutant injuries to vegetation since synergistic effects of multiple pollutants can occur. Also factors such as nutrient deficiencies, insecticide damage, and plant physiological disturbances complicate true evaluation of air pollution damage.

Part 5: The management of air quality requires the regulation of pollutant emissions to achieve some clearly defined set of ambient air quality standards or goals. This implies the existence of control techniques which invariably are constrained by their economic and technical feasibility. This chapter is concerned with the measurement, documentation, and control techniques in air pollution abatement.

Emission inventories provide a basis for planning control strategies. Basic data on source category, pollutant characteristics, and location of industrialized areas are now collected in many countries. Emissions may be classified as to whether they are stationary or moving, or, whether they constitute a point, line, or area source.

Control techniques for limiting pollutant formation and emission fall into three classes: (1) purification or substitution of raw materials and fuels to decrease pollutants, (2) process change and equipment modification, for example, to make combustion more efficient, (3) cleaning of emissions before release to the atmosphere.

Of these three types of control techniques, the cleaning of source emissions is the most widely used. Control devices are generally designed to control either gaseous pollutants or particulate pollutants, since few devices are efficient in the control of both. Some devices are designed for a specific type of pollutant or specific size of particulate. The following considerations may be of importance where a particular control device is to be selected: identification of specific gases and particulates to be controlled; the recovery of product; the recovery of heat; the disposal facilities involved.

Part 6: The goal of air quality modelling is to supply information for decisions to be made within the framework of environmental planning and management systems. The fundamental problem is to calculate the atmospheric concentrations of pollutants in space and time given such independent variables and parameters as meteorological variables, emission rates, transformation, and removal rates. Atmospheric dispersion and modelling is concerned with the solution of this problem.

For the purpose of introduction to this voluminous topic, models are classified into three broad categories: (1) physical models, (2) statistical models, (3) mathematical models of atmospheric transport.

Physical models are intended to simulate the behavior of pollutants in the atmosphere using scaled-down models of physical reality in wind tunnels. Fluid models have the advantage that scale-model geometry, flow velocities, and other essential variables can be easily controlled. The critical area of concern in this type of modelling is to ensure that the scaling criteria adequately represents the full scale case. Physical modelling is discussed only briefly in Part 6, in relation to the aerodynamic effects of structures on pollutant dispersion.

Statistical models attempt to establish statistical relationships based on past air quality monitoring data. For example, a particular relationship might describe the current pollutant concentration at a location as a function of lagged concentrations at other locations and the prevailing meteorological conditions. Statistical models are also useful in dealing with probabilistic questions such as determining the probability that a concentration level will be exceeded given a certain source emission level. Statistical models are used when a relatively large amount of monitoring data are available. This could amount to several years of data taken at frequent intervals at a number of monitoring stations. Because of data constraints and the specialized conditions under which statistical models are appropriate, their introduction is delayed until Part 8, where a particular implementation is discussed relating to the design of air quality monitoring networks.

The focus in Part 6 is on mathematical models of atmospheric transport that are based on the equations of mass conservation for individual pollutants. They are characterized by a fundamental description of atmospheric motion and physical-chemical processes which influence the mass balance and continuity of an air parcel. A model of this type requires, as part of its formulation or as data input, information on emissions, meteorology, and atmospheric chemistry and removal processes. Mass conservation models may be formulated as time-varying or steady state and both temporal and spatial scales can vary widely.

There are two fundamental modelling approaches that are based on the mass conservation equation. The

Eulerian approach describes the pollutant concentration relative to a fixed coordinate system. The Lagrangian description of concentration change is made relative to the moving fluid. The two approaches produce different mathematical representation with different solution characteristics.

The most frequently used approach to dispersion modelling is the Gaussian diffusion formulation. This approach is based on the solution to Ficks' diffusion equation which is the well-known Gaussian distribution function. The Gaussian formulation has a number of different versions. The point source form can be integrated in one or two dimensions to yield line and area representations.

Part 7: Sampling and analysis of air pollutants are usually treated as a single topic due to the interdependence of the steps needed to arrive at reliable quantitative determinations. In general, the sequence of steps consists of: sample collection using appropriate instruments and methods for particular pollutants; special treatment or refinement of the sample; sample analysis and quantitative determination of the pollutants collected. This Section contains a survey of some of the more important techniques currently available to obtain samples and to analyse them.

Sampling is the most critical of the previously mentioned steps since a representative sample is needed in order to obtain reliable results. Furthermore, due to the low concentrations of some components it is normal to preconcentrate them during the sampling of much larger volumes of air. After this step the problem is to transfer the components in the sample to be analytically processed without any change in their physical or chemical state. Thus, any artifact formation during sampling must be avoided.

Measures must often be taken in the case of liquid samples to avoid decomposition. For example some possible methods to stabilize rain water involve the addition of acids or biocides and storing the samples at a temperature of 4°C. In the case where reactive trace constituents are present in the rain water the samples should be analysed immediately after collection.

It is sometimes convenient to remove suspended particulate matter from liquid samples by centrifuging or by filtration. In filtering, the proper filter material must be carefully chosen because filters may absorb trace constituents from a solution or may contaminate a sample.

A typical operation carried out as part of sample pretreatment is the desorption of collected gases from solid sorbents. If this is done by heating, the temperature must be kept low enough to avoid decomposing the substance of interest or the sorbent itself.

After sampling and pretreatment many methods exist for the quantitative analysis of the collected gaseous, liquid, and solid material. A summary of those techniques is given which lend themselves to a broad range of applications.

Two statistical studies which illustrate the careful application of sampling and analysis methods are discussed in Part 7. The first was carried out in Beijing, China and involves the estimation of atmospheric heavy metal exposure from re-entrained dust and the degree to which the dust had been contaminated above natural concentrations. The second study deals with the collection and use of time sequenced concentration measurements to investigate the transport of particulate matter from sources in Asia to Japan and Hawaii.

Part 8: Air quality monitoring network design involves selecting and locating observation sites and scheduling observations in order to meet network objectives. A systematic approach to the design of monitoring networks is dealt with in this section. The associated design criteria of network accuracy and precision are an important part of the design procedure. Also involved is the setting of specifications for individual stations in the network, including the selection of equipment, instrumentation, sampling and analysis procedures, and data recording methods.

The main emphasis in this part is on the basic concepts and representative techniques of monitoring network systems design. The task of monitoring ambient air quality involves the sampling of a dynamic and complex environmental process which exhibits large variations in space and time. It is desirable to reduce both spatial and temporal variability by locating observation sites and scheduling observations so that they are representative of the space and time scales specified as part of the network objective.

A primary consideration in air monitoring survey design is cost. Total costs depend on the number of sampling stations, sampling frequency, and the period over which sampling takes place. Because of the nature of these costs it is important to minimize the number of sites and the sampling frequency while maintaining a spatially distributed sample size to obtain timely information with the desired level of confidence.

Methods used to perform air quality network design may be classified into three broad categories: modelling methods, statistical methods, and a combination of these two approaches. The method used will depend upon the monitoring objective and the availability of adequate data.

Part 9: The last section contains six papers all contributed by scientists from Middle East countries. Problems of atmospheric pollution dealt within these papers are not significantly different from the types of problems encountered elsewhere. They are, for the most part, concerned with the identification and solution of the problems associated with increasing industrialization and population densities.

One difference that is apparent is the absence of air pollution regulations and standards on a country-wide basis. Studies of the type described in this part will do much to help in establishing the proper ground work upon which to base a reasonable set of standards and controls.

In conclusion, it may be of interest for the reader to know that this volume is not the result of an isolated event. For the past several years the Arab School of Science and Technology has presented Summer Sessions on topics of current technological and scientific significance. The session on Atmospheric Pollution was a continuation of the tradition and represented the second session in the "Environment Series" of the School. It was held in Zabadani, near Damascus in August 1985. The location and surrounding region provided an excellent setting for lecturers and participants alike to experience professional, cultural, and social growth. The program committee comprised of Drs. A. Habel and F. Abousamra designed an agenda that provided an excellent balance between theory and application using formal lectures, panel discussions, and supplementary sessions. The School director, Dr. Nabil Harfouch, and his very efficient staff supplied the advice, assistance, and administrative functions so necessary to mount a program of this nature. It has been a pleasure to have been associated with this project.

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ATMOSPHERIC POLLUTION AND PLANNING STUDIES

Planning of Air Pollution Studies

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1 INTRODUCTION

The main subject of this presentation will be the planning of air pollution studies, i.e. how we use emission surveys, meteorological data and chemical measurements of gases, aerosols and precipitation to examine the air pollution situation in a given area in order to obtain a realistic basis for future abatement policies. In the first instance this requires a general understanding of the main chemical and physical processes taking place. Much of this will be discussed in greater detail in other presentations. The introductory chapters on sources and atmospheric dispersion are therefore limited to a brief description of conditions which are essential in the planning of air pollution studies.

Air pollution from the combustion of fuels and noxious odours from various production processes have for a long time been a characteristic of major cities. Since the middle of this century, it has been realized that combustion products also are affecting atmospheric chemistry outside urban and industrialized areas, and during the last decade it has become evident that air pollutants may contaminate substantial parts of the global atmosphere.

Since the industrial revolution started 200 years ago with coal as the main energy source, the introduction of oil and gas in this century has tremendously increased the industrial production capacity of our world, and the mobility of man. The accompanying spread of information and technology has led to higher demands on all kinds of consumer's goods. This general development has given air pollution a new dimension during the last 30-40 years.

As a consequence, the concentration of carbon dioxide has increased by 15% from approximately 295 ppm by the turn of the century, to a level of about 345 ppm at present. The haze formed over industrialized areas such as the European continent and the eastern part of North America and the adjoining seas, is easily recognized from aircraft and satellite observations (1,2), and the long range transport of gases and aerosol particles and the acidification of the precipitation are now well documented (3,4). Other industrialized areas also contribute significantly to the world wide pollution of the atmosphere (5).

Actually, the amounts of atmospheric contaminants released by the combustion of fossil fuels, and the extensive use of many chemicals, make it necessary to consider seriously the capacity of the atmosphere and our environment to receive these pollutants without substantial adverse effects. In order to do this, the emission sources, atmospheric transport mechanisms and sinks must be known.

In this connection it should, however, be born in mind that also the clean atmosphere, in addition to its main components of nitrogen, oxygen, water and carbon dioxide, contains highly variable amounts of gaseous and particulate components from wind erosion, biological activities, forest fires, lightning, volcanic eruptions, etc. By air pollutants we mean substances which are emitted to the atmosphere by the activities of man. The air is polluted if the concentrations reach levels which may have negative effects on our health, our environment, or materials we use. Thus, the significance of many pollutants depends on the amounts emitted in relation to the natural background and their chemical persistency, as well as their dispersion and rate of deposition. In particular, this applies to several trace elements and the many polychlorinated and other halogenated hydrocarbons. The latter, which do not occur naturally at all, have physical properties unknown in nature, and some, i.e. the dioxins, are among the most poisonous substances we know.

2 THE AIR POLLUTANTS

The major emissions of air pollutants are due to the use of fossil fuels for heat and energy production, including motorized traffic. In addition come the emissions from industrial production processes which in some areas may be the dominating source, and the large scale use of many industrial products, e.g., fertilizers, solvents, pesticides, freons, etc. The agglomeration of the population in cities and densely populated areas has added to the problems. While the smoke from a single country house worries no one, the fumes from thousands of fireplaces and motor vehicles crammed together in a city, may grow into a serious problem. The concentration of cities and industries in certain regions leads to similar problems on a larger scale.

The growing use of fossil fuels in Europe during this century is illustrated in Figure 1 (6). The early sulphur dioxide emissions in Europe were mainly due to the combustion of sulphur containing coals and in some areas the processing of sulphidic ores. The increased demand for energy after 1950 was met with a wide-spread introduction of petroleum products, and the result was a doubling of the sulphur dioxide emissions in Europe during the period 1950-75. Later they have not changed much. During the last 10 years the emissions of sulphur dioxide have become less in many Western European countries, mainly because of a shift from coal to gas, while increased energy production based on traditional fuels, has led to larger