

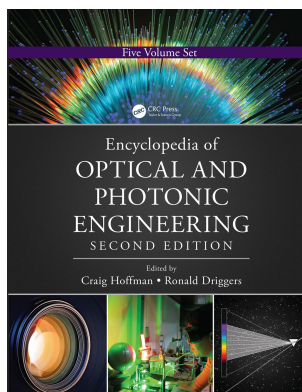
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Publisher: *CRC Press*

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Encyclopedia of Optical and Photonic Engineering

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Publication details

<https://www.routledgehandbooks.com/doi/10.1081/E-EOE2-120009502>

Arkadi Zilberman

How to cite :- Arkadi Zilberman. 22 Sep 2015, *Aerosols from: Encyclopedia of Optical and Photonic Engineering* CRC Press

Accessed on: 14 Jun 2021

<https://www.routledgehandbooks.com/doi/10.1081/E-EOE2-120009502>

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Aerosols

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Abstract

Atmospheric aerosols comprise a dispersed system of small solid and liquid particles suspended in air. They remain suspended for varying periods of time, and are then transported by vertical and horizontal wind currents, frequently to great distances. Aerosols are formed by a primary source that includes dispersion of particulates from the Earth's surface and a secondary source resulting from atmospheric chemical reactions, condensation, or coagulation processes. This entry summarizes the knowledge about aerosols and their size distribution, and present some measurements and measurement techniques.

INTRODUCTION

Atmospheric air is never free from particles having a variety of origins, sizes, and chemical compositions. Atmospheric aerosols comprise a dispersed system of small solid and liquid particles suspended in air. Remaining suspended for varying periods of time, they are transported by vertical and horizontal wind currents, frequently to great distances. Aerosols are formed by two main processes: a primary source which includes dispersion of particulates from the Earth's surface (like soil and deserts, oceans, volcanoes, biomass burning, industrial injection) and a secondary source resulting from atmospheric chemical reactions, condensation, or coagulation processes. Aerosol concentrations and properties depend on the intensity of the sources, on the atmospheric processes that affect them, and on the particle transportation from one region to another. The size distribution of the atmospheric aerosol is one of its core physical parameters. It determines the various properties like mass and number density, or optical scattering, as a function of particle radius. For the atmospheric aerosols, this size range covers more than five orders of magnitude, from 10 nm to several hundred micrometers. Like air masses or atmospheric clouds, the aerosol-size distribution varies from place to place, with altitude and with time.

OVERVIEW

Aerosols play important roles in weather, climate, and air quality. Even for clear skies, the particulates affect the heating or cooling of the atmosphere through scatter and absorption of energy. Increases in aerosol loading of the atmosphere can lead either to an increase or a decrease in the mean global temperature of the Earth. Indeed, the interaction with solar and terrestrial radiation by aerosols perturbs the radiative budget via scattering and absorption of sunlight.^[1,2] By acting as cloud condensation nuclei or ice

nuclei,^[3,4] aerosol particles also modify the cloud microphysics. As a result, aerosol particles may change the cloud radiative properties.^[5,6] The direct effect of aerosols on radiation budget due to reflection of sunlight to space and the indirect effect on cloud albedo by the modification of cloud properties may cause a cooling effect that may counterbalance the warming due to the increase in carbon dioxide concentration.^[7,8] In atmospheric chemistry, aerosols also serve as the liquid phase that increases the speed of chemical reactions.^[9]

Large urban areas often are sources of extreme heavy air pollution, affecting both the radiation balance and the climate for extended regions around them and the population's health within them. Since the 1970s and 1980s, important new data have been gathered about the direct health effects of aerosol particles.^[10] There are indications that micrometer- and submicrometer-sized particles can cause increases in morbidity in urban dwellers, possibly more than any gaseous air pollutant at current concentrations.

The performance of electrooptical systems is substantially affected by aerosol particles that scatter and absorb electromagnetic radiation.^[11,12] Knowledge of the parameters that determine the optical properties of atmospheric aerosols is essential for development of techniques for optical communication and imaging through the atmosphere; laser weaponry; remote sensing, in particular, from space; or the necessary correction of atmospheric effects in satellite imagery. The estimation of the performance of electrooptical systems depends on the accuracy of the atmospheric models being used in the propagation prediction codes. Large uncertainties remain in the models for aerosol constituents. Difficulties in estimating the influence of aerosols on electrooptical system performance and their climatic impact arise from the high spatial and temporal variability of aerosol concentrations, and the physical and the chemical properties. In addition, the large size range makes the measurements of aerosols very difficult, time-consuming, and susceptible to error.

Our knowledge about atmospheric aerosols has grown considerably, but still remains fractional and limited, especially as a function of altitude.

There is vast literature in the area of aerosol studies^[13–17] and remote sensing,^[18–20] including data and models of aerosol extinction, size distribution, and properties for different latitudes. Many works introduce new questions about aerosol sources, transformation, transport, and sink processes as well as influence on performance of electrooptical systems that are still unresolved.

This entry will summarize the knowledge about the aerosol and its size distribution, and presents some measurements and measurement techniques.

AEROSOL SOURCES AND CLASSIFICATIONS

Aerosols may be introduced into the atmosphere as wind-raised dust and sea salt, products of combustion, such as soot, ash, condensed organics, and products formed by chemical reactions within the atmosphere involving gaseous materials such as sulfates, nitrates, H₂S, NH₃, etc.

Oceans serve as a major source of natural aerosols and through air–sea exchange, contribute greatly to the global cycles of carbon, nitrogen, and sulfur aerosols.^[21] Liquid water from oceans can also be transferred to the atmosphere through air bubbles at the surface as well as sea-salt aerosols when water evaporates. Biomass burning is an important source of organic particles, while arid and semiarid regions are mainly the sources for mineral dust.^[22] Industrial and urban emissions are a major source of aerosols, which consist of complex and variable mixtures of primary solids, such as mineral dust and graphitic carbon, and of secondary, mainly water-soluble, particulate substances like sulfates, nitrates, and organic matter formed from gaseous emissions.^[23] These anthropogenic aerosols are recognized to be of paramount importance in the Earth's radiative budget due to both their optical and cloud condensation properties. Moreover, urban aerosols have also a major influence on the chemistry of trace gases by modifying the solar radiative fluxes.

In a first attempt to sort into geographically distinct atmospheric aerosols, Junge^[24] classified aerosols depending on their location in space and sources into background, maritime, remote continental, and rural. This classification later was expanded and quantified.^[25]

Some studies were devoted to specific aerosols types: desert aerosols,^[26,27] urban aerosols,^[28] or aerosols resulting from biomass burning in tropical regions,^[29] and stratospheric aerosols.^[30] Of course, all classification models only reflect certain average values. Individual distributions vary depending on local weather and wind, vertical mixing, horizontal transport, gas-to-particle conversion, season, etc. On a global scale, aerosol climatology was summarized by d'Almeida et al.^[17] It includes a compilation of a large

amount of data and tabulates the dominant type of aerosols as a function of latitude, longitude, and season.

From all classifications of atmospheric aerosols, the most commonly used one is according to size. General classification suggests three modes of aerosols:^[14] 1) a *nuclei* mode which is generated by spontaneous nucleation of the gaseous material for particles less than 0.1 μm in diameter; 2) the *accumulation* mode for particles between 0.1 and 1 μm diameter, mainly resulting from coagulation and in cloud processes; and 3) the *coarse* mode for particles larger than 1.0 μm in diameter originating from the Earth's surface (land and ocean). The classification is quite similar to the Junge's designation^[24] referred to as Aitken, large, and giant particles. The particles vary not only in chemical composition and size but also in shape (spheres, ellipsoids, rods, etc.).

Sea-salt particles are important components of all aerosols. The salt is injected into the air as small droplets of seawater by the bursting of innumerable bubbles formed by cresting waves at all ocean surfaces. On the average, each bubble produces a few large droplets and hundreds or more small droplets. The largest droplets are of such a size that the salt residue, when the water is evaporated, has an effective diameter of 2–3 μm. The small droplets tend to yield residues less than 0.3 μm in diameter. Most of the droplets are caught up by the wind, carried aloft, and transported great distances. Each bit of sea salt is a condensation nucleus, alternating between the crystalline and aqueous states as the relative humidity decreases and increases. Sea-salt nuclei are not restricted to maritime aerosols but are also found well inland. The sea-salt number concentrations in clean air masses ranged from about 30 to 100 cm⁻³, with somewhat higher concentration observed at higher wind speed.^[31]

From the optical standpoint, to underline scattering properties of the atmosphere the term *haze aerosol* was introduced.^[32,33] Hazes are polydisperse aerosols in which the size range of particles extends from about 0.01 to 10 μm. Haze is a condition wherein the scattering property of the atmosphere is greater than that attributable to the gas molecules but is less than that of fog. Haze scattering imparts a distinctive gray hue to the sky, which would otherwise appear a deep blue, and is usually the determining factor of visibility. Haze can include all types of aerosols. Cosmic dust, volcanic ash, foliage exudations, combustion products, bits of sea salt—all these are found to varying degrees in haze.

Certain types of haze particles, such as ordinary dust grains, are nonhygroscopic, while other types, such as bits of salt, are highly hygroscopic and display a strong dependence on relative humidity and are thus more highly variable. Dust particles are a relatively passive constituent of atmospheric haze.^[34] They are derived from a number of terrestrial and extraterrestrial sources. Extraterrestrial sources include planetary accretion and meteoroids. Particles due to these two sources are the predominant haze

components in the outermost atmosphere. Volcanoes generate a huge volume of fine ash and gases. These particles are injected high into the stratosphere, where they remain for extended periods of time. In the lowest levels of the atmosphere, surface winds play an important role in sweeping up soil-based dust. Finally, industry and construction are important sources of dust particles.

Hygroscopic particles are derived from a number of sources, primarily vegetation and the sea. Vegetation exudes aromatic hydrocarbons that under the influence of ozone and sunlight oxidize and condense or nucleate into tiny droplets of complex tars and resins. These particles have an average radius of approximately $0.15\ \mu\text{m}$. Their size depends on the ambient relative humidity and available supply of water vapor, and on the extent to which they coagulate by collisions. The hygroscopic particles act as centers or nuclei for condensation of water vapor. Clouds and precipitation are the meteorological results of nuclei that began as haze aerosols.

The smoke aerosol has been introduced as individual aerosol type. Previous attempts to create generalized aerosol models did not include smoke aerosols due to lack of measurements.^[35] The reasons for the difference between smoke and urban/industrial aerosols are complex. Urban/industrial aerosols are affected by gas-phase oxidation that generates particles and, because of a large humidification factor, swelling from humidity. In contrast, smoke has a much smaller humidification factor; thus, particle growth from water intake is much smaller.^[36] Also, the smoke aging process is a short-lived phenomenon that results in increases of mass of roughly 20–50%, at most a 15% increase in radius. The smoke aerosol appears to be bimodal with an accumulation mode peaking at $0.12\text{--}0.16\ \mu\text{m}$ and a coarse mode at roughly $1.5\text{--}10\ \mu\text{m}$.^[37] Smoke aerosols from biomass burning can cover one-third of the South American continent during the burning season and wide areas of tropical Africa and the South Atlantic. Because

smoke aerosols are a global phenomenon, they are suspected of playing a role in Earth's global energy balance. Smoke aerosols absorb and scatter solar radiation and, therefore, may act as a negative direct force by scattering solar radiation back to space.

The range of size for different types of aerosols and their interaction with radiation is summarized in Fig. 1.

AEROSOL REMOVAL PROCESS

Aerosol removal processes include coagulation, fallout, and washout. Because of the nature of these removal processes, residence times may vary widely, from minutes to weeks in the troposphere up to years in the stratosphere, where the removal processes resulting from condensation and precipitation are normally absent.

Coagulation occurs when particles collide and coalesce, forming fewer but larger particles. The small particles in still air coagulate by Brownian motion alone. Collision for larger particles is caused by small-scale turbulence. Aerosols coagulate faster when the particles are of nonuniform size than when they are nearly of the same size. This process affects the population of small particles more than the population of the large particles.

Fallout or sedimentation is due to effects of gravity. The settling of particles is offset to winds and convective currents. The velocity of particles is the equilibrium between acceleration due to gravity and viscous drag due to the air. Because of the vertical variability of atmospheric pressure, there is a large altitude dependence on the terminal velocity. In any case, however, particles larger than approximately $20\ \mu\text{m}$ are rare. In settling through undisturbed air, the particle attains a terminal or equilibrium velocity such that the gravitational force (negative buoyancy) is balanced by the opposing force due to viscous drag. The terminal velocity, v_t , is given by the Stokes–Cunningham

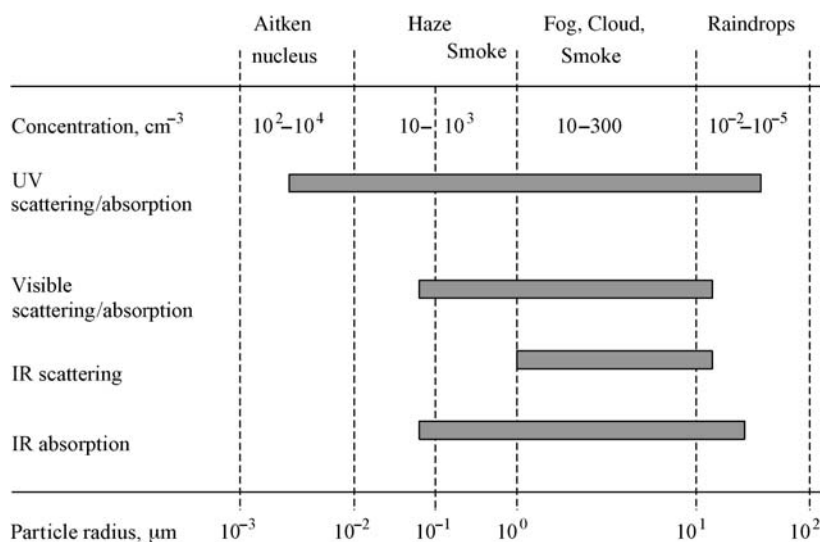


Fig. 1 Aerosol types and their interaction with radiation.

expression:^[38]

$$v_t = \frac{2r^2}{9\eta} g(\rho_p - \rho_a) \left(1 + \frac{B\bar{l}}{r}\right) \quad (1)$$

where ρ_p and ρ_a are the densities of the particle and air, B is a factor whose value lies between 1.25 for $\bar{l}/r \leq 0.1$ and 1.65 for $\bar{l}/r \geq 10$, g is the gravity constant, \bar{l} is the molecular mean free path (6×10^{-6} cm), and η the viscosity of air (1.8×10^{-4} g cm⁻¹ sec⁻¹).

Washout is the removal of particles by rain and snow. Snow, which has smaller terminal velocity and rougher surfaces, may be more effective as a purging effect.

Washout implies some kind of joining action between haze particles and raindrops. Actual physical contact between these objects of such dissimilar size may not be necessary. In fact, a small particle likely is pushed aside by the airstream of a falling raindrop. The washout can be accomplished in several steps. First, some of the nuclei grow into cloud droplets by condensation of water vapor. Next, some of the particles that have not grown coagulate with these droplets. Many of these droplets, having masses far greater than those of the particles, then merge with falling raindrops by the complex processes responsible for raindrop growth.

The coagulation, fallout, and washout continually carry both dust and hygroscopic particles to the ground, tending to prevent an unlimited increase in the atmospheric particle content.

There are different concepts for characterizing the time aerosol particles spend in the atmosphere: relaxation time, residence time, and lifetime. From a combination of individual estimates, an empirical size-dependent model for aerosol residence time has been developed.^[25]

$$\frac{1}{\tau} = \frac{1}{1.28 \times 10^8} \left(\frac{r}{0.3}\right)^2 + \frac{1}{1.28 \times 10^8} \left(\frac{r}{0.3}\right)^{-2} + \frac{1}{\tau_{\text{wet}}} \quad (2)$$

where τ is the residence time (sec), τ_{wet} is the wet removal time (sec), and r is the particle radius (μm). Fig. 2 shows results for various wet removal times, as they are typical for the atmospheric boundary layer, free troposphere, and the stratosphere. It can be seen that the residence time for very large and very small particles is rather short. This is the result of large settling velocities of the giant particles and coagulation of the Aitken particles. This model agrees with observed residence times.

AEROSOL-SIZE DISTRIBUTION MODELS

The manner in which the particle population is spread over the range of sizes is defined by the size distribution function. The size distribution of the atmospheric aerosol is one of its core physical parameters. It determines how the various properties like mass and number density, or optical scattering, are distributed over the particle radius.

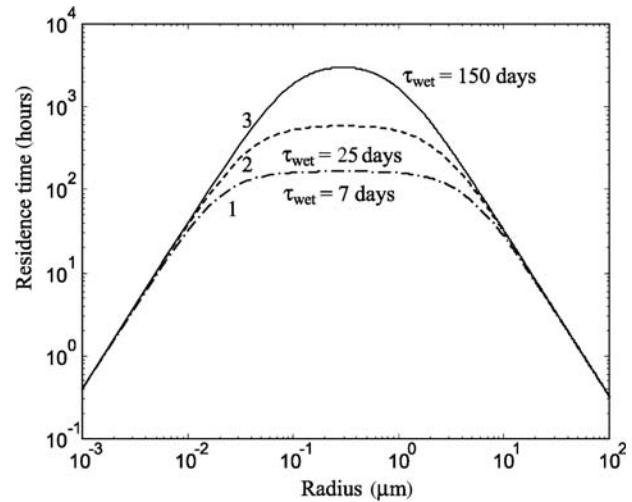


Fig. 2 Residence time of aerosols in boundary layer (1), free troposphere (2), and stratosphere (3) for different wet removal times, τ_{wet} .

Knowledge of the size distribution of atmospheric aerosol is of interest in many areas of aerosol research, e.g., for investigation of aerosol sources and aging processes, for propagation of electromagnetic radiation through the atmosphere. Particle-size distributions are necessary inputs for models used to predict the attenuation and scatter of radiation between the transmitter and receiver in different applications (optical communication, satellite image restoration, weapon-based electrooptical systems, etc.).

The techniques employed to count and measure particles usually provide data on the number of particles per specified interval of radius. The concentration, N , which is the total number of particles per unit volume, is equal to the sum of the class populations

$$N = \sum_{i=0}^k n_i(r_i) \quad (3)$$

If the size classes are narrow, a continuous function can be devised to fit the histogram. When a distribution can thus be expressed by a continuous function, the number $n(r)$ of particles per unit interval of radius and per unit volume is given by

$$n(r) = \frac{dN(r)}{dr} \quad (4)$$

The differential quantity $dN(r)$ expresses the number of particles having a radius between r and $r + dr$, per unit volume, according to the distribution function, $n(r)$.

Because of the many orders of magnitude present in atmospheric aerosol concentrations and radii, a logarithmic size distribution function is often used

$$n(r) = \frac{dN(r)}{d \log(r)} \quad (5)$$

The literature on the subject is quite lengthy, providing useful input to certain theoretical models of $n(r)$ that have been developed to fit the bulk of the observations.

The much used distribution function is the power-law distribution first presented by Junge.^[24,39]

Junge's model is

$$n(r) = \frac{dN(r)}{d \log r} = cr^{-\beta}, \quad r \geq r_{\min} \quad (6)$$

Eq. 6 can be put in a nonlogarithmic form as

$$n(r) = \frac{dN(r)}{dr} = 0.434cr^{-(\beta+1)} \quad (7)$$

Here, c is the normalizing constant to adjust the total number of particles per unit volume, and β is the shaping parameter. Most measured size distributions can best be fit by values of β in the range $2 \leq \beta \leq 4$, for hazy and clear atmospheric conditions and for aerosols whose radii lie between 0.1 and 20 μm .^[40,41] Actual particle-size distributions may differ considerably from a strict power-law form. On the average, however, the power law seems to be a good representation of aerosols having a wide variety of origins and compositions.

The power-law distribution does not permit to show us modulations in the shape of the distribution. To allow for the drop-off of particle number density at small radii, the modified gamma distribution function was introduced.^[42] It has the form:

$$n(r) = \frac{dN(r)}{dr} = ar^\alpha \exp(-br^\beta) \quad (8)$$

where a is the total number density; and α , β , and b are the shaping parameters.

The total particle concentration, given by the integral over all particle radii, for this distribution is^[41]

$$N = a\beta^{-1}b^{-(\alpha+1)/\beta}\Gamma\left(\frac{\alpha+1}{\beta}\right) \quad (9)$$

The mode radius for this distribution is given by

$$r_c^\beta = \frac{\alpha}{b\beta} \quad (10)$$

The value of the distribution at the mode radius is

$$n(r_c) = ar_c^\alpha \exp(-\alpha/\beta) \quad (11)$$

Because it has four adjustable constants, Eq. 8 can be fitted to various aerosol models.

Still another commonly used distribution is the log-normal distribution given by the expression:^[43,44]

$$\frac{dN(r)}{dr} = \frac{N_0}{r\sqrt{2\pi} \ln \sigma} \exp\left[-\frac{1}{2} \frac{\ln(r/r_m)^2}{\ln \sigma}\right] \quad (12)$$

where σ is the standard deviation of $\ln r$, r_m is the mode radius, and N_0 is the total particle concentration.

This form of distribution function permits fitting the multimodal nature of the atmospheric aerosols. Harris^[45] and Davies^[46] suggested to use the sum of four log-normal distributions and the sum of as many as seven distributions to fit a measured aerosol-size distribution. Remer and Kaufman,^[28] Shettle and Fenn,^[44] and Nakajima et al.^[47] have shown that two modes are generally adequate to characterize the gross features of most aerosol distributions. Three superimposed log-normal distributions are also often used. This permits in many cases testing of three modes of aerosol distribution.^[48]

$$\frac{dN(r)}{dr} = \sum_{i=1}^3 \frac{N_i}{r\sqrt{2\pi} \ln \sigma_i} \exp\left[-\frac{1}{2} \frac{\ln(r/r_{mi})^2}{\ln \sigma_i}\right] \quad (13)$$

where the terms are as defined following Eq. 12.

If needed for special cases, volume, mass, or surface distributions are presented. The equations are

$$\frac{dM(r)}{dr} = \frac{4}{3}\pi r^3 \rho \frac{dN(r)}{dr} \quad (14)$$

$$\frac{dV(r)}{dr} = \frac{4}{3}\pi r^3 \frac{dN(r)}{dr} \quad (15)$$

$$\frac{dS(r)}{dr} = 4\pi r^2 \frac{dN(r)}{dr} \quad (16)$$

where $M(r)$ is the mass concentration (g cm^{-3}), $S(r)$ is the surface concentration ($\text{cm}^2 \text{cm}^{-3}$), $V(r)$ is the volume concentration ($\text{cm}^3 \text{cm}^{-3}$), and ρ is the bulk density of the particles (g cm^{-3}). As for most atmospheric aerosols, the surface distribution has its maximum around 0.1–1 μm , and the volume (mass) distribution has the giant particle-size range, $r > 1 \mu\text{m}$.

From the aerosol-size distributions, other aerosol-size parameters may be calculated: effective mean radius, r_e ; linear mean radius, \bar{r} ; and the rms radius, r_{rms} , also called the surface mean radius.

The linear mean radius, \bar{r} , is obtained by integrating over the sizes and dividing by the concentration:

$$\bar{r} = \frac{\int_0^\infty rn(r)dr}{\int_0^\infty n(r)dr} \approx \frac{1}{N} \sum_{i=0}^k r_i N_i \quad (17)$$

where the integral in the denominator represents the total number of particles. Eq. 17 shows that \bar{r} is a weighted mean value.

The rms radius, r_{rms} , is defined by

$$r_{\text{rms}} = \left[\frac{\int_0^\infty r^2 n(r)dr}{\int_0^\infty n(r)dr} \right]^{1/2} = \left(\frac{1}{N} \sum_{i=0}^k n_i r_i^2 \right)^{1/2} \quad (18)$$

In size distributions of atmospheric particles, r_{rms} is affected by the presence of very small and very large particles.

The effective radius, r_e , is

$$r_e = \frac{\int_0^\infty r^3 n(r) dr}{\int_0^\infty r^2 n(r) dr} = \frac{\sum_{i=0}^k r_i^3 n_i}{\sum_{i=0}^k r_i^2 n_i} \quad (19)$$

For a polydispersion having a relatively narrow size distribution, r_e is equivalent to the radius required for a monodispersion to exhibit the same total scattering characteristics.

The following list summarizes the concepts of radius and several others encountered in the literature:

\bar{r} —Linear mean radius.

r_{rms} —Root-mean-square radius or surface mean radius.

r_e —Effective mean radius.

r_m —Median radius. One-half of the particles have radii smaller than r_m .

r_c —Critical or mode radius. This is the radius of the size class containing the greatest population and corresponds to the maximum of $n(r)$. Some size distributions may be bimodal or trimodal.

It can be noted that such model distributions only reflect certain average values. Instantaneous distribution may vary, depending on local weather and wind, vertical mixing, horizontal transport, gas-to-particle conversion conditions, season, etc.

AEROSOL LOADING IN THE ATMOSPHERE

Aerosol particles transported into the atmosphere by convection and advection from different origins or formed by gas-to-particle conversion or photochemical processes have very complex and variable structure in their height and spatial distributions. Because of the large variability of aerosols over continents, our knowledge about aerosol-size distributions, number, and mass concentrations, especially as a function of altitude, still remains fractional and limited. Large variations exist in the data from different locations. Mass concentrations range from $2 \mu\text{g m}^{-3}$ in polar regions to $10 \mu\text{g m}^{-3}$ for the background, $30 \mu\text{g m}^{-3}$ for remote and rural; and from $170 \mu\text{g m}^{-3}$ in polluted urban environments to $10^5 \mu\text{g m}^{-3}$ in sandstorms.^[49] Variability of aerosol number concentration at different locations can be seen from measurements. Aerosol number concentrations observed in the city of Santiago de Chile ranged from 2×10^3 to $1.6 \times 10^4 \text{ cm}^{-3}$ and geometric mean diameters from 0.1 to 0.16 μm .^[50] Number concentrations in the range 10^4 to $4.5 \times 10^4 \text{ cm}^{-3}$ were measured in an urban area in Taipei for aerosols in the size range 0.017–0.886 μm in diameters.^[51] For a subtropical urban atmosphere (the City of Brisbane, Australia), the average particle concentration of 7400 cm^{-3} was measured (diameter of particles from 0.016 to 0.626 μm).^[52] The typical aerosol number

concentrations in urban area of the Negev desert (city of Beer-Sheva, Israel) ranged from 1.5×10^3 to $5 \times 10^3 \text{ cm}^{-3}$ for normal conditions and exceed $3 \times 10^4 \text{ cm}^{-3}$ for disturbed conditions (particle diameters from 0.3 to 9 μm). Continental, lower tropospheric aerosols have been measured in Southern Finland for different aerosol modes. Total particle concentrations between 300 and 3×10^3 particles cm^{-3} were observed.^[53]

Aerosol observations during clean marine conditions have been reported for southern hemisphere mid-latitude site and sites on the Antarctic continent (Cape Grim, Tasmania, and Macquarie Island, along coastal Antarctica), and during shipboard research programs. Marine number concentrations between 200 and 700 cm^{-3} for clean and Antarctic influenced periods, respectively, were obtained. Particle number concentrations in the Arctic haze varied between 50 and 600 particles cm^{-3} in the accumulation mode.

Because the aerosol in the atmosphere exhibits a considerable variation in location, height, time, and constitution, different conceptions exist for describing the aerosol loading in the atmosphere.

Models for the vertical variability of atmospheric aerosols are generally broken into a number of distinct layers. In each of these layers, a dominant physical mechanism determines the type, number density, and size distribution of particles. Generally accepted layer models consist of the following:^[41,54] a boundary layer that goes from 0 to 2–2.5 km, free tropospheric region running from 2.5 to 6–7 km, a stratospheric layer from 8 to 30 km, and layers above 30 km composed mainly of particles that are extraterrestrial in origin. Fig. 3 is an example of such a description.

The average thickness of the aerosol-mixing region is approximately 2–2.5 km. Within this region, one would expect the aerosol concentration to be influenced strongly by conditions at ground level. Consequently, aerosols in this region display the highest variability with meteorological condition, climate, etc.

In the tropospheric layer that extends from 2.5 to 7 km, an exponential decay of aerosol number density is observed. One often sees total number densities vary as follows:^[38]

$$N(z) = N(0) \exp\left(-\frac{z}{z_s}\right) \quad (20)$$

where the scale height, z_s , ranges from 1 to 1.4 km.

In the stratospheric regions of the atmosphere, an important component of aerosols is sulfuric acid. These aerosols are distributed by large volcanic eruptions that inject large ash particles into the stratosphere. Several months after eruption, sulfuric acid aerosols are formed by a gas-to-particle conversion mechanism, and complicated size distributions with a bimodal or multimodal structure are constructed. The aerosols above 30 km are considered to be primarily meteoric dust.

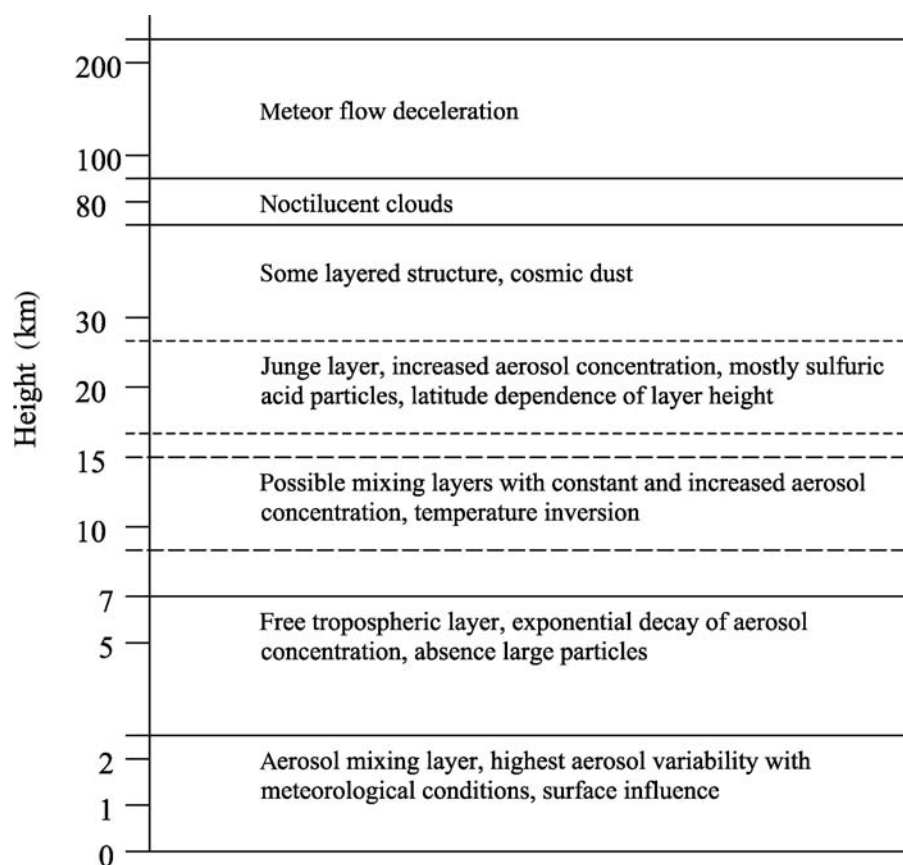


Fig. 3 Vertical variability of atmospheric aerosols.

The few data available show that in the atmospheric boundary layer (at 0–2 km) and in the lower stratosphere (at 9–14-km altitude), there may exist different layers (known as mixing layers) with constant and increased aerosol concentration.^[55,56] These layers can be caused by temperature inversions at ground level and by tropopause effect where the temperature gradient changes sign. Moreover, in the stratospheric region, there is some latitude dependence of aerosol layer height. The concentration maximum of stratospheric aerosols near the equator is located at 22–26-km elevation altitude, but about at 17–18-km height in the polar region.

To utilize the extensive measurements, a series of aerosol models for different environmental conditions and seasons were constructed.^[54] The models have been divided into four altitude regimes as described above. For the boundary layer, 10 models were introduced for several surface visibilities and for rural, urban, and maritime environments. For the tropospheric region, the spring–summer and fall–winter models were described. For the stratosphere, up to 30 km, in which background conditions, as well as moderate, high, and extreme volcanic conditions, for the two seasons were described. For the altitudes above 30 km, two models were described, one for background conditions and one for high aerosol concentrations.

Aerosols in the planetary boundary layer generally have a bimodal size distribution. A function which is the sum of two log-normal distributions has been chosen.^[28,37,57,58] In this distribution, the first broad peak occurs for particle diameters between 0.1 and 1 μm and another between 5 and 100 μm . The large mode consists mainly of soil-derived particles or sea-salt particles, whereas the accumulation mode consists of anthropogenic particles such as sulfates or nitrates. Maximal values of the individual modes change corresponding to different seasons or to different atmospheric conditions. For the urban model, the combustion and industrial aerosols to the general rural model in the lowest 2 km were added.^[44,59] The model that is frequently used for the prediction of aerosols in the marine atmosphere is the U.S. Navy Aerosol Model (NAM).^[60] However, NAM is intended only for application at a level of about 10 m above the air–sea interface. The Naval Oceanic Vertical Aerosol Model (NOVAM) extends the NAM predictions to higher altitudes.^[61] Such models are reasonably successful for the North Atlantic and Pacific Oceans, but not for the Mediterranean Sea.

The smoke aerosol in the boundary layer appears to be bimodal with an accumulation mode peaking at 0.12–0.16 μm and a coarse mode at roughly 3–10 μm .^[37] In contrast, a trimodal volume-size distribution ($r_m = 0.13\text{--}0.15, 0.5$

and $>1.5 \mu\text{m}$) was found in aircraft measurements.^[62] Ground-based sampling suggests a bimodal distribution with modes peaking at approximately 0.15 and $1.5 \mu\text{m}$, respectively. Other measurements of smoke also find an accumulation mode with mean volume radius in the 0.12–0.17 μm range.

In the troposphere, above the boundary layer, the rural aerosol model was used,^[44] but without the large-particle component of the size distribution because of the longer residence of aerosols above the boundary and the expected differential loss of the larger particles. This leaves the log-normal distribution with the small-particle component. Therefore, the tropospheric parameters are $N = 1$, $\bar{r} = 0.02\text{--}0.05 \mu\text{m}$ depending on relative humidity, and $\sigma = 0.35$.

Stratospheric aerosols appear to have a Junge-type size distribution or a unimodal one with a small-mode radius composed of sulfuric acid particles in the background condition although the size distribution varies depending on the altitude.

Friend^[63] has suggested that the stratospheric aerosol may best be described by a log-normal distribution function. Toon and Pollack^[64] determined that a best fit to the data is $\sigma = 2.0$ and $\bar{r} = 0.035 \mu\text{m}$. Herman et al.,^[54] on the other hand, a modified gamma function was chosen to represent the stratospheric aerosol although with different values of the parameters than those used earlier. In the Junge layer (at 18–22 km), the aerosol distribution

was fitted well with parameters $N = 10$, $\bar{r} = 0.0725 \mu\text{m}$, and $\sigma = 1.86$.^[65] In general, the total particle concentrations just above the tropopause (at about 15-km height) are between 10 and 100cm^{-3} , but decrease to 1cm^{-3} or less above about 20 km. The most comprehensive attempt at defining a stratospheric aerosol model has been that of satellite- and balloon-borne measurements, which included measurements of stratospheric aerosols in six size ranges.^[30,66]

The aerosols above 30 km are considered to be primarily meteoric dust, and the size distribution is taken as log-normal with parameters $N = 1.0$, $\bar{r} = 0.0725 \mu\text{m}$, and $\sigma = 0.5$.^[54] The upper atmosphere region is represented with a normal aerosol distribution with height and an extreme model. The extreme model is used to represent those occasions when this region is invaded with micrometeoritic dust or noctilucent clouds.

Toon and Pollack^[64] present a global average model for atmospheric aerosols. This model is intended to be a composite average based on available data at that moment.

On a global scale, aerosol climatology was summarized later by d'Almeida et al.^[17] A compilation of a large amount of data and tables of the dominant types of tropospheric aerosols as a function of the latitude, the longitude, and the season are shown. The new data for aerosol models were added from measurements.^[48] Table 1 summarizes the parameters used for some models of aerosol-size distributions at different altitudes.

Table 1 Parameters of model aerosol size distribution

Log-normal distribution	$N_1 (\text{cm}^{-3})$	$r_1 (\mu\text{m})$	$\log \sigma_1$	$N_2 (\text{cm}^{-3})$	$r_2 (\mu\text{m})$	$\log \sigma_2$	$N_3 (\text{cm}^{-3})$	$r_3 (\mu\text{m})$	$\log \sigma_3$
Background (tropospheric)	129	3.6×10^{-3}	0.645	6	0.127	0.253	0.635	0.259	0.425
Maritime	133	3.9×10^{-3}	0.657	67	0.133	0.21	3.06	0.29	0.396
Remote continental	3,200	0.01	0.161	2,900	0.58	0.217	0.3	0.9	0.38
Rural	6,650	7.39×10^{-3}	0.23	147	2.69×10^{-2}	0.58	1,990	0.042	0.27
Urban	99,300	6.5×10^{-3}	0.25	1,110	7.14×10^{-3}	0.67	36,400	0.025	0.34
Stratospheric	5	0.22	0.25						
	54	0.07	0.57						
Modified gamma distribution	a		α		β		b		
<i>Stratospheric</i>									
Background	324		1		1		18		
Fresh volcanic	341.33		1		0.5		8		
Aged volcanic	5461.33		1		0.5		16		
<i>Hazes</i>									
H (stratosphere)	4e5		2		1		20		
L (continental)	5e6		2		0.5		15.1		
M (marine)	5.3e4		1		0.5		8.9		
Free troposphere (power law)		$r < 0.045 \mu\text{m}$, $\beta = -1$; $0.045 < r < 5 \mu\text{m}$, $\beta = 2.6$; $5 < r < 30 \mu\text{m}$, $\beta = 4.6$							

Source: Adapted from Thomas & Duncan,^[41] Jaenicke,^[48] and Herman, LaRocca, et al.^[54]

Many experiments were carried out to test and modify the present aerosol models. The new data available show that the distribution varies dramatically with altitude, often within meters. Presently, no information about the parameterization of such vertical distributions exists.

AEROSOL MEASUREMENT TECHNIQUES

As discussed earlier, the aerosol-size distribution covers about five orders of magnitude in radius and approximately seven to eight orders of magnitude in the ordinate. Such extended size ranges can hardly be measured with one measurement system; thus, several have to be used simultaneously or different measurement principles must be used. Besides, the problem of the rather short residence time of the atmospheric aerosol exists as does its dependence on particle size. The short residence times produce large variations in the particle concentration.

The nature of the particles requires different physical principles to be used for measurements in situ or remote sensing. Examples of instruments for in situ measurements might be diffusion batteries, electrical mobility analyzers, optical particle counters (white light as well as lasers), cascade impactors, etc.^[67] These devices are designed to sample a relatively small volume of air at a sample point and to count and sort by size single aerosol particles in the sampled air. These instruments are capable of counting and sizing thousands of particles per second with size range from 0.003 to 40 μm . The fundamental characteristics of optical particle counter technology basically have remained unchanged for nearly 30 years. These devices sample a small volume of air, which is directed through a well-defined illuminated measurement region near the focus of a beam of light. Individual particles pass through the beam and scatter light in proportion to their size. The optical system collecting and focusing the scattered light to a photodetector is designed to provide a near-monotonic response function of photodetector voltage or current output vs. particle size. In situ measurements of aerosols by aircraft or balloon are restricted in time and space so that some remote-sensing techniques are required, and these should complement other techniques.

Instruments used in optical remote sensing of the atmosphere fall into two classifications, active and passive. In general, measurements obtained by active and passive sensors are complementary. Active instruments supply their own radiation source (laser, etc.), while passive instruments rely on naturally occurring radiation (direct and scattered solar radiation reaching the Earth's surface). All these methods have one thing in common: they measure appropriate optical properties of the aerosol particles and then infer their size distribution by an appropriate procedure from these optical properties.

Passive optical remote-sensing instruments include radiometers historically known as sun photometers.^[68,69]

They have a long history of operation for atmospheric studies including operation in space for a number of years.^[70] The active remote-sensing instruments are light detection and ranging (LIDAR) systems and polar nephelometers.^[19,71]

The passage of light through the atmosphere produces scattering and absorption, which is dependent on the atmospheric particulate properties. Particulates scatter light according to their size, shape, index of refraction, and the wavelength, intensity, and polarization of the incident light. The information encoded by the aerosol on the scattered light is a major source for aerosol analysis. The retrieved size distributions of course are based only on the optically active particles, which means particles in the range 0.1–10- μm radius.

Even in the clearest air, the sun's disk appears surrounded by a halo of bright sky called aureole, which can be several degrees in diameter. This is caused by the forward-scattering characteristics of the particles along the solar ray path. The measurements of aureole radiance analyzed in terms of scattering theory gives information on the average particle size. Because large particles (dimensions \geq the wavelength of the incident light) scatter very strongly in the forward direction because of diffraction by the particles, radiometer measurements made near the sun's disk are ideal for the extraction of aerosol particle-size information. The measurements, if performed at wavelengths in the visible region, are more sensitive to the smaller aerosol particle sizes because these particles contribute significantly more to the scattering cross section (outscattering in every direction) than to the small angle scattering.

Sun/sky scanning spectral radiometers, i.e., radiometers that measure the spectral sky radiance at known angular distances from the sun, are used successfully to retrieve total column, ambient spectral aerosol optical thickness, and volume-size distribution. The radiometers use sun measurements at difference wavelengths. The instruments invert sky measurements to obtain volume aerosol-size distribution and phase function. This technique requires precise aureole measurements near the solar disk and good stray-light rejection. Because the measurements are directional and represent conditions of the total column atmosphere, there are direct applications to satellite- and airborne observations as well as atmospheric processes.

The technology of ground-based atmospheric aerosol measurements using sun photometry has changed substantially because the first handheld analog instrument was introduced in the 1960s. Modern digital units of laboratory quality and field hardiness can collect data more accurately and quickly and are often interfaced with on-board processing.

Within the optically active region (where atmospheric aerosols scatter light appreciably), the size distribution is basic parameter, which can be inferred with a polar nephelometer.^[71] Typically, the terminology nephelometer encompasses those devices, which determine particle size

and/or concentration with scattered light. They are small-scale devices that incorporate their own light source (flash lamp with known spectral distribution). One group includes those instruments, which have a wide viewing angle set at a fixed position. A second group consists of polar nephelometers, which have a small field of view of scatter, but the angle at which the scatter is observed may be changed. These instruments typically measure the scatter from a small-volume element of the atmosphere.

Laser radar, more popularly known as LIDAR, is the most powerful techniques for active remote sensing of the atmosphere. Ground-based, airborne and satellite-borne LIDAR systems exist.^[19,20,72] LIDAR has the desirable feature of being able to measure aerosols at various heights in the atmosphere.

In LIDAR systems, a laser is used as a transmitter and the optical telescope is used in a receiver. As usually emplaced, the transmitter and receiver are at the same site so that the measuring signal of the LIDAR is mostly determined by the backscattering of the laser pulse by the aerosol particles and also by the extinction (absorption and scattering) of the pulse along the path between the LIDAR and the backscattering volume. Some of the scattered radiation is collected by the telescope and detected by a sensitive detector. 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 information about the presence, range, and concentration of aerosols. The spectral distribution of the backscattering is strongly influenced by the particle-size distribution so that by using LIDAR at more than one wavelength, one can obtain information about the aerosol-size distribution at different elevation heights. Fig. 4 shows results of aerosol distribution measurement at different altitudes with LIDAR.

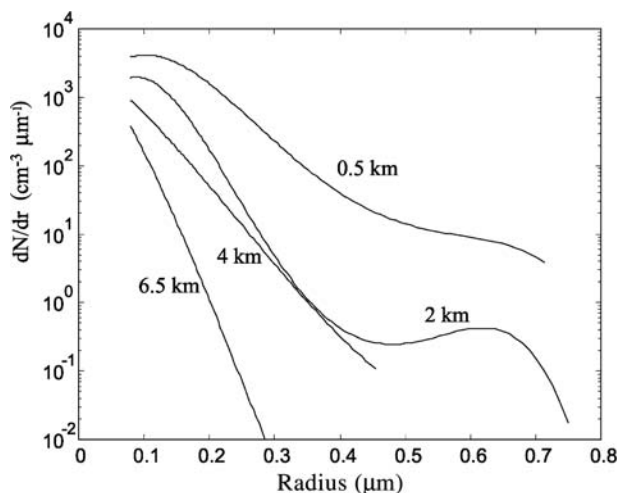


Fig. 4 Variation of the aerosol size distribution (accumulation mode) with altitude measured with LIDAR on July 26, 2001 in Beer-Sheva (Israel) at 01:30 A.M. (local time).

Both LIDAR and solar radiation measurements have the advantage that aerosols can be observed without disturbing their characteristics, and necessary information can be obtained by combining these measurements. Compared to airborne sampling methods, they are able to monitor continuously the vertical aerosol structure. In addition, by avoiding requirements for balloons or aircraft, they tend to be more economical and convenient in use. Passive methods can indeed be carried out with little effort, and the particle-size distribution can be deduced quite well if the wavelengths and scattering angles of such measurements are chosen adequately. However, the solar radiation method only derives average values of the aerosol-size distribution for the totality of aerosol particles within the path of the sunlight through the atmosphere.

For describing aerosol physical properties, extensive sets of both in situ and remote measurements are needed. In situ sampling or ground-based remote sensing are very informative, but on a relatively local scale. In order to obtain global coverage and a good sampling of the aerosol characteristics at climatic scales, it is necessary to use satellite data. Remote sensing of the Earth from satellites has grown to become a fundamental tool for climate and environmental studies. Present satellite-borne sensors use passive and active techniques and provide a capability to monitor aerosol loading on a global scale for long-time periods.

Knowledge of the parameters that determine the optical properties of atmospheric aerosols is essential not only for the determination of their climatic effects but also for the development of techniques for remote sensing of the atmosphere, or the necessary correction of atmospheric effects in satellite imagery, especially the effects of aerosols.

CONCLUSION

Our knowledge about atmospheric aerosols has grown considerably, but still remains fractional and limited. Many works introduce new questions about aerosol sources, transformation, transport, and sink processes as well as influence on performance of electrooptical systems that are still unresolved. Large uncertainties remain in the models for aerosol constituents.

In this entry, knowledge about aerosols, their sources and types, size distributions, spatial and temporal variability have been summarized. Some aerosol measurement techniques have been presented, and their limitations are discussed.

Difficulties in estimating the influence of aerosols on electrooptical system performance and their climatic impact arise from the high spatial and temporal variability of aerosol concentrations, and physical and chemical properties. In addition, the large size range makes the measurements of aerosols very difficult, time-consuming, and susceptible to error.

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