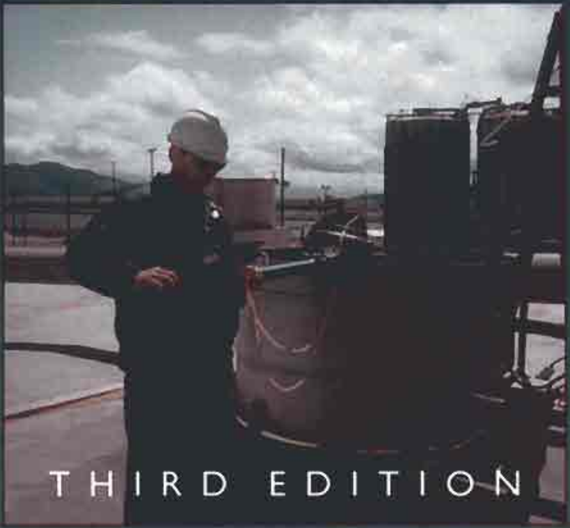
Fundamentals of Air Pollution



THIRD EDITION

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Fundamentals of Air Pollution

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Dedication

Dedicated to Arthur C. Stern, the pioneer in air pollution science and engineering, by three humble followers. He was our mentor, taskmaster, and friend.

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Preface

The authors of this book include a chemist (Donald L. Fox), a meteorologist (D. Bruce Turner), and a mechanical engineer (Richard W. Boubel). This 1:1:1 ratio has some relevance in that it approximates the ratio of those professionally involved in the field of air pollution. In the environmental protection and management field, the experience of the recent past has been that physicists and electrical engineers have been most attracted to the radiation, nuclear, and noise areas; biologists and civil engineers to the aquatic and solid waste areas; chemists, meteorologists, and chemical and mechanical engineers to the area of air pollution and its control. These remarks are not intended to exclude all others from the party (or from this course). The control of air pollution requires the combined efforts of all the professions mentioned, in addition to the input of physicians, lawyers, and social scientists. However, the professional mix of the authors, and their expectation of a not-too-dissimilar mix of students using this book, forewarns the tenor of its contents and presentation.

Although this book consists of six parts and three authors, it is not to be considered six short books put together back-to-back to make one large one. By and large, the several parts are the work of more than one author. Obviously, the meteorologist member of the author team is principally responsible for the part of the book concerned with the meteorology of air pollution, the chemist author for the chapters on chemistry, and the engineer author for those on engineering. However, as you will see, no chapters are signed, and all authors accept responsibility for the strengths and weaknesses of the chapters and for the book as a whole.

In the twenty years since publication of the first edition of *Fundamentals of Air Pollution* (1973), and the nine years since the second edition (1984), the fundamentals have not changed. The basic physics, chemistry, and engineering are still the same, but there is now a greater in-depth understanding of their application to air pollution. This edition has been edited, revised, and updated to include the new technology available to air pollution practitioners. Its contents are also influenced to a great extent by the passage of the U.S. Clean Air Act Amendments of 1990 (CAAA 90). These amendments have changed the health and risk-based regulations of the U.S. Clean Air Act to technology-driven regulations with extensive penalty provisions for noncompliance.

We have added more detailed discussion of areas that have been under intensive study during the past decade. There has been a similar need to add discussion of CAAA 90 and its regulatory concepts, such as control of air toxics, indoor air pollution, pollution prevention, and trading and banking of emission rights. Ten more years of new data on air quality have required the updating of the tables and figures presenting these data.

We have expanded some subject areas, which previously were of concern to only a few scientists, but which have been popularized by the media to the point where they are common discussion subjects. These include "Global Warming," "The Ozone Hole," "Energy Conservation," "Renewable Resources," and "Quality of Life."

With each passing decade, more and more pollution sources of earlier decades become obsolete and are replaced by processes and equipment that produce less pollution. At the same time, population and the demand for products and services increase. Students must keep these concepts in mind as they study from this text, knowing that the world in which they will practice their profession will be different from the world today.

By virtue of its division into six sections, this text may be used in several ways. Part I, by itself, provides the material for a short course to introduce a diverse group of students to the subject—with the other five parts serving as a built-in reference book. Parts I, II, and II, which define the problem, can provide the basis for a semester's work, while Parts IV, V, and VI, which resolve the problem, provide the material for a second semester's work. Part IV may well be used separately as the basis for a course on the meteorology of air pollution, and the book as a whole may be used for an intensive one-semester course.

The viewpoint of this book is first that most of the students who elect to receive some training in air pollution will have previously taken courses in chemistry at the high school or university level, and that those few who have not would be well advised to defer the study of air pollution until they catch up on their chemistry.

The second point of view is that the engineering design of control systems for stationary and mobile sources requires a command of the principles of chemical and mechanical engineering beyond that which can be included in a one-volume textbook on air pollution. Before venturing into the field of engineering control of air pollution, a student should, as a minimum, master courses in internal combustion engines, power plant engineering, the unit processes of chemical engineering, engineering thermodynamics, and kinetics. However, this does not have to be accomplished before taking a course based on this book but can well be done simultaneously with or after doing so.

The third point of view is that *no one*, regardless of their professional background, should be in the field of air pollution control unless they sufficiently understand the behavior of the atmosphere, which is the feature that differentiates *air* pollution from the other aspects of environmental protection and management. This requires a knowledge of some basic atmospheric chemistry in addition to some rather specialized air pollution meteorology. The viewpoint presented in the textbook is that very few students using it will have previously studied basic meteorology. It is hoped that exposure to air pollution meteorology at this stage will excite a handful of students to delve deeper into the subject. Therefore, a relatively large proportion of this book has been devoted to meteorology because of its projected importance to the student.

The authors have tried to maintain a universal point of view so that the material presented would be equally applicable in all the countries of the world. Although a deliberate attempt has been made to keep American provincialism out of the book, it has inevitably crept in through the exclusive use of English language references and suggested reading lists, and the preponderant use of American data for the examples, tables, and figures. The saving grace in this respect is that the principles of chemistry, meteorology, and engineering are universal.

As persons who have dedicated all or significant parts of their professional careers to the field of air pollution, the authors believe in its importance and relevance. We believe that as the world's population increases, it will become increasingly important to have an adequate number of well-trained professions engaged in air pollution control. If we did not believe this, it would have been pointless for us to have written this textbook.

We recognize that, in terms of short-term urgency, many nations and communities may rightly assign a lower priority to air pollution control than to problems of population, poverty, nutrition, housing, education, water supply, communicable disease control, civil rights, mental health, aging, or crime. Air pollution control is more likely to have a higher priority for a person or a community already reaping the benefits of society in the form of adequate income, food, housing, education, and health care than for persons who have not and may never reap these benefits.

However, in terms of long-term needs, nations and communities can ignore air pollution control only at their peril. A population can subsist, albeit poorly, with inadequate housing, schools, police, and care of the ill, insane, and aged; it can also subsist with a primitive water supply. The ultimate determinants for survival are its food and air supplies. Conversely, even were society to succeed in providing in a completely adequate manner all of its other needs, it would be of no avail if the result were an atmosphere so befouled as not to sustain life. The long-term objective of air pollution control is to allow the world's population to meet all its needs for energy, goods, and services without sullying its air supply.

Part I

The Elements of Air Pollution

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1

The History of Air Pollution

I. BEFORE THE INDUSTRIAL REVOLUTION

One of the reasons the tribes of early history were nomadic was to move periodically away from the stench of the animal, vegetable, and human wastes they generated. When the tribesmen learned to use fire, they used it for millennia in a way that filled the air inside their living quarters with the products of incomplete combustion. Examples of this can still be seen today in some of the more primitive parts of the world. After its invention, the chimney removed the combustion products and cooking smells from the living quarters, but for centuries the open fire in the fireplace caused its emission to be smoky. In AD 61 the Roman philosopher Seneca reported thus on conditions in Rome:

> As soon as I had gotten out of the heavy air of Rome and from the stink of the smoky chimneys thereof, which, being stirred, poured forth whatever pestilential vapors and soot they had enclosed in them, I felt an alteration of my disposition.

Air pollution, associated with burning wood in Tutbury Castle in Nottingham, was considered "unendurable" by Eleanor of Aquitaine, the wife of King Henry II of England, and caused her to move in the year 1157. One hundred-sixteen years later, coal burning was prohibited in London; and in 1306, Edward I issued a royal proclamation enjoining the use of *sea-coal* in furnaces. Elizabeth I barred the burning of coal in London when Parliament was in session. The repeated necessity for such royal action would seem to indicate that coal continued to be burned despite these edicts. By 1661 the pollution of London had become bad enough to prompt John Evelyn to submit a brochure "Fumifugium, or the Inconvenience of the Aer, and Smoake of London Dissipated (together with some remedies humbly proposed)" to King Charles II and Parliament. This brochure has been reprinted and is recommended to students of air pollution (1). It proposes means of air pollution control that are still viable in the twentieth century.

The principal industries associated with the production of air pollution in the centuries preceding the Industrial Revolution were metallurgy, ceramics, and preservation of animal products. In the bronze and iron ages, villages were exposed to dust and fumes from many sources. Native copper and gold were forged, and clay was baked and glazed to form pottery and bricks before 4000 BC Iron was in common use and leather was tanned before 1000 BC. Most of the methods of modern metallurgy were known before AD 1. They relied on charcoal rather than coal or coke. However, coal was mined and used for fuel before AD 1000, although it was not made into coke until about 1600; and coke did not enter metallurgical practice significantly until about 1700. These industries and their effluents as they existed before 1556 are best described in the book "De Re Metallica" published in that year by Georg Bauer, known as Georgius Agricola (Fig. 1-1). This book was translated into English and published in 1912 by Herbert Clark Hoover and his wife (2).

Examples of the air pollution associated with the ceramic and animal product preservation industries are shown in Figs. 1-2 and 1-3, respectively.

II. THE INDUSTRIAL REVOLUTION

The Industrial Revolution was the consequence of the harnessing of steam to provide power to pump water and move machinery. This began in the early years of the eighteenth century, when Savery, Papin, and Newcomen designed their pumping engines, and culminated in 1784 in Watt's reciprocating engine. The reciprocating steam engine reigned supreme until it was displaced by the steam turbine in the twentieth century.

Steam engines and steam turbines require steam boilers, which, until the advent of the nuclear reactor, were fired by vegetable or fossil fuels. During most of the nineteenth century, coal was the principal fuel, although some oil was used for steam generation late in the century.

Fig. 1-1. Lead smelting furnace. Source: G. Agricola, "De Re Metallica," Book X, p. 481, Basel, Switzerland, 1556. Translated by H. C. Hoover and L. H. Hoover, *Mining Magazine*, London, 1912. Reprinted by Dover Publications, New York, 1950.

The predominant air pollution problem of the nineteenth century was smoke and ash from the burning of coal or oil in the boiler furnaces of stationary power plants, locomotives, and marine vessels, and in home heating fireplaces and furnaces. Great Britain took the lead in addressing this problem, and, in the words of Sir Hugh Beaver (3):

> By 1819, there was sufficient pressure for Parliament to appoint the first of a whole dynasty of committees "to consider how far persons using steam engines and furnaces could work them in a manner less prejudicial to public health and comfort." This committee confirmed the practicability of smoke prevention, as so many succeeding committees were to do, but as was often again to be experienced, nothing was done.
>
> In 1843, there was another Parliamentary Select Committee, and in 1845, a third. In that same year, during the height of the great railway boom, an act of Parliament disposed of trouble from locomotives once and for all (!) by laying down the dictum that they must consume their own smoke. The Town Improvement Clauses Act

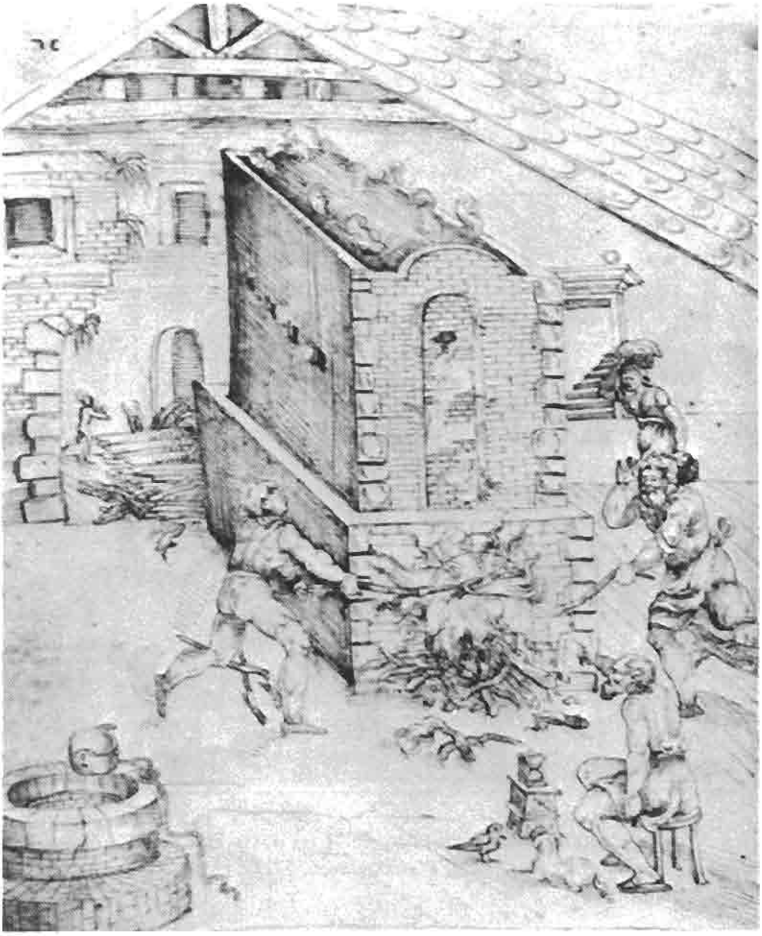

Fig. 1-2. A pottery kiln. Source: Cipriano Piccolpasso, "The Three Books of the Potters's Art," fol. 35C, 1550. Translated by B. Rackham and A. Van de Put, Victoria and Albert Museum, London, 1934.

> two years later applied the same panacea to factory furnaces. Then 1853 and 1856 witnessed two acts of Parliament dealing specifically with London and empowering the police to enforce provisions against smoke from furnaces, public baths, and washhouses and furnaces used in the working of steam vessels on the Thames.

Smoke and ash abatement in Great Britain was considered to be a health agency responsibility and was so confirmed by the first Public Health Act of 1848 and the later ones of 1866 and 1875. Air pollution from the emerging chemical industry was considered a separate matter and was made the responsibility of the Alkali Inspectorate created by the Alkali Act of 1863.

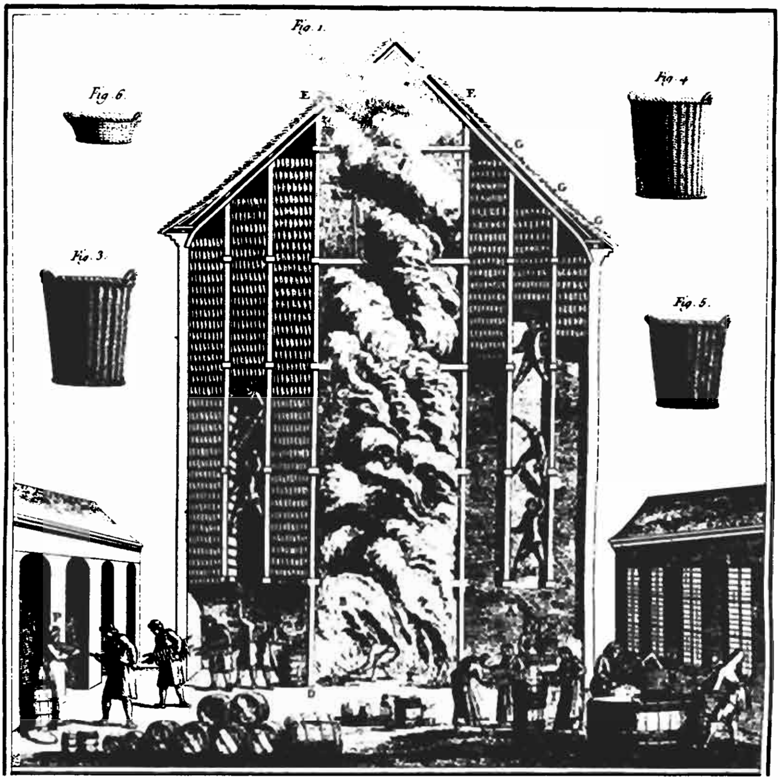

Fig. 1-3. A kiln for smoking red herring. Source: H. L. Duhamel due Monceau, "Traité général des pêches," Vol. 2, Sect. III, Plate XV, Fig. 1, Paris, 1772.

In the United States, smoke abatement (as air pollution control was then known) was considered a municipal responsibility. There were no federal or state smoke abatement laws or regulations. The first municipal ordinances and regulations limiting the emission of black smoke and ash appeared in the 1880s and were directed toward industrial, locomotive, and marine rather than domestic sources. As the nineteenth century drew to a close, the pollution of the air of mill towns the world over had risen to a peak (Fig. 1-4); damage to vegetation from the smelting of sulfide ores was recognized as a problem everywhere it was practiced.

The principal technological developments in the control of air pollution by engineering during the nineteenth century were the stoker for mechanical firing of coal, the scrubber for removing acid gases from effluent gas streams, cyclone and bag house dust collectors, and the introduction of physical and chemical principles into process design.

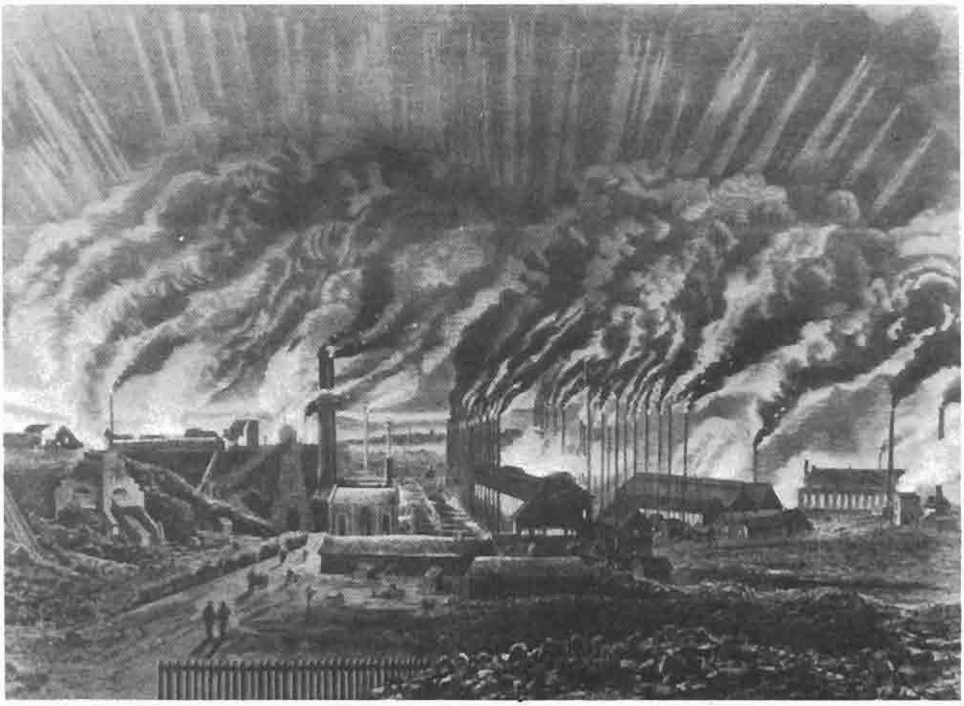

Fig. 1-4. Engraving (1876) of a metal foundry refining department in the industrial Saar region of West Germany. Source: The Bettmann Archive, Inc.

III. THE TWENTIETH CENTURY

A. 1900–1925

During the period 1900–1925 there were great changes in the technology of both the production of air pollution and its engineering control, but no significant changes in legislation, regulations, understanding of the problem, or public attitudes toward the problem. As cities and factories grew in size, the severity of the pollution problem increased.

One of the principal technological changes in the production of pollution was the replacement of the steam engine by the electric motor as the means of operating machinery and pumping water. This change transferred the smoke and ash emission from the boiler house of the factory to the boiler house of the electric generating station. At the start of this period, coal was hand-fired in the boiler house; by the middle of the period, it was mechanically fired by stokers; by the end of the period, pulverized coal, oil, and gas firing had begun to take over. Each form of firing produced its own characteristic emissions to the atmosphere.

At the start of this period, steam locomotives came into the heart of the larger cities. By the end of the period, the urban terminals of many railroads had been electrified, thereby transferring much air pollution from the railroad right-of-way to the electric generating station. The replacement of coal by oil in many applications decreased ash emissions from those sources. There was rapid technological change in industry. However, the most significant change was the rapid increase in the number of automobiles from almost none at the turn of the century to millions by 1925 (Table 1-1).

The principal technological changes in the engineering control of air pollution were the perfection of the motor-driven fan, which allowed large-scale gas-treating systems to be built; the invention of the electrostatic precipitator, which made particulate control in many processes feasible; and the development of a chemical engineering capability for the design of process equipment, which made the control of gas and vapor effluents feasible.

B. 1925–1950

In this period, present-day air pollution problems and solutions emerged. The Meuse Valley (Belgium) episode (4) occurred in 1930; the Donora, Pennsylvania, episode (5) occurred in 1948; and the Poza Rica, Mexico, episode (6) in 1950. Smog first appeared in Los Angeles in the 1940s (Fig. 1-5). The Trail, British Columbia, smelter arbitration (7) was completed in 1941. The first National Air Pollution Symposium in the United States was held in Pasadena, California, in 1949 (8), and the first United States Technical Conference on Air Pollution was held in Washington, D.C., in 1950 (9). The first large-scale surveys of air pollution were undertaken—Salt Lake City, Utah (1926) (10); New York, New York (1937) (11); and Leicester, England (1939) (12).

Air pollution research got a start in California. The Technical Foundation for Air Pollution Meteorology was established in the search for means of disseminating and protecting against chemical, biological, and nuclear warfare agents. Toxicology came of age. The stage was set for the air pollution scientific and technological explosion of the second half of the twentieth century.

A major technological change was the building of natural gas pipelines, and where this occurred, there was rapid displacement of coal and oil as home heating fuels with dramatic improvement in air quality; witness the much publicized decrease in black smoke in Pittsburgh (Fig. 1-6) and St. Louis. The diesel locomotive began to displace the steam locomotive, thereby slowing the pace of railroad electrification. The internal combustion engine bus started its displacement of the electrified streetcar. The automobile continued to proliferate (Table 1-1).

Fig. 1-5. Los Angeles smog. Source: Los Angeles County, California.

During this period, no significant national air pollution legislation or regulations were adopted anywhere in the world. However, the first state air pollution law in the United States was adopted by California in 1947.

C. 1950–1980

In Great Britain, a major air pollution disaster hit London in 1952 (13), resulting in the passage of the Clean Air Act in 1956 and an expansion of the authority of the Alkali Inspectorate. The principal changes that resulted were in the means of heating homes. Previously, most heating was done by burning soft coal on grates in separate fireplaces in each room. A successful effort was made to substitute smokeless fuels for the soft coal used in this manner, and central or electrical heating for fireplace heating. The outcome was a decrease in "smoke" concentration, as measured by the blackness of paper filters through which British air was passed from 175 $\mu g/m^3$ in 1958 to 75 $\mu g/m^3$ in 1968 (14).

During these two decades, almost every country in Europe, as well as Japan, Australia, and New Zealand, experienced serious air pollution in its larger cities. As a result, these countries were the first to enact national air pollution control legislation. By 1980, major national air pollution research centers had been set up at the Warren Springs Laboratory, Stevenage, England; the Institut National de la Santé et de las Recherche Medicale at

Le Visinet, France; the Rijksinstituut Voor de Volksgezondheid, Bilthoven and the Instituut voor Gezondheidstechniek-TNO, Delft, The Netherlands; the Statens Naturvardsverk, Solna, Sweden; the Institut für Wasser-Boden-und Luft-hygiene, Berlin and the Landensanstalt für Immissions und Bodennutzungsshutz, Essen, Germany. The important air pollution research centers in Japan are too numerous to mention.

In the United States, the smog problem continued to worsen in Los Angeles and appeared in large cities throughout the nation (Fig. 1-7). In 1955 the first federal air pollution legislation was enacted, providing federal support for air pollution research, training, and technical assistance. Responsibility for the administration of the federal program was given to the Public Health Service (PHS) of the United States Department of Health, Education, and Welfare, and remained there until 1970, when it was transferred to the new United States Environmental Protection Agency (EPA). The initial federal legislation was amended and extended several times between 1955 and 1980, greatly increasing federal authority, particularly in the area of control (15). The automobile continued to proliferate (Table 1-1).

As in Europe, air pollution research activity expanded tremendously in the United States during these three decades. The headquarters of federal research activity was at the Robert A. Taft Sanitary Engineering Center of the PHS in Cincinnati, Ohio, during the early years of the period and at the National Environmental Research Center in Triangle Park, North Carolina, at the end of the period.

An International Air Pollution Congress was held in New York City in 1955 (16). Three National Air Pollution Conferences were held in Washington, D.C., in 1958 (17), 1962 (18), and 1966 (19). In 1959, an International Clean Air Conference was held in London (20).

TABLE 1-1

Annual Motor Vehicle Sales in the United States[a]

Year	Total	Year	Total
1900	4,192	1945	725,215
1905	25,000	1950	8,003,056
1910	187,000	1955	9,169,292
1915	969,930	1960	7,869,221
1920	2,227,347	1965	11,057,366
1925	4,265,830	1970	8,239,257
1930	3,362,820	1975	8,985,012
1935	3,971,241	1980	8,067,309
1940	4,472,286	1985	11,045,784
		1990	9,295,732

[a] Data include foreign and domestic sales for trucks, buses, and automobiles.

Fig. 1-6. (Right) Pittsburgh before the decrease in black smoke. Source: Allegheny County, Pennsylvania. (Left) Pittsburgh after the decrease in black smoke. Source: Allegheny County, Pennsylvania

In 1964, the International Union of Air Pollution Prevention Associations (IUAPPA) was formed. IUAPPA has held International Clean Air Congresses in London in 1966 (21); Washington, D.C., in 1970 (22); Dusseldorf in 1973 (23); Tokyo in 1977 (24); Buenos Aires in 1980 (25); Paris in 1983 (26); Sydney in 1986 (27); The Hague in 1989 (28); and Montreal in 1992 (29).

Technological interest during these 30 years has focused on automotive air pollution and its control, on sulfur oxide pollution and its control by sulfur oxide removal from flue gases and fuel desulfurization, and on control of nitrogen oxides produced in combustion processes.

Air pollution meteorology came of age and, by 1980, mathematical models of the pollution of the atmosphere were being energetically developed. A start had been made in elucidating the photochemistry of air pollution. Air quality monitoring systems became operational throughout the world. A wide variety of measuring instruments became available.

Fig. 1-7. Smog in New York City. Source: Wide World Photos.

IV. THE 1980s

The highlight of the 1970s and 1980s was the emergence of the ecological, or total environmental, approach. Organizationally, this has taken the form of departments or ministries of the environment in governments at all levels throughout the world. In the United States there is a federal Environmental Protection Agency, and in most states and populous counties and cities, there are counterpart organizations charged with responsibility for air and water quality, sold waste sanitation, noise abatement, and control of the hazards associated with radiation and the use of pesticides. This is paralleled in industry, where formerly diffuse responsibility for these areas is increasingly the responsibility of an environmental protection coordinator. Similar changes are evident in research and education.

Pollution controls were being built into pollution sources—automobiles, power plants, factories—at the time of original construction rather than later on. Also, for the first time, serious attention was directed to the problems caused by the "greenhouse" effect of carbon dioxide and other gases building up in the atmosphere, possible depletion of the stratospheric ozone layer by fluorocarbons, long-range transport of pollution, prevention of significant deterioration (PSD), and acidic deposition.

V. THE 1990s

The most sweeping change, in the United States at least, in the decade of the 1990s was the passage of the Clean Air Act Amendments on November 15, 1990 (29). This was the only change in the Clean Air Act since 1977, even though the U.S. Congress had mandated that the Act be amended much earlier. Michigan Representative John Dingell referred to the amendments as "the most complex, comprehensive, and far-reaching environmental law any Congress has ever considered." John-Mark Stenvaag has stated in his book, "Clean Air Act 1990 Amendments, Law and Practice" (30), "The enormity of the 1990 amendments begs description. The prior Act, consisting of approximately 70,000 words, was widely recognized to be a remarkably complicated, unapproachable piece of legislation. If environmental attorneys, government officials, and regulated entities were awed by the prior Act, they will be astonished, even stupefied, by the 1990 amendments. In approximately 145,000 new words, Congress has essentially tripled the length of the prior Act and geometrically increased its complexity."

The 1990s saw the emergence, in the popular media, of two distinct but closely related global environmental crises, uncontrolled global climate changes and stratospheric ozone depletion. The climate changes of concern were both the warming trends caused by the buildup of greenhouse gases in the atmosphere and cooling trends caused by particulate matter and sulfates in the same atmosphere. Some researchers have suggested that these two trends will cancel each other. Other authors have written (31) that global warming may not be all bad. It is going to be an interesting decade as many theories are developed and tested during the 1990s. The "Earth Summit," really the U.N. Conference of Environment and Development, in Rio de Janeiro during June 1992 did little to resolve the problems, but it did indicate the magnitude of the concern and the differences expressed by the nations of the world.

The other global environmental problem, stratospheric ozone depletion, was less controversial and more imminent. The U.S. Senate Committee Report supporting the Clean Air Act Amendments of 1990 states, "Destruction of the ozone layer is caused primarily by the release into the atmosphere of chlorofluorocarbons (CFCs) and similar manufactured substances—persistent chemicals that rise into the stratosphere where they catalyze the destruction of stratospheric ozone. A decrease in stratospheric ozone will allow more ultraviolet (UV) radiation to reach Earth, resulting in increased rates of disease in humans, including increased incidence of skin cancer, cataracts, and, potentially, suppression of the immune system. Increased UV radiation has also been shown to damage crops and marine resources."

The Montreal Protocol of July 1987 resulted in an international treaty in which the industrialized nations agreed to halt the production of most ozone-destroying chlorofluorocarbons by the year 2000. This deadline was hastily changed to 1996, in February 1992, after a U.S. National Aeronautics and Space Administration (NASA) satellite and high-altitude sampling aircraft found levels of chlorine monoxide over North America that were 50% greater than that measured over Antarctica.

VI. THE FUTURE

The air pollution problems of the future are predicated on the use of more and more fossil and nuclear fuel as the population of the world increases. During the lifetime of the students using this book, partial respite may be offered by solar, photovoltaic, geothermal, wind, nonfossile fuel (hydrogen and biomass), and oceanic (thermal gradient, tidal, and wave) sources of energy. Still, many of the agonizing environmental decisions of the next decades will involve a choice between fossil fuel and nuclear power sources and the depletion of future fuel reserves for present needs. Serious questions will arise regarding whether to conserve or to use these reserves—whether to allow unlimited growth or to curb it.

Other problems concerning transportation systems, waste processing and recycling systems, national priorities, international economics, employment versus environmental quality, and personal freedoms will continue to surface. The choices will have to be made, ideally by educated citizens and charismatic leaders.

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QUESTIONS

1. Discuss the development of the use of enclosed space for human occupancy over the period of recorded history.
2. Discuss the development of the heating and cooling of enclosed space for human occupancy over the period of recorded history.
3. Discuss the development of the lighting of enclosed space for human occupancy over the period of recorded history.
4. Discuss the development of means to supplement human muscular power over the period of recorded history.
5. Discuss the development of transportation over the period of recorded history.
6. Discuss the development of agriculture over the period of recorded history.
7. Discuss the future alternative sources of energy for light, heat, and power.
8. Compare the so-called soft (i.e., widely distributed small sources) and hard (i.e., fewer very large sources) paths for the future provision of energy for light, heat, and power.
9. What have been the most important developments in the history of the air pollution problem since the publication of this edition of this book?

2

The Natural versus Polluted Atmosphere

Pollution, *n:* the action of polluting; the condition of being polluted. Pollute, *vt:* [L. *pollutus,* past part. or *polluere,* to make physically impure or unclean]; to defile, desecrate, profane-syn. see CONTAMINATE. Air, *n:* [fr. L *aer,* fr. Gr. *aer*] 1. the mixture of invisible tasteless gases which surrounds the earth . Atmosphere, *n:* [fr. Gr. (*atmo* and *sphaira*) 2. the whole mass of air surrounding the earth . . .[1]

I. THE ATMOSPHERE

On a macroscale (Fig. 2-1) as temperature varies with altitude, so does density (1). In general, the air grows progressively less dense as we move upward from the troposphere through the stratosphere and the chemosphere to the ionosphere. In the upper reaches of the ionosphere, the gaseous molecules are few and far between as compared with the troposphere.

The ionosphere and chemosphere are of interest to space scientists because they must be traversed by space vehicles en route to or from the

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[1] Definitions are from "Webster's Tenth New Collegiate Dictionary," 10th ed. Merriam, Springfield, MA, 1993.

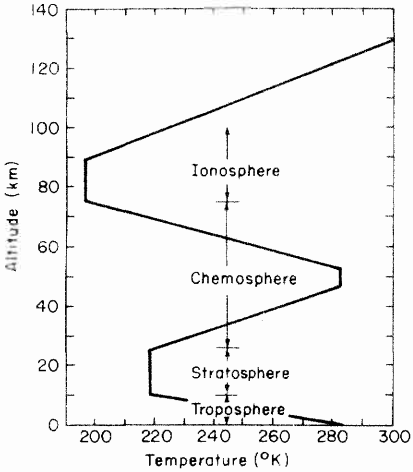


Fig. 2-1. The regions of the atmosphere.

moon or the planets, and they are regions in which satellites travel in the earth's orbit. These regions are of interest to communications scientists because of their influence on radio communications, and they are of interest to air pollution scientists primarily because of their absorption and scattering of solar energy, which influence the amount and spectral distribution of solar energy and cosmic rays reaching the stratosphere and troposphere.

The stratosphere is of interest to aeronautical scientists because it is traversed by airplanes; to communications scientists because of radio and television communications; and to air pollution scientists because global transport of pollution, particularly the debris of aboveground atomic bomb tests and volcanic eruptions, occurs in this region and because absorption and scattering of solar energy also occur there. The lower portion of this region contains the stratospheric ozone layer, which absorbs harmful ultraviolet (UV) solar radiation. Global change scientists are interested in modifications of this layer by long-term accumulation of chlorofluorocarbons (CFCs) and other gases released at the earth's surface or by high-altitude aircraft.

The troposphere is the region in which we live and is the primary focus of this book.

II. UNPOLLUTED AIR

The gaseous composition of unpolluted tropospheric air is given in Table 2-1. Unpolluted air is a concept, i.e., what the composition of the air would be if humans and their works were not on earth. We will never know the precise composition of unpolluted air because by the time we had the means and the desire to determine its composition, humans had been polluting the air for thousands of years. Now even at the most remote locations at sea, at the poles, and in the deserts and mountains, the air may be best described as dilute polluted air. It closely approximates unpolluted air, but differs from it to the extent that it contains vestiges of diffused and aged human-made pollution.

The real atmosphere is more than a dry mixture of permanent gases. It has other constituents—vapor of both water and organic liquids, and particulate matter held in suspension. Above their temperature of condensation, vapor molecules act just like permanent gas molecules in the air. The predominant vapor in the air is water vapor. Below its condensation temperature, if the air is saturated, water changes from vapor to liquid. We are all familiar with this phenomenon because it appears as fog or mist in the air and as condensed liquid water on windows and other cold surfaces exposed to air. The quantity of water vapor in the air varies greatly from almost complete dryness to supersaturation, i.e., between 0% and 4% by weight. If Table 2-1 is compiled on a wet air basis at a time when the water vapor concentration is 31,200 parts by volume per million parts by volume of wet air (Table 2-2), the concentration of condensable organic vapors is seen to be so low compared to that of water vapor that for all practical purposes the difference between wet air and dry air is its water vapor content.

Gaseous composition in Tables 2-1 and 2-2 is expressed as parts per million by volume—ppm (vol). (When a concentration is expressed simply

TABLE 2-1

The Gaseous Composition of Unpolluted Air (Dry Basis)

	ppm (vol)	μg/m^3
Nitrogen	780,000	8.95×10^8
Oxygen	209,400	2.74×10^8
Water	—	—
Argon	9,300	1.52×10^7
Carbon dioxide	315	5.67×10^5
Neon	18	1.49×10^4
Helium	5.2	8.50×10^2
Methane	1.0–1.2	$6.56–7.87 \times 10^2$
Krypton	1.0	3.43×10^3
Nitrous oxide	0.5	9.00×10^2
Hydrogen	0.5	4.13×10^1
Xenon	0.08	4.29×10^2
Organic vapors	ca. 0.02	—

TABLE 2-2

The Gaseous Composition of Unpolluted Air (Wet Basis)

	ppm (vol)	$\mu g/m^3$
Nitrogen	756,500	8.67×10^8
Oxygen	202,900	2.65×10^8
Water	31,200	2.30×10^7
Argon	9,000	1.47×10^7
Carbon dioxide	305	5.49×10^5
Neon	17.4	1.44×10^4
Helium	5.0	8.25×10^2
Methane	0.97–1.16	$6.35\text{–}7.63 \times 10^2$
Krypton	0.97	3.32×10^3
Nitrous oxide	0.49	8.73×10^2
Hydrogen	0.49	4.00×10^1
Xenon	0.08	4.17×10^2
Organic vapors	ca. 0.02	—

as ppm, one is in doubt as to whether a volume or weight basis is intended.) To avoid confusion caused by different units, air pollutant concentrations in this book are generally expressed as micrograms per cubic meter of air ($\mu g/m^3$) at 25°C and 760 mm Hg, i.e., in metric units. To convert from units of ppm (vol) to $\mu g/m^3$, it is assumed that the ideal gas law is accurate under ambient conditions. A generalized formula for the conversion at 25°C and 760 mm Hg is

$$\begin{aligned} 1 \text{ ppm (vol) pollutant} &= \frac{1 \text{ liter pollutant}}{10^6 \text{ liter air}} \\ &= \frac{(1 \text{ liter}/22.4) \times \text{MW} \times 10^6\ \mu\text{g/gm}}{10^6 \text{ liters} \times 298°\text{K}/273°\text{K} \times 10^{-3}\ \text{m}^3/\text{liter}} \\ &= 40.9 \times \text{MW}\ \mu\text{g/m}^3 \qquad (2\text{-}1) \end{aligned}$$

where MW equals molecular weight. For convenience, conversion units for common pollutants are shown in Table 2-3.

A minor problem arises in regard to nitrogen oxides. It is common practice to add concentrations of nitrogen dioxide and nitric oxide in ppm (vol) and express the sum as "oxides of nitrogen." In metric units, conversion from ppm (vol) to $\mu g/m^3$ must be done separately for nitrogen dioxide and nitric oxide prior to addition.

III. PARTICULATE MATTER

Neither Table 2-1 nor Table 2-2 lists among the constituents of the air the suspended particulate matter that it always contains. The gases and vapors exist as individual molecules in random motion. Each gas or vapor

TABLE 2-3

Conversion Factors between Volume and Mass Units of Concentration (25°C, 760 mm Hg)

	To convert from	
Pollutant	ppm (vol) to $\mu g/m^3$, multiply by:	$\mu g/m^3$ to ppm (vol), multiply by ($\times 10^{-3}$):
Ammonia (NH_3)	695	1.44
Carbon dioxide	1800	0.56
Carbon monoxide	1150	0.87
Chlorine	2900	0.34
Ethylene	1150	0.87
Hydrogen chloride	1490	0.67
Hydrogen fluoride	820	1.22
Hydrogen sulfide	1390	0.72
Methane (carbon)	655	1.53
Nitrogen dioxide	1880	0.53
Nitric oxide	1230	0.81
Ozone	1960	0.51
Peroxyacetylnitrate	4950	0.20
Sulfur dioxide	2620	0.38

exerts its proportionate partial pressure. The particles are aggregates of many molecules, sometimes of similar molecules, often of dissimilar ones. They age in the air by several processes. Some particles serve as nuclei upon which vapors condense. Some particles react chemically with atmospheric gases or vapors to form different compounds. When two particles collide in the air, they tend to adhere to each other because of attractive surface forces, thereby forming progressively larger and larger particles by agglomeration. The larger a particle becomes, the greater its weight and the greater its likelihood of falling to the ground rather than remaining airborne. The process by which particles fall out of the air to the ground is called *sedimentation*. Washout of particles by snowflakes, rain, hail, sleet, mist, or fog is a common form of agglomeration and sedimentation. Still other particles leave the air by impaction onto and retention by the solid surfaces of vegetation, soil, and buildings. The particulate mix in the atmosphere is dynamic, with continual injection into the air from sources of small particles; creation of particles in the air by vapor condensation or chemical reaction among gases and vapors; and removal of particles from the air by agglomeration, sedimentation, or impaction.

Before the advent of humans and their works, there must have been particles in the air from natural sources. These certainly included all the particulate forms of condensed water vapor; the condensed and reacted forms of natural organic vapors; salt particles resulting from the evaporation of water from sea spray; wind-borne pollen, fungi, molds, algae, yeasts, rusts, bacteria, and debris from live and decaying plant and animal life; particles eroded by the wind from beaches, desert, soil, and rock; particles from volcanic and other geothermal eruption and from forest fires started by lightning; and particles entering the troposphere from outer space. As mentioned earlier, the true natural background concentration will never be known because when it existed humans were not there to measure it, and by the time humans started measuring particulate matter levels in the air, they had already been polluting the atmosphere with particles resulting from their presence on earth for several million years. The best that can be done now is to assume that the particulate levels at remote places—the middle of the sea, the poles, and the mountaintops—approach the true background concentration. The very act of going to a remote location to make a measurement implies some change in the atmosphere of that remote location attributable to the means people used to travel and to maintain themselves while obtaining the measurements. Particulate matter is measured on a dry basis, thereby eliminating from the measurement not only water droplets and snowflakes but also all vapors, both aqueous and organic, that evaporate or are desiccated from the particulate matter during the drying process. Since different investigators and investigative processes employ different drying procedures and definitions of dryness, it is important to know the procedures and definition employed when comparing data.

There are ways of measuring particulate matter other than by weight per unit volume of air. They include a count of the total number of particles in a unit volume of air, a count of the number of particles of each size range, the weight of particles of each size range, and similar measures based on the surface area and volume of the particles rather than on their number or weight. Some particles in the air are so small that they cannot be seen by an optical microscope, individually weighing so little that their presence is masked in gravimetric analysis by the presence of a few large particles. The mass of a spherical particle is

$$w = \tfrac{4}{3}\pi p r^3 \tag{2-2}$$

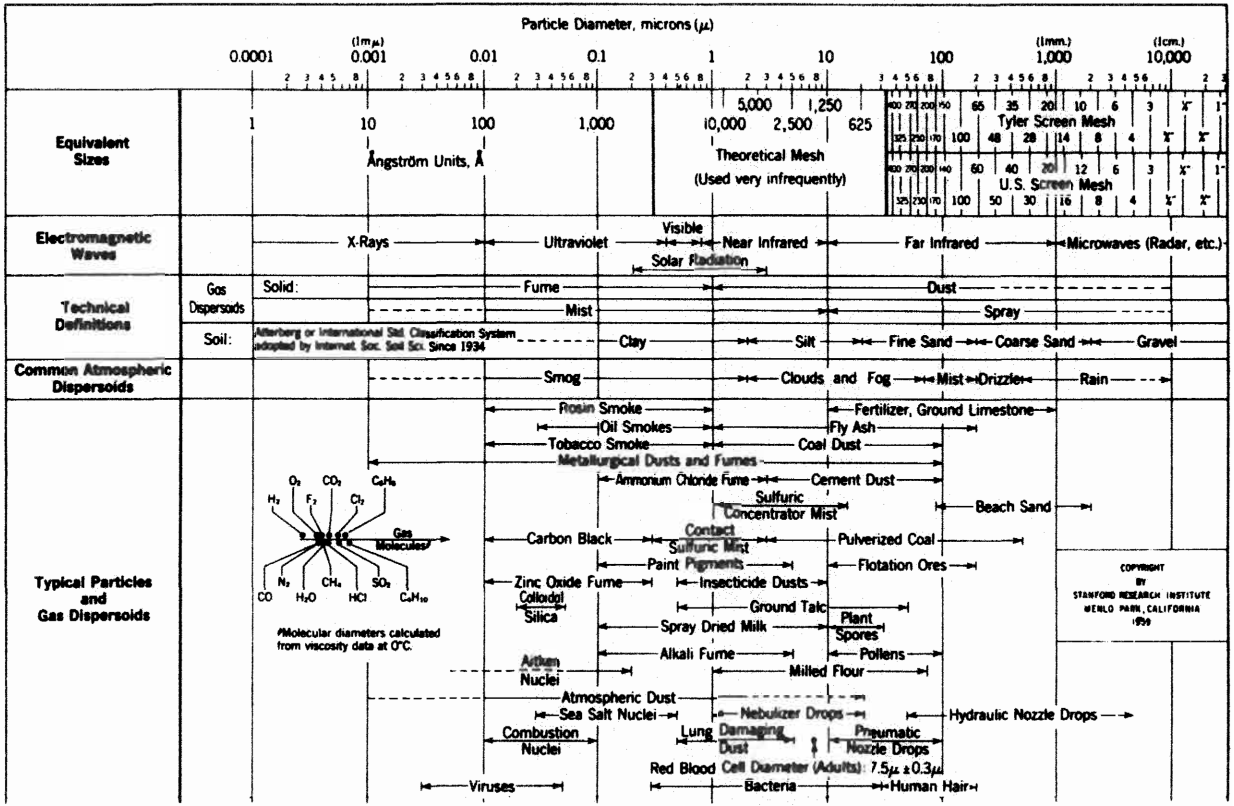
where w is the particle mass (gm), r is the particle radius (cm), and p is the particle density (gm/cm^3).

The size of small particles is measured in microns (μm). One micron is one-millionth of a meter or 10,000 Å (angstrom units)—the units used to measure the wavelength of light (visible light is between 3000 and 8000 Å) (Fig. 2-2) (2). Compare the weight of a 10-μm particle near the upper limit of those found suspended in the air and a 0.1-μm particle which is near the lower limit. If both particles have the same density (p), the smaller particle will have one-millionth the weight of the larger one. The usual gravimetric procedures can scarcely distinguish a 0.1-μm particle in the presence of a 10-μm particle. To measure the entire size range of particles in the atmosphere, several measurement techniques must therefore be combined, each most appropriate for its size range (Table 2-4). Thus the smallest particles—those only slightly larger than a gas molecule—are measured by the electric charge they carry and by electron microscopy. The next larger size range is measured by electron microscopy or by the ability of these particles to act as nuclei upon which water vapor can be condensed in a cloud chamber. (The water droplets are measured rather than the particles themselves.) The still larger size range is measured by electron or optical microscopy; and the largest size range is measured gravimetrically, either as suspended particles separated from the air by a sampling device or as sedimented particles falling out of the air into a receptacle.

By measuring each portion of the particle size spectrum by the most appropriate method, a composite diagram of the size distribution of the atmospheric aerosol can be produced. Figure 2-3 shows that there are separate size distributions with respect to the number, surface area, and volume (or mass) of the particles. The volume (mass) distribution is called *bimodal* because of its separate maxima at about 0.2 and 10 μm, which result from different mechanisms of particle formation. The mode with the 0.2-μm maximum results from coagulation and condensation formation mechanisms. These particles are created in the atmosphere by chemical reaction among gases and vapors. They are called *fine* particles to differentiate them from the particles in the 10-μm maximum mode, which are called *coarse*. These fine particles are primarily sulfates, nitrates, organics, ammonium, and lead compounds. The mode with the 10-μm maximum are particles introduced to the atmosphere as solids from the surface of the earth and the seas, plus particles from the coagulation–condensation mode which have grown larger and moved across the saddle between the modes into the larger size mode. These are primarily silicon, iron, aluminum, sea salt, and plant particles. Thus there is a dynamism that creates small particles, allows them to grow larger, and eventually allows the large particles to be scavenged from the atmosphere by sedimentation (in the absence of precipitation), plus washout and rainout when there is precipitation.

The majority of particles in the atmosphere are spherical in shape because they are formed by condensation or cooling processes or they contain core nuclei coated with liquid. Liquid surface tension draws the material in the particle into a spherical shape. Other important particle shapes exist in the atmosphere; e.g., asbestos is present as long fibers and fly ash can be irregular in shape.

The methods just noted tell something about the physical characteristics of atmospheric particulate matter but nothing about its chemical composition. One can seek this kind of information for either individual particles or all particles en masse. Analysis of particles en masse involves analysis of a mixture of particles of many different compounds. How much of


Particle Diameter, microns (μ)
0.0001
(1mμ) 0.001
0.01
0.1
1
10
100
(1mm.) 1,000
(1cm.) 10,000
Equivalent Sizes
1
10
100
1,000
Ångström Units, Å
10,000
5,000
2,500
1,250
625
Theoretical Mesh
(Used very infrequently)
Tyler Screen Mesh
U.S. Screen Mesh
Electromagnetic Waves
X-Rays
Ultraviolet
Visible
Near Infrared
Far Infrared
Microwaves (Radar, etc.)
Solar Radiation
Technical Definitions
Gas Dispersoids
Solid:
Fume
Dust
Mist
Spray
Soil:
Atterberg or International Std. Classification System adopted by Internat. Soc. Soil Sci. Since 1934
Clay
Silt
Fine Sand
Coarse Sand
Gravel
Common Atmospheric Dispersoids
Smog
Clouds and Fog
Mist
Drizzle
Rain
Typical Particles and Gas Dispersoids
Rosin Smoke
Fertilizer, Ground Limestone
Oil Smokes
Fly Ash
Tobacco Smoke
Coal Dust
Metallurgical Dusts and Fumes
Ammonium Chloride Fume
Cement Dust
Sulfuric Concentrator Mist
Beach Sand
Carbon Black
Contact Sulfuric Mist
Pulverized Coal
Paint Pigments
Flotation Ores
Zinc Oxide Fume
Insecticide Dusts
Colloidal Silica
Ground Talc
Spray Dried Milk
Plant Spores
Alkali Fume
Pollens
Aitken Nuclei
Milled Flour
Atmospheric Dust
Sea Salt Nuclei
Nebulizer Drops
Hydraulic Nozzle Drops
Combustion Nuclei
Lung Damaging Dust
Pneumatic Nozzle Drops
Red Blood Cell Diameter (Adults): 7.5μ ± 0.3μ
Viruses
Bacteria
Human Hair
H_2
O_2
CO_2
C_6H_6
F_2
Cl_2
Gas Molecules*
CO
N_2
H_2O
CH_4
HCl
SO_2
C_4H_{10}
*Molecular diameters calculated from viscosity data at 0°C.
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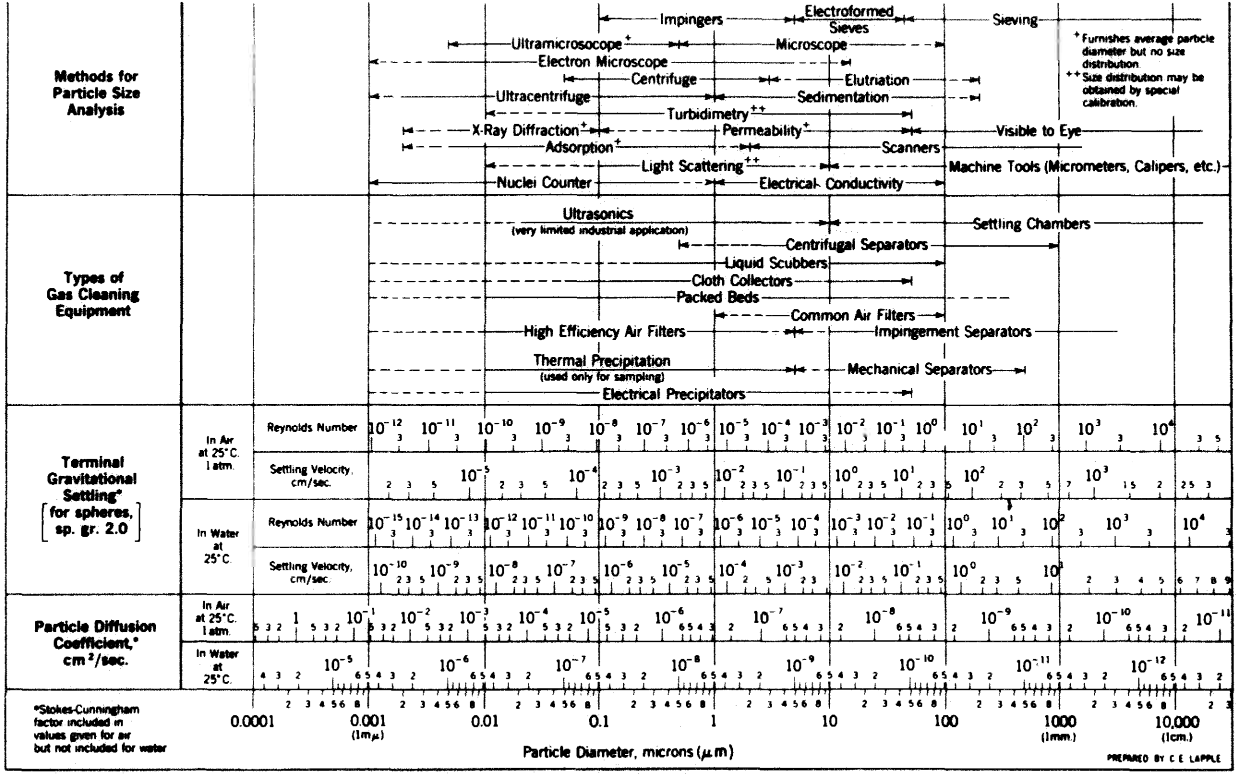

Fig. 2-2. Characteristics of particles and particle dispersoids. Reproduced by permission of SRI International, Menlo Park, California, 1959.

TABLE 2-4

Particle Size Ranges and Their Methods of Measurement

Particle size range (um)	Ions	Nuclei	Visability	Suspended or settleable; nonairborne	Dispersion aerosol	Condensation aerosol	Pollen and spores	Sedimentation, diffusion, and settling
10^{-4}–10^{-3}	Small	—	—	Suspended	—	Gas molecules	—	Diffusion
10^{-3}–10^{-2}	Intermediate and large	Aitken nuclei	Electron microscope	Suspended	—	Vapor molecules	—	Diffusion
10^{-2}–10^{-1}	Large	Aitken and condensation nuclei	Electron microscope	Suspended	—	Fume–mist	—	Diffusion
Air pollution								
10^{-1}–10^{-0}	—	Condensation nuclei	Microscope: electron and optical	Suspended	Dust–mist	Fume–mist	—	Diffusion and sedimentation
10^{0}–10^{1}	—	—	Microscope: optical	Suspended and settleable	Dust–mist	Fume–mist	—	Sedimentation
10^{1}–10^{2}	—	—	Eye, sieves	Settleable	Dust–mist	Mist–fog	Pollen and spores	
10^{2}–10^{3}	—	—	Eye, sieves	Nonairborne	Dust–spray	Drizzle–rain	—	Sedimentation
10^{3}–10^{4}	—	—	Eye, sieves	Nonairborne	Sand–rocks	Rain	—	Sedimentation

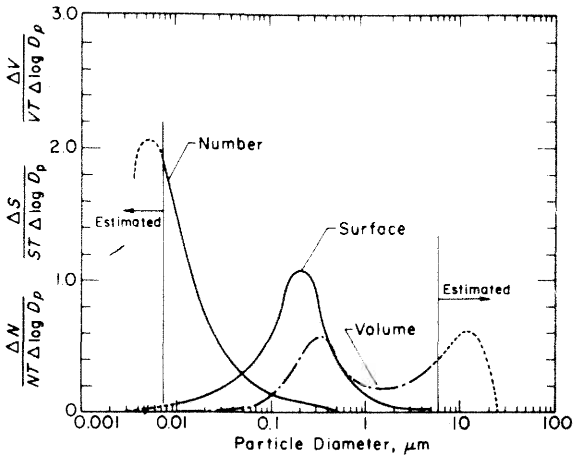

Fig. 2-3. Grand average number (N), surface area (S), and volume (V) distribution of Los Angeles smog. The linear ordinate normalized by total number (NT), area (ST), or volume (VT) is used so that the apparent area under the curves is proportional to the quantity in that size range. Source: Corn, M., Properties of non-viable particles in the air, *In* "Air Pollution," 3rd ed., Vol. I (A. C. Stern, ed.). Academic Press, New York, 1976, p. 123.

each element or radical, anion, or cation is present in the mixture can be determined. Specific organic compounds may be separated and identified. Individual particles may be analyzed by electron microscopy techniques.

Much of the concern about particulate matter in the atmosphere arises because particles of certain size ranges can be inhaled and retained by the human respiratory system. There is also concern because particulate matter in the atmosphere absorbs and scatters incoming solar radiation. For a detailed discussion of the human respiratory system and the defenses it provides against exposure of the lungs to particulate matter, see Chapter 7.

IV. CONCEPTS

A. Sources and Sinks

The places from which pollutants emanate are called *sources*. There are natural as well as anthropogenic sources of the permanent gases considered to be pollutants. These include plant and animal respiration and the decay of what was once living matter. Volcanoes and naturally caused forest fires are other natural sources. The places to which pollutants disappear from the air are called *sinks*. Sinks include the soil, vegetation, structures, and water bodies, particularly the oceans. The mechanisms whereby pollutants