

## 2. Remote Sensing for Air Quality Management

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With 3 Figures

In recent years, it has become clear that man's activities are producing significant and measurable effects on the environment. Two isolated events dramatically brought this to the attention of the world: The air pollution incidents in Donora, Pennsylvania, during 1948 (20 dead and several hundred ill), and London during December 1952 (4000 dead in a few days). It became clear that the environment is not a bottomless sink, but is fragile and could not continue to be abused. As a result of these tragedies and other significant environmental insults, public opinion demanded that the environment be considered a valued resource and as such, be protected. The U.S. Congress passed a number of laws which dictate that environmental pollution be reduced to socially accepted levels.

To accomplish this mandate for atmospheric pollutants requires a detailed assessment of a number of interrelated factors. First, an evaluation of the effects of specific pollutants is performed which points out the critical receptor of that pollutant and the maximum allowable pollutant level to minimize effects on health or welfare. Next, pollutant sources and their respective strengths are identified. This information is incorporated in pollutant transport and dispersion models to predict ambient concentrations. High ambient concentrations may require controls on the sources, and the efficacy of these controls must be measured. These processes are iterated until ambient levels are below the established maximum level. Most of these factors require pollutant concentration monitoring.

Historically, air pollution monitoring has been performed using wet chemical techniques and grab sampling for later laboratory analysis. During the last decade, physical and automated methods have been developed which satisfy many monitoring requirements. These instruments are generally precise and accurate, but are limited to single-point measurements. Remote monitoring instrumentation, especially active techniques incorporating laser sources, holds promise for providing three-dimensional pollution concentration measurements. These systems have the potential for being used in:

a) Monitoring sources—Rapid, non-interfering measurement of pollutant effluents.

b) Measuring transport of pollutants—Monitoring at ground level and aloft over wide geographical areas.

c) Monitoring ambient pollutant concentrations—Spatial resolution similar to model predictions and other applications which are discussed in Section 2.4.

The following sections of this chapter will provide a review of some basic information concerning laser probing of the atmosphere.

A general discussion of the atmosphere is presented in Section 2.1 providing a review of atmospheric constituents, their significance, and their concentration ranges. This is followed in Section 2.2 with a brief overview of mathematical modeling for predicting pollutant transport and diffusion. The next Section 2.3 provides a general overview of laser probing techniques and systems to monitor the atmosphere; and finally, the last Section 2.4 describes advantages of remote monitoring and points out specific applications for laser sensing systems.

## 2.1 The Atmosphere

This section will briefly describe the atmosphere in relationship to laser probing. A brief description of the medium (the air) in which the measurements are made is in order, along with a description of the atmospheric constituents which are candidates for laser monitoring.

The atmosphere consists primarily of nitrogen and oxygen. These molecules make up 99% of the dry air, with argon comprising the majority of the remainder (nearly 1%). Water vapor, which is highly variable, is the next major gaseous constituent, followed by carbon dioxide. Many other gases are present in trace concentrations and will be discussed later in this section. In addition to these, the atmosphere consists of solid particles, aerosols and liquid and/or solid water.

Most of the constituents modify the transmission of electromagnetic radiation through the atmosphere. It is just this modification (attenuation, absorption, and scattering) which is the measurable parameter in the application of laser probing; and it is, therefore, important to have a clear understanding of the air to appreciate such applications. The discussion will treat separately the two major regions of the lower atmosphere—the troposphere and stratosphere.

### 2.1.1 Structure of the Atmosphere

The atmosphere is generally divided into a number of layers, each of which is characterized by its temperature structure. The mean temperature profile of the atmosphere, shown in Fig. 2.1, demonstrates this

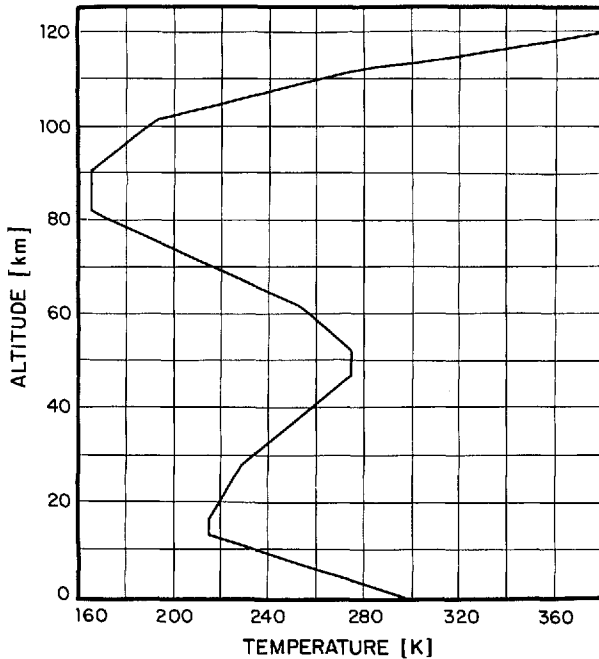


Fig. 2.1. Mean temperature as a function of altitude at 45° North latitude during July. Adapted from [2.1]

division. In the lowest layer, the temperature decreases from the Earth's surface to an altitude of approximately 10–12 km. Above this altitude, temperature increases with increasing altitude. The altitude at which this change occurs is referred to as the *tropopause* and defines the boundary between the *troposphere* (the lowest layer) and the *stratosphere* above. Within the troposphere, the air is generally near neutral stability (controlled by the decreasing temperature with altitude), and turbulent mixing is good; whereas, in the stratosphere, because of the increasing temperature with altitude, the air is very stable and mixing is discouraged. Because of the difference in stability and, thereby, difference in mixing, and since washout due to rainfall generally occurs in the troposphere, the residence time for minor constituents in the stratosphere is significantly longer. Residence time in the stratosphere may range from a few weeks to years, whereas in the troposphere, residence time may be as low as a few hours to days. The other significant difference is the high level of ultra-violet radiation available to promote photochemical reactions in the upper portions of the stratosphere.

Table 2.1. Composition of "clean" dry air near sea level. Adapted from [2.2] and [2.3]

Component [% by volume]		Content [ppm]	Component [% by volume]		Content [ppm]
Nitrogen	78.09	780900	Hydrogen	0.00005	0.5
Oxygen	20.94	209400	Methane	0.00015	1.5
Argon	0.93	9300	Nitrogen	0.0000001	0.001
Carbon	0.0318	318	dioxide		
dioxide			Ozone	0.000002	0.02
Neon	0.0018	18	Sulfur	0.00000002	0.0002
Helium	0.00052	5.2	dioxide		
Krypton	0.0001	1	Carbon	0.00001	0.1
Xenon	0.000008	0.08	mon-		
Nitrous	0.000025	0.25	oxide		
oxide			Ammonia	0.000001	0.01

*Note:* The concentrations of some of these gases may differ with time and place, and the data for some are open to question. Single values for concentrations, instead of ranges of concentrations, are given above to indicate order of magnitude, not specific and universally accepted concentrations.

### 2.1.2 Troposphere

The normal dry-air constituents of the troposphere and their respective concentrations are listed in Table 2.1. As was mentioned earlier, nitrogen and oxygen comprise 99% of the air in the lower atmosphere, followed in order by argon (1%) and carbon dioxide (0.03%). Within the troposphere, water vapor is highly variable, ranging in concentration from 1 to 3%.

Some of the minor constituents listed in Table 2.1 as well as other pollutants, vary both temporally and spatially in the troposphere. The significance of these constituents to air quality management, along with their sources and concentration ranges, will be discussed within one of the following categories:

- 1) Aerosols and particles
- 2) Oxides of carbon
- 3) Sulfur compounds
- 4) Nitrogen compounds
- 5) Hydrocarbons
- 6) Ozone.

#### *Aerosols and Particles*

The most noticeable effect aerosols and particles have on the troposphere is their contribution to the reduction in visibility. In addition, they provide reaction sites for pollutant gases, modify precipitation by

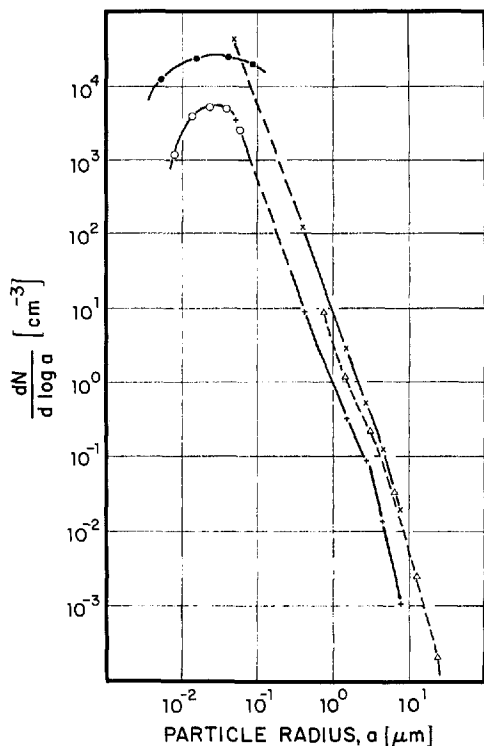


Fig. 2.2. Complete size distributions of aerosol particles at various stations in Germany. Data for the lower curve were obtained from ion mobility measurements, whereas those for the upper curve were based on impactor measurements. Individual sets of marked data points represent averages of many measurements which were not necessarily simultaneous.  $N$  is the total concentration of aerosol particles of radius smaller than  $a$ . After JUNGE [2.4]

functioning as condensation nuclei, and act as carries to transport pollutants into the lungs, eyes, and other sensitive organs of man and onto the leaves of plants and crops. The natural sources of aerosols and particles include sea spray, wind-blown soil, pollen, forest fires, volcanoes, and the reaction products of naturally occurring chemicals (e.g. the Great Smokey Mountains). Man contributes to this natural burden through the combustion of fossil fuels, the reactions of man-made pollutants, and other activities such as construction and agriculture.

Particulate matter in the troposphere varies in composition from solid particles such as fly ash, lead, sulfates, and nitrates, to liquid aerosols, including sulfuric and nitric acid and other dissolved salts. In between these two extremes are solid particles with liquid cladding.

Table 2.2. Summary of NASN (National Air Surveillance Network) suspended-particle samples for urban stations, by population class, during the period 1967-63. From [2.5]

Pop. class	No. of samples	No. of stations <sup>a</sup>	Min. [ $\mu\text{g}/\text{m}^3$ ]	Max. [ $\mu\text{g}/\text{m}^3$ ]	Arith. mean [ $\mu\text{g}/\text{m}^3$ ]	Geo. mean [ $\mu\text{g}/\text{m}^3$ ]
3 million and over	316	2	57	714	182	167
1-3 million	519	3	34	597	161	146
0.7-1.0 million	1191	7	14	658	129	113
0.4-0.7 million	3053	19	18	977	128	112
0.1-0.4 million	9531	92	10	1706	113	100
50 000-100 000	5806	81	6	982	111	93
25 000-50 000	1606	23	5	679	85	71
10 000-25 000	484	6	11	539	80	63
< 10 000	150	5	22	396	100	84

<sup>a</sup> 64 Stations participate every year: the remaining stations participated 1 or more years during the 7-year period.

The shapes of the aerosols range from the very irregular fly ash to the spherical liquid droplets. Also, the size of the aerosols vary, but under steady-state conditions normally range from 0.01 to 20  $\mu\text{m}$  in radius. These limits are generally controlled by natural processes: the lower limit because smaller particles tend to coagulate, and the upper limit due to gravitational settling. A typical distribution of aerosol sizes in the atmosphere, measured by JUNGE [2.4], is shown in Fig. 2.2. Concentrations of aerosols and particles vary considerably and depend on meteorological conditions and the locations of local sources. Two *in-situ* techniques are used to monitor concentration: An electro-optical method which counts particles in certain size ranges, and provides a measure of aerosol number density; and a filter technique which monitors the total mass of particles collected (in  $\mu\text{g}/\text{m}^3$ ). Concentrations vary from background levels of 5  $\mu\text{g}/\text{m}^3$  to high levels of over 1000  $\mu\text{g}/\text{m}^3$  in urban and industrialized areas. Table 2.2 presents a summary of particulate mass concentration measurements in urban areas of varying sizes.

### *Oxides of Carbon*

Carbon dioxide is not generally considered a pollutant, but because its concentration is increasing globally, it may be a significant factor in the Earth's heat balance. Carbon monoxide, on the other hand, is a toxic pollutant and is especially dangerous to the health of humans.

Carbon dioxide and CO are produced both naturally and by man's activities. Natural sources of  $\text{CO}_2$  and CO include, volcanoes, forest fires, and vegetation decay. In addition, CO is produced in nature by

photochemical oxidation of organic matter. Man's activities contribute significantly to the concentration of CO and, to a lesser extent, to the levels of CO<sub>2</sub>. In both cases, burning of fossil fuels produces the greatest impact. It has been estimated that the automobile is responsible for nearly 80% of the CO in the troposphere [2.3].

The background level of CO<sub>2</sub> (neglecting the direct effects of man's activities) is approximately 320 ppm, and it is generally agreed to be increasing at a rate of approximately 0.7 ppm/year. In urban and industrialized areas concentrations as high as 500 ppm have been observed [2.2]. The background level for CO is considerably lower, and is estimated to be in the range 0.05 to 0.1 ppm. Urban extremes for CO are closely correlated with vehicular traffic patterns, and may range from a few ppm in residential areas to a few tens of ppm in urban centers. Maximum instantaneous measurements of CO have been as high as 100 ppm [2.2].

### *Sulfur Compounds*

The sulfur compounds in the atmosphere considered to be pollutants include SO<sub>2</sub>, H<sub>2</sub>S and sulfate aerosols. These compounds are toxic to man in high concentrations and also produce vegetation damage due both to direct contact and as a result of acid rainfall. In nature, volcanoes produce the majority of SO<sub>2</sub>, while decaying vegetation is responsible for most of the H<sub>2</sub>S. Man's contribution to the levels of sulfur compounds stems mainly from fossil fuel combustion. It has been estimated that nearly 50% of the man-made SO<sub>2</sub> is produced by fossil-fuel-burning electrical generating plants [2.6].

The residence time in the atmosphere for SO<sub>2</sub> is typically less than one week—its concentration is, therefore, strongly dependent on source locations. Even in industrialized regions the concentration is generally less than 0.1 ppm. However, much higher average concentrations have been observed. During the London episode in December 1952, the average concentration over a two-day period reached 1.34 ppm [2.2]. Hydrogen sulfide's background concentration is much lower, and has been estimated to be approximately 0.2 ppb [2.6]. In isolated locations H<sub>2</sub>S has been measured at concentrations greater than 1 ppm [2.4]. Sulfate aerosol concentrations vary widely from a background of a few µg/m<sup>3</sup> to a high of over 600 µg/m<sup>3</sup> during the 1952 London episode [2.5].

### *Nitrogen Compounds*

a) *Nitrogen Oxides.* The oxides of nitrogen considered most toxic include both NO and NO<sub>2</sub>, often referred to cumulatively as nitrogen

oxides ( $\text{NO}_x$ ). They are toxic to man in the ppm range and cause plant damage even at lower concentrations.  $\text{NO}_x$  are the basis for the formation of nitrate aerosols and contribute to the formation of photo-chemical smog through their reaction with hydrocarbons. In nature,  $\text{NO}_x$  are formed as NO through biological activity. Man's contribution to the levels of  $\text{NO}_x$  is primarily due to the production of NO during high-temperature combustion. Both natural and man-made NO are further oxidized in the atmosphere to  $\text{NO}_2$ .

The concentration levels of  $\text{NO}_x$  are highly variable in the atmosphere, depending on source strengths and locations, and the conditions of the atmosphere. For example, the concentration of  $\text{NO}_x$  in Los Angeles ranges between 0.05 ppm (non-smoggy day) to recorded maxima of nearly 4.0 ppm [2.2].

b) *Ammonia*. The significance of ammonia is its role in the formation of ammonium sulfate aerosols. Its major source is the natural decay of animal and vegetable matter. It has been estimated that man produces less than 1% of the total ammonia in the atmosphere, mainly in combustion by-products and chemical manufacturing. Ammonia concentrations range from a low of a few ppb to highs of a few ppm near local sources [1.2].

c) *Organic Nitrogen Compounds*. Organic nitrogen compounds such as PAN and other peroxyacetyl nitrates are considered to be the principal irritants in photo-chemical smog. Their concentrations range downward from 35 ppb [2.2].

### *Hydrocarbons*

The term hydrocarbons refers to a wide range of hydrogen-carbon-containing compounds. Hydrocarbons are generally classed in terms of their reactivity—with the olefins being most reactive, benzene being slightly reactive, and methane considered to be nearly non-reactive. The significance of hydrocarbons is the role they play in the formation of photo-chemical smog. They are often referred to as oxidant precursors because of the reaction  $\text{HC} + \text{NO}_x + h\nu \rightarrow \text{O}_3 + \text{byproducts}$ . The natural sources of hydrocarbons include terpenes from forests and vegetation, and methane from decomposition of organic matter. On the average, man's activities produce nearly 15% of the total hydrocarbons present in the atmosphere through the combustion of fossil fuels and the processing and utilization of petroleum products. Because of the low reactivity of methane, our primary interest is with non-methane hydrocarbons. The non-methane component may be as high as 50% of the total concentration. Background levels of methane have been measured



to be approximately 1.5 ppm [2.3]. However, in urban areas, total hydrocarbons range from a few ppm to maximum instantaneous concentrations in Los Angeles of 40 ppm [2.2].

### *Ozone*

The significance of ozone in the troposphere is its toxicity to man in concentrations as low as 100 ppb. The natural sources of ozone include transport from the stratosphere, lightning, and volcanic eruptions. Man contributes to this natural level by releasing into the atmosphere hydrocarbons and  $\text{NO}_x$  which react in the presence of sunlight to produce ozone by the reaction discussed in Subsection 2.1.2 under *Hydrocarbons*.

Background levels of ozone at sea level range between 10 and 30 ppb; however, during air pollution episodes, concentrations as high as 500 ppb have been observed [2.6].

### **2.1.3 Stratosphere**

The basic constituents of the atmosphere,  $\text{N}_2$ ,  $\text{O}_2$ , argon and  $\text{CO}_2$ , are mixed uniformly from the earth's surface through the stratosphere to altitudes of approximately 100 km. Water vapor, other trace gases, and aerosols or particles are variable in concentration both spatially and temporally within the stratosphere. The variable constituents of the stratosphere will be discussed in the following sub-sections.

#### *Aerosols and Particles*

Aerosols and particles exist throughout the stratosphere. The concentration peaks near an altitude of 20 km, referred to as the "20 km" or "Junge" aerosol layer. Examples of early measurements of this layer are shown in Fig. 2.3. The primary significance of aerosols in the stratosphere is their possible effects on radiation transfer through the atmosphere and subsequent effect on the total radiation balance of the earth. Chemically the particles have been found to be primarily sulfates, probably due to oxidation, at stratospheric altitudes, of sulfur-containing gases such as  $\text{SO}_2$  and  $\text{H}_2\text{S}$ . The source of  $\text{SO}_2$  and  $\text{H}_2\text{S}$  is a combination of man-made and naturally occurring gases transported to the stratosphere from the troposphere. Another major source of particulate matter in the stratosphere is sulfur-containing gases and particles emitted by volcanic eruptions and injected into the atmosphere up to stratospheric altitudes. It has been suggested that volcanoes are the principle source of particulate matter in the stratosphere: An example is

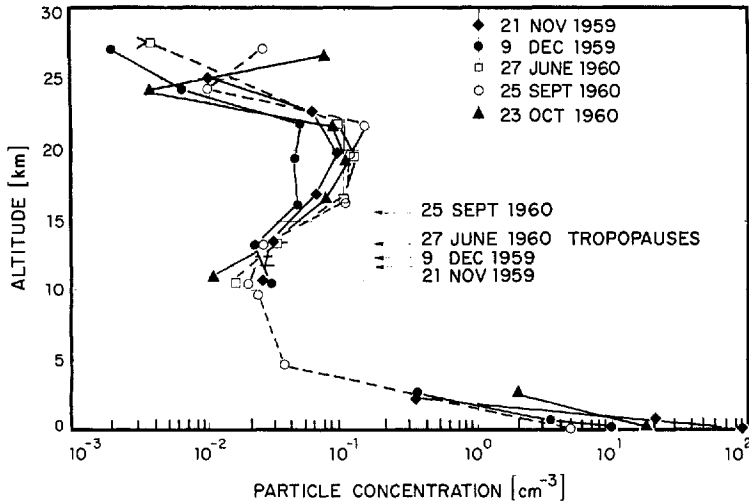


Fig. 2.3. Vertical distribution of stratospheric particles collected by inertial impactors. From [Ref. 2.6, p. 199]

the eruption of Mt. Agung in March 1963. During the winter of 1963–64, the aerosol concentrations were measured to be as much as a factor of 20 above the data shown in Fig. 2.3. By 1970 the stratospheric aerosol concentration had receded to the pre-Agung levels. More recently, the eruption of Volcan de Fuego in Guatemala again produced an elevated stratospheric aerosol concentration [2.7]. Fluctuations of this magnitude which are correlated with volcanic activity strongly indicate that natural processes determine the major quantity of particulate matter in the upper atmosphere.

#### *Minor Molecular Constituents*

a) *Sulfur Components.* As mentioned in the preceding subsection, the significance of sulfur compounds such as  $\text{SO}_2$  and  $\text{H}_2\text{S}$  is the role they play in the formation of the sulfate aerosol.

b)  $\text{NO}_x$ . The significance of  $\text{NO}$  and  $\text{NO}_2$  in the stratosphere is their possible contribution to the depletion of ozone. The concentration of  $\text{NO}_x$  varies from approximately  $10^8$  to  $10^{10}$  molecules/ $\text{cm}^3$  in the altitude range from 10 to 30 km.

c) The significance of nitric acid in the stratosphere is its role in the removal of  $\text{NO}_2$  and its formation of nitric acid aerosols. The distribution of nitric acid is similar to ozone, peaking in concentration at approxima-

tely 20 km. The concentration is between  $10^9$  and  $10^{10}$  molecules/cm<sup>3</sup> between 10 and 30 km.

d) *Methane*. It has been suggested that methane in the stratosphere may be involved in ozone photo-chemistry [2.6]. The reaction results in methane being oxidized to form water vapor and CO<sub>2</sub>. The most significant effect, then, is methane's contribution to the increase in stratospheric water vapor. Methane varies in concentration between  $10^{11}$  and  $10^{13}$  molecules/cm<sup>3</sup> in the altitude range 10 to 30 km [2.8].

e) *Ozone*. The importance of ozone in the stratosphere is that it provides the earth with a shield against high energy ultraviolet radiation from the sun. The mixing ratio of ozone peaks at approximately 8 ppm in the altitude range between 25 and 30 km. The peak value and distribution of ozone, however, is highly variable both spatially and temporally.

f) *Chlorofluorocarbons*. It has been postulated that chlorofluorocarbons in the stratosphere may undergo photo-dissociation due to the presence of high-energy ultraviolet radiation, and their by-products may significantly deplete the ozone concentration. Chlorofluorocarbons, particularly the freons, are used as refrigerants and aerosol can propellants. Because of their inertness in the troposphere, the global concentration of chlorofluorocarbons has been steadily increasing, approximately doubling in the three years between 1972 and 1975; and it is feared that an increasing amount is being transported into the stratosphere [2.9]. Recent measurements indicate that the stratospheric chlorofluorocarbon concentration is of the order of a few hundred parts per trillion at the present time [2.8].

## 2.2 General Pollution Transport Modeling

As mentioned in the introductory section of this chapter, the connection between sources of air pollution and the location of a critical receptor of the pollutant involves transport, transformation, and dispersion through the atmosphere. A complete and thorough treatment of the transport and diffusion of a number of pollutants in a given region is desirable in order to uniquely determine the cumulative concentrations at all points within the region as a function of time, but generally impossible to obtain since it requires data approaching the spatial scale of molecules and temporal scale of molecular collision times. Fortunately, for most applications an understanding of atmospheric transport and diffusion between sources and receptor to this extent is not required. In an attempt to understand and predict transport and diffusion

through the atmosphere, a number of mathematical models have been developed. These models are used to:

- a) Predict ambient concentrations due to existing pollutant sources.
- b) Establish maximum source strengths while maintaining safe ambient levels.
- c) Assess the impact of proposed new or modified sources on ambient concentrations.

### **2.2.1 Description of Mathematical Models**

The simplest mathematical model for predicting the transport and dispersion of pollutants through the atmosphere is referred to as the box model. In the development of this model it is assumed that the area under study can be approximated as rectangular. The atmosphere above this area is contained in a hypothetical box whose height above ground level corresponds to the height of the mixed layer. The sides of the box correspond either to significant geological features, such as mountain ranges, or are positioned so that two sides are perpendicular to the prevailing winds. In more advanced models the area box is divided into a large number of smaller boxes whose square bases are located at ground level and whose height either matches the mixed layer height or some fraction of that height, thereby providing a number of stacked boxes over each square base. Thus, the larger area box is divided into a large number of small boxes both horizontally and vertically.

In the simplest application of the multi-box model to predict pollutant transport, the format below is followed:

- 1) An accurate pollutant source inventory is made providing source strengths and locations.
- 2) Pollutant source strengths in each small box are summed to yield an approximate average concentration for the small box.
- 3) Wind data and estimated atmospheric stability are used to provide the trajectory and dispersion of pollutants from each small box.
- 4) The resultant average concentration of pollutants in any given small box, as a function of time, is estimated to be the summation of its own average source strength and the contribution of pollutants to and from other small boxes, transported by the wind.

This model, and other more sophisticated ones, will continue to be improved, thus providing a better understanding of pollutant transport. The treatment of chemical reactions and other transformation processes during transport, which act as either sources or sinks of pollutants, is an example of recent improvements in some mathematical models.

One important characteristic of this class of models, as far as laser monitoring is concerned, is that the *average* pollutant concentration is predicted for spatial dimensions equal to the size of the small boxes. In most applications of these models, the dimensions are typically 1 km horizontally and 0.2–1 km vertically.

### 2.2.2 Monitoring Needed for Model Development and Validation

In order to provide data for the validation and development of models, dedicated methods for monitoring pollutants, which provide spatial and temporal resolution similar to model predictions, are required. Present *in-situ* techniques monitor at a single point in most cases, and thus do not provide data which are representative of the *average* concentration over the spatial extent of the model predictions. Integrated horizontal measurements at ground level and integrated vertical measurements through the mixed layer matched to the spatial scales of the models will provide data to either validate developed models or act as feedback to improve model predictions. Measurements of this type can be performed either by mobile *in-situ* instruments which are moved horizontally and/or vertically over the prediction grid in a time comparable to the temporal scale of the model or, more preferably, by remote-monitoring instrumentation which measures integrated concentration (long-path laser absorption), or a profile concentration which can subsequently be integrated (various LIDAR applications).

Until air pollution monitoring provides data on the proper spatial and temporal scales, the utility of atmospheric transport models cannot be firmly established. Remote monitoring techniques hold promise for cost-effectively providing these data.

## 2.3 Remote Monitoring Capabilities of Laser Systems

This section will present a brief overview of the capabilities of remote-monitoring laser instrumentation. Many of the techniques discussed will be covered in detail in subsequent chapters.

Remote monitoring can be defined as sensing qualitatively and/or quantitatively a chemical or physical parameter in the environment where the monitoring instrument and the parameter under investigation are spatially separated. In the atmosphere, remote sensing techniques can be divided into two broad categories: active and passive, depending on the source of radiation. Passive techniques make use of available radiation in the atmosphere (e.g., solar and earth-reflected or emitted

radiation). The interaction of this radiation with the species under investigation (absorption and scattering) or the thermal emission of the species is observed to infer concentration. Active techniques, on the other hand, are characterized by the introduction of specific radiation into the atmosphere, typically incorporating lasers as the source of this radiation. The interaction (scattering, absorption, fluorescence) with the atmosphere is observed to infer species concentration. The remaining discussion of this section will be limited to active techniques which use lasers as the source of radiation<sup>1</sup>. Active techniques, for the purpose of this discussion, will be further subdivided into single-ended and double-ended systems.

### 2.3.1 Single-Ended Systems

Single-ended systems, as the name implies, co-locate the laser source and telescope receiver. Generally single-ended systems depend on scattering as the primary means for interrogating the atmosphere. This may be either direct scattering by the species of interest, or scattering by other atmospheric constituents, with the species of interest modifying the transmission of the laser beam in a measurable manner. Direct scattering techniques include LIDAR, Raman LIDAR and resonance LIDAR, whereas, indirect scattering techniques include: differential absorption scattering LIDAR and an additional application of Raman LIDAR. Each of these techniques will be discussed separately.

## LIDAR

LIDAR is an acronym for Light Detection And Ranging. All single-ended LIDAR systems consist of a laser and telescope whose optical axes are aligned parallel such that the telescope field-of-view includes the laser beam as it propagates through the atmosphere. The differences in the LIDAR systems mentioned above are in the selection of the laser wavelength, the receiver wavelength, and the data analysis and interpretation.

In the most basic system, referred to simply as LIDAR, the detector wavelength is matched to the laser wavelength. As the laser radiation propagates through the atmosphere, it interacts with aerosols and molecules. The principle interaction of interest is elastic scattering by the aerosols. Some of the scattered radiation is collected by the telescope

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<sup>1</sup> Lasers can also be used in the passive mode as local oscillators to detect radiation by high-resolution, sensitive heterodyne techniques, as discussed in Chapter 7.

and detected by a sensitive photomultiplier. The detected signal is recorded as a function of time to provide a range-resolved measure of atmospheric scattering. Analysis of the recorded data is then performed to provide an indication of aerosol distribution in both the troposphere and stratosphere.

### *Raman LIDAR*

In Raman LIDAR the receiver is made sensitive, through the use of a spectrometer or interference filter, to Raman-shifted wavelengths. Most molecules scatter electromagnetic radiation not only at the excitation wavelength but also at specific shifted wavelengths by the phenomenon referred to as Raman scattering. The magnitude of the shift is unique to the scattering molecule, and the intensity of the Raman band is proportional to the scattering molecule's concentration. Raman LIDAR has been applied to the measurement of a number of atmospheric molecules, including H<sub>2</sub>O, SO<sub>2</sub> and CO<sub>2</sub>. The most significant limitation of the technique arises from the low cross section for Raman scattering. Thus, the application requires the use of high-power lasers, large telescopes, long integration times, and is generally limited to measurements where high molecular concentrations are present.

### *Resonance LIDAR*

Resonance LIDAR is characterized by a careful selection of both laser and receiver wavelength so that they are matched to an absorption wavelength of the atmospheric species of interest. Stimulating a molecule at an absorption frequency produces resonant scattering which may be significantly more intense than non-resonant scattering. The utility of resonance LIDAR is limited in the lower atmosphere because radiationless quenching reduces the scattering intensity in a nonlinear fashion dependent on the concentration of the quenching molecules. However, this technique has been successfully applied to the measurement of high-altitude concentrations of atomic sodium and potassium.

### *Differential Absorption Scattering LIDAR*

This system is the first of the indirect scattering techniques to be discussed. It depends on scattering by aerosols in the atmosphere but measures selected molecular species due to their absorption. The technique makes use of at least two laser beams at different wavelengths which are sequentially or simultaneously transmitted along the same path in the atmosphere. One laser beam is absorbed by the molecular

species of interest, whereas the other at a nearby wavelength is not strongly absorbed. Since the beams are spectrally separated by a small wavelength increment, the cross-section for aerosol scattering may be essentially identical for each. To the extent that this is valid, the difference in scattered intensity of the beams as they propagate is due to the difference in absorption by the molecule under investigation. Analysis of the detected signals from both beams as a function of time provides a range-resolved measurement of the absorbing molecule's concentration.

### *Raman LIDAR for Visibility*

Raman LIDAR, as described above, can also be applied to the measurement of atmospheric attenuation. In this application Raman signals from either  $N_2$  or  $O_2$  in the atmosphere are monitored. Since the distribution of these major constituents is well known, a comparison of the Raman signal, which includes attenuation, with the known molecular distribution provides a range-resolved measure of double-path attenuation. In many cases, especially under low-visibility, attenuation can be related to visibility.

### **2.3.2 Double-Ended Systems**

Double-ended systems are characterized by either having the laser transmitter and receiver telescope located separately or having the laser and telescope co-located with a physical reflector located at a distance. The two principle double-ended systems are Bistatic LIDAR and Long Path Absorption. These techniques will be discussed separately.

#### *Bistatic LIDAR*

Bistatic LIDAR is similar to the LIDAR system discussed above except that the laser and telescope are separated by a distance. In the utilization of this technique, both the laser and telescope are aimed toward the same point in the atmosphere. The beam divergence of the laser and the field-of-view of the telescope together define the scattering volume at the point of intersection. As the laser pulse propagates through the scattering volume, some of the energy scattered elastically by atmospheric aerosols is collected by the telescope and detected by a photomultiplier. The output of the photomultiplier is recorded. By carefully varying the pointing angle of both the laser and telescope, measurements of scattering at a fixed altitude can be observed for a variety of scattering angles. The bistatic LIDAR technique can provide valuable scattering angle data which can be interpreted to infer the refractive index and/or size distribution of atmospheric aerosols.



### *Long Path Absorption*

This technique utilizes the absorption of a laser beam as it propagates through the atmosphere as the measurable parameter to infer molecular concentration. The laser source and telescope receiver can be separated and pointed toward each other; but, for ease of operation, the laser and telescope are generally co-located with their optical axes aligned and pointing to a retro-reflector or topographical target. As in the Differential Absorption Scattering LIDAR discussed above, at least two laser beams at slightly different wavelengths are used to interrogate the intervening atmosphere between the system and the reflector. The two beams are adjusted in wavelength to be “on” and “off” an absorption band of the molecule under investigation. Comparison of the two signals collected by the telescope provides a measure of the integrated concentration of the molecular species along the transmission path. In some applications of the technique more than two beams of differing wavelength are required because of multiple molecular absorption. The number of beams required is at least one more than the number of significant molecular absorbers. A promising outgrowth of this technique is the use of topographical targets instead of cooperative retro-reflectors. The targets may be either trees, hills, or buildings when using a horizontal ground-based system, or the earth’s surface for an airborne system.

With this discussion of the large variety of active remote-sensing techniques and systems which are available and/or being developed for monitoring the atmosphere, the question remains: How will these and other remote-monitoring systems be used in air quality management? The answer to this question is the subject of the next section.

## **2.4 Remote Monitoring in Air Quality Management**

As mentioned in the introductory section of this chapter, remote monitoring holds promise for providing three-dimensional pollution concentration data of the atmosphere. The large number of pollutants of concern and their significance, distribution, and concentration levels were reviewed in Section 2.1. Section 2.2 described mathematical transport models which attempt to predict the movement of pollutants from their source to the critical receptor, and a discussion of the various laser techniques for remotely sensing the atmosphere was presented in the last Section 2.3. The question to be addressed here is: What are the unique advantages of remote atmospheric monitoring, when compared to present methods, which will insure that these techniques will be

incorporated into air quality management systems of the future? To answer this question adequately, the unique advantages will be considered, and specific applications based on these advantages will be listed.

### **2.4.1 Advantages of Remote Monitoring**

Although it is reasonable to assume that remote monitoring instrumentation may generally provide as accurate or precise measurements as *in-situ* instrumentation, they do, as a class of techniques, provide certain unique advantages: namely, they permit specific applications which would be difficult, if not impossible, using standard instrumentation. Some of these advantages are listed below and will be discussed individually:

- 1) Non-interfering for source effluent monitoring.
- 2) Integrated-path measurements.
- 3) Measurements at ground level and aloft.
- 4) Perspective in monitoring.
- 5) Measurements over large geographical areas.

#### *Non-Interfering*

Remote monitoring of pollutant emissions at their source is non-interfering in two ways. Since it is a probeless technique, no extraction of source gases is required, thus eliminating the possibility of modifying the sample during the measurement. In addition, it has the potential for providing specific measurements of effluent concentration without interfering with the operation of the industrial facility under investigation. Measurements made off the property of the facility provide an opportunity to perform unannounced inspections of emission concentrations.

#### *Integrated-Path Measurements*

The utility of integrated measurements for pollution transport model verification and development has been previously discussed (Sec. 2.2). In this case, remote-monitoring laser techniques provide measurements on a spatial scale comparable with model predictions. Another need for integrated measurements is related to the accurate estimation of total pollutant dose to humans. Remote monitoring can provide this information with integrated measurements over an area which approximates the motion area of the population under study.

#### *Measurements at Ground Level and Aloft*

To obtain a better understanding of air quality, it is becoming increasingly recognized that measurements at ground level must be supple-

mented by measurement aloft. Laser monitoring may prove to be the cost-effective method to monitor pollutant concentrations aloft. Three-dimensional measurements are also needed to assess the impact of photo-chemical reactions and to trace pollutants from their source to the receptors. Measurements as a function of altitude are required for model development and verification, since most models do not specifically predict ground-level pollution concentration.

#### *Perspective in Monitoring*

Conventional *in-situ* instruments normally provide an accurate measurement at only one location. Pollutant concentrations in the atmosphere surrounding this location may vary significantly, thus invalidating the measurement as being representative of the larger area. Local sources, meteorology, topography and man-made structures all contribute to possible variability. A single remote monitoring instrument located in the area has the potential for providing accurate representative measurements. In addition, remote sensing can be used in initial surveys to optimally locate *in-situ* monitoring networks.

#### *Measurement Over Large Geographical Areas*

There are many large air sheds which require monitoring. This monitoring would be performed initially to establish a baseline so that, in the future, changes in air quality could be determined. Laser instrumentation mounted on mobile platforms (aircraft, trucks) may prove to be the most cost-effective technique to measure air quality over these large geographical areas.

### **2.4.2 Applications of Remote Monitoring**

As mentioned and discussed in the previous section, there are a number of unique advantages in employing laser remote-sensing instrumentation for monitoring air quality. By making use of these advantages, remote monitoring techniques can significantly complement the present *in-situ* methods. A number of specific applications are listed below:

- 1) Monitoring opacity of a source plume.
- 2) Monitoring specific effluents at the source.
- 3) Measuring plume transport and diffusion.
- 4) Monitoring to determine representativeness of point measurements (pollutant variability studies).
- 5) Surveying to design optimum *in-situ* monitoring networks.
- 6) Monitoring for verification and development of pollution transport models.

- 7) Measuring mixed height over large air sheds.
- 8) Monitoring long-range transport of pollutants from urban and industrialized areas.
- 9) Monitoring expansive wilderness areas to determine air quality trends.
- 10) Assessing proposed sites for new sources.
- 11) Monitoring during air pollution episodes.

These are some of the applications of laser remote-monitoring instrumentation currently under consideration. It is certain that others will be recognized as the technology is applied to the requirements of air quality management.

## 2.5 Conclusion

There is now, and will continue to be, a need for understanding atmospheric processes in connection with the transport and conversion of pollutants from both natural and man-made sources. Not only is the lower atmosphere which we breathe important, but so is the upper atmosphere which contains gases and particles which affect the spectrum and intensity of solar radiation impinging on the Earth's surface. A case has been made here for the use of laser remote-sensing techniques for achieving some of the monitoring goals associated with model development and atmospheric surveillance. Specific monitoring techniques based on the use of laser radiation will be considered in detail in the following chapters.

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