AEROSOLS

Contents

Climatology of Tropospheric Aerosols

J L Gras, CSIRO Atmospheric Research, Aspendale, Victoria, Australia

Introduction

Although the idea of a climatology of tropospheric aerosol appears relatively straightforward, practical implementation of a comprehensive global-scale climatology turns out to be very difficult; arguably, it is far from well established. Most problems stem from the inhomogeneous nature and spatial distribution of the aerosol, combined with sparse and relatively uncoordinated measurements. Despite this, much is known about the distribution of aerosol throughout the troposphere and how it varies seasonally. New remote-sensing technologies and analysis methodologies allow detailed observations and global mapping of some aerosol parameters. Determination of the global distribution of aerosol intrinsic properties, such as chemical composition, and the long-term trends of all aerosol properties remains a major challenge.

An aerosol is a stable suspension of a solid or liquid in a gas, in this case air. In practical terms, for the ambient atmosphere this includes particles from nanometers to tens and sometimes, hundreds of micrometers in diameter (around 10^{-9} to 10^{-4} m, or around five decades in size, and spanning an even greater range of concentrations). Constituent particles in the aerosol may be primary, that is, emitted as a particle, or secondary, being produced from gases in the atmosphere (by condensation or chemical reaction of certain species such as dimethyl sulfide (DMS), sulfur dioxide, and volatile organic compounds). They may include inorganic, organic, and biological entities such as spores, pollens, viruses, bacteria, waxes, and plant debris. Most particles include some water, although cloud elements are usually excluded from the description. Aerosol particles have both natural sources (e.g., wind-blown sea spray, dusts, and volcanic debris) and anthropogenic sources (such as smokes, fumes, and exhausts). In some cases, such as wind-blown dusts from poorly managed agricultural areas, this distinction may be unclear. In many regions, particularly Northern Hemisphere mid-latitudes, anthropogenic components now frequently dominate particle number and mass. In general, an aerosol comprises particles of mixed composition both internally (i.e., within one particle) and externally (between particles); this makes aerosol unlike a gas, which is the same wherever it is measured. Particle lifetimes are dependent on size and location in the atmosphere. Typically, lifetimes range from days near the surface to months in the upper troposphere. There are many sources of particles, ranging from point sources including volcanoes, fires, and industrial plants, through to cities, clouds, entire regions, oceans, and even the atmosphere itself. In general, the spatial and temporal distribution of aerosol is notably inhomogeneous. Substantial changes in aerosol properties can occur over distances of only a few kilometers horizontally and much less vertically. The atmospheric aerosol is extremely dynamic, evolving and changing properties from the point of production until its ultimate removal by sedimentation or wet processes. During its lifetime, a particle may amalgamate with other particles, exchange material through gas-phase reactions, and most likely pass through a number of cloud cycles where it is incorporated into cloud droplets, reappearing in a modified form when the droplets evaporate.

The troposphere is that part of the atmosphere lying between the surface and the stratosphere. The name derives from the Greek *tropos* for 'turning', and this part of the atmosphere is characterized by air motion and mixing. Near the poles the troposphere reaches to about 8 km altitude and in the tropics to around 18 km

altitude. The concept of a well-mixed troposphere is somewhat misleading though, since many of the important tropospheric aerosol features are associated with discrete layers in the atmosphere and the transport of aerosol in layers, often in large quantities.

Tropospheric aerosols impact on global climate, the atmospheric environment, and even human health. Aerosols are a significant component in the global radiation balance, scattering and absorbing solar radiation and changing the properties of clouds. Their climatic effect is of similar magnitude to the greenhouse effect of carbon dioxide, but the net effect of particles is a cooling. Aerosols are important in the precipitation process: all cloud drops form on aerosol particles, known as cloud condensation nuclei, and ice nuclei play a similar role in ice clouds. Aerosols interact with reactive trace gases (heterogenous processes) and are implicated in biogeochemical cycling in the atmosphere, including transport of trace nutrients such as iron to the world's oceans and of wind-generated sea-salt to the land, and playing roles in the sulfur, nitrogen, and carbon cycles. At the urban and rural levels they are the dominant cause of reduction of visibility and add to acidification and transport significant quantities of chemicals, such as minerals, sulfate, nitrate, and carbonaceous material. Some aerosol components are toxic, others are carcinogenic. Epidemiological studies have shown significant correlation between various aerosol properties, including certain mass fractions, and human mortality and morbidity. Bioaerosol (molds, spores, pollens, dust mite feces, bacteria and viruses, and possibly fragments) are implicated in diseases including asthma.

How do aerosol properties vary across the globe and as a function of time, location, and altitude in the atmosphere? These factors are usually studied through development of a 'climatology'. Strictly speaking, a climatology is a statistical description of a defined system, composed of various elements. These elements exhibit spatial and temporal variations to which the climate concept applies. For the global climate, for example, this would include temperature and rainfall. The most fundamental elements for the tropospheric aerosol system are the size-dependent concentration and aerosol intrinsic properties; the latter properties are independent of concentration and include chemical composition, refractive index, and shape. Other elements or means of descriptions of the aerosol are also possible, such as light extinction coefficient and integral mass.

Unfortunately, no single definition of an aerosol climatology is universally accepted. The definition above, which describes an 'observational' aerosol climatology, requires comprehensive measurements of the aerosol properties or elements. For tropospheric aerosol on the global scale, no truly comprehensive 'observational' climatology exists for any intrinsic aerosol property and it may be unachievable in the foreseeable future. None exists even for properties as apparently fundamental as total mass or number. Sufficient reliable data exist for certain aerosol properties at selected sites or in certain regions for the development of limited 'observational' aerosol climatologies. Very few records of any aerosol parameter are multidecadal, and establishing these records remains an important task for international science.

Contrasting with this is a 'fully modeled' aerosol climatology. A comprehensive aerosol model should be able to distribute precursor material between new particles and existing particles and generate a full description of the distribution of mass and intrinsic aerosol properties, as a function of size, from around 10^{-9} m to 10^{-4} m diameter, giving the three-dimensional spatial distribution of these properties over time. Aerosol dynamical models and chemical transport models exist but none currently has all these capabilities.

An intermediate approach, in effect generating a 'hybrid' aerosol climatology, has also been used. It comprises a model, based on an external mixture of aerosols from a set of generalized sources (e.g., biomass burning emissions, mineral dust, and sulfate), each with a given emissions inventory. Each component contributes its own generic properties, such as descriptions of the size distribution and optical properties, derived from a variety of *in situ* or remotely sensed measurements.

Data quality is central to all climatologies. Measurements with unknown accuracy or poorly specified conditions of measurement are ultimately of little or no value.

In addition to the spatial or temporal variation of aerosol properties, other useful descriptions include the amount of material or mass flux emitted by various sources. Major aerosol flux contributors include mineral aerosol, sea-salt, smoke from biomass burning, anthropogenic emissions (as sulfate and carbonaceous material), and secondary aerosol sources (including natural precursor gases). **Table 1** gives typical estimates of the annual flux of various sources from the 1970s and 1990s, although the relative importance for both environmental and climate impacts is not simply related to these mass fluxes. Particles in the range 50 nm to 1 μm, for example, interact strongly with solar radiation and have the biggest effect on cloud properties. For mineral and sea-salt aerosol, massive particles with very short lifetimes complicate the definition of the flux. Flux estimates

Table 1   Estimated strength of tropospheric aerosol sources

	Early 1970s (Tg/y^{-1})	1990s (Tg/y^{-1})
Natural		
Mineral	10–500	1000–3000[a]
Sea-salt	200–1000	1000–3000[a]
Volcanic	3–150	15–90
Biogenic		50
Gas to particle	300–2000	200–1300
Extraterrestrial	0.1–50	10
Anthropogenic		
Industry, fossil fuel, carbon, sulfate, nitrate, organics	100–400	300
Biomass burning	3–150	100–450

[a]Depends strongly on upper size limit.

have been evolving since the early 1970s as information on aerosol emission rates and the extent of sources improve.

Spatial and Temporal Distribution of Particle Properties

Convection and advection control the transport of atmospheric particles. Most particle sources are near the surface and concentrations generally decrease by about two-thirds for each 1 km altitude to about 5 km over the continents and 2 km over oceans. Above this, particle mixing ratios are relatively constant with altitude to near the tropopause. Concentrations of larger particles increase again in the lower stratosphere and total particle concentrations decline. Tropospheric air enters the stratosphere at low latitudes, returning to the troposphere, via tropopause perturbations (folding), polewards of about 30° N and 30° S. This return air carries some stratospheric particles into the upper troposphere and sedimentation provides a flux of larger particles into the upper troposphere at all latitudes. Stratospheric particles can be identified to about 1–2 km below the tropopause. Extraterrestrial particle sources are relatively minor.

Very small particles have an altitude profile more indicative of a distributed source in the upper free troposphere, and freshly nucleated particles have been observed in the upper troposphere when the integrated aerosol surface area becomes small. Both features are consistent with a relatively homogeneous tropospheric aerosol away from major sources, the so-called tropospheric background. Adding to this are remnant aerosol from various sources and the many major enhancements in tropospheric aerosol concentration associated with layer transport, particularly of mineral dusts, smoke plumes, and anthropogenic material.

Satellite-based remote sensing methodologies are very effective for geographically mapping various measures of aerosol 'amount', but the determination of intrinsic size-dependent properties such as composition and particle shape require *in situ* determination. Consequently, there are far fewer data available for these properties, and the intercomparability of sampling protocols becomes very important. Geographic distributions of some species, such as sulfate, have received considerable attention and, while no really comprehensive global climatology of intrinsic properties currently exists, there are a growing number of national networks addressing the issue of aerosol composition, including sulfate, nitrate, and carbonaceous components.

In the following sections the troposphere is considered in two altitudes ranges, greater than and less than 5 km, with emphasis placed on aerosol mass and number concentration.

Lower Troposphere

The altitude range from the surface to around 5 km includes the most intensively studied region of the troposphere through surface networks, mountain top observatories, and aircraft measurements. This part of the atmosphere is complex, involving the diurnally varying planetary boundary layer, nocturnal inversions, the marine boundary layer, clouds, and precipitation. Surface measurements are not always good indicators of the free troposphere or even the boundary layer. Measurement networks are also not uniformly distributed spatially and have widely different measurement parameters and methodologies. Remote sensing of column-integrated aerosol properties from satellites using spectrally resolved, scattered solar radiation also primarily senses lower-tropospheric aerosol properties, with roughly equal contributions from the boundary layer and free troposphere.

From late in the 19th century, systematic recording of meteorological observations by ships' crews included aerosol phenomena (particularly haze). Compiled in the 1930s, these represent one of the most extensive aerosol climatologies of the period and several decades following. The geographic distribution and seasonal variation from more than 50 years' observations show most features evident in today's satellite observations. These include Saharan dust plumes over the Atlantic ocean, the eastern Atlantic European plume, a North American summer plume in the North Atlantic, spring dust over the north-western Pacific, a summer plume over the Arabian sea, and dry-season plumes in north-western Australia and Indonesia. Currently, satellite-based remote measurements

afford the most extensive geographic mapping of those components of the aerosol that interact appreciably with visible or near-visible wavelengths, although surface-based measurements of column-integrated optical properties afford higher precision and the stability needed to assess subtle long-term changes.

Examples of dust and smoke distributions are shown in **Figure 1** in a monthly average 'UV absorption aerosol index' from the Total Ozone Mapping Spectrometer (TOMS), for October 1997. This demonstrates the extensive smoke plumes from fires in Indonesia and Brazil as well as some dust over Africa. **Figure 2** shows extensive mineral aerosol from Africa extending over the Atlantic Ocean during April 2000, as well as other dust including some over eastern China. Individual daily mapping, such as for the 1997 Indonesian fires, is also possible, as shown in **Figure 3**. High-resolution spectrally resolved measurements of scattered solar radiation, for example, from NOAA's AVHRR (Advanced Very High Resolution Radiometer) satellites, also allow detailed mapping of equivalent column-integrated aerosol light extinction (optical depth) over ocean regions. The major aerosol plumes can be seen clearly in maps of the distribution of optical depth, such as **Figure 4**.

Particle Mass

As indicated by remote sensing and *in situ* measurements, aerosol mass concentration varies strongly with location and time, depending on the proximity to sources and the effectiveness of dispersal and removal mechanisms. Particle total mass concentrations range from around $100 \, \text{ng m}^{-3}$ in the upper troposphere to greater than several hundred $\mu\text{g m}^{-3}$ in many large
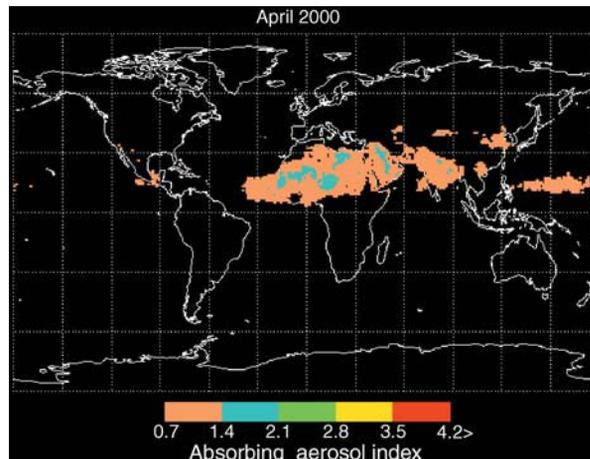

Figure 2 TOMS UV-absorbing aerosol index for April 2000. (Image courtesy NASA.)

cities in developing countries. Over ocean regions, concentrations vary with wind speed and proximity to continental sources, but values down to $10 \, \mu\text{g m}^{-3}$ are observed. Total aerosol mass is measured less extensively than mass in aerosol size ranges associated with health or climate effects. In some jurisdictions the mass loading of particles with aerodynamic diameter $D_a < 10 \, \mu\text{m}$ (PM10) is regulated. Concentrations of PM10 in many cities in developing regions exceed $100 \, \mu\text{g m}^{-3}$ (24-hour average), but in developed countries are typically less than $50 \, \mu\text{g m}^{-3}$. In more remote continental regions, such as around the American Rocky Mountains, PM10 concentrations are usually less than $10 \, \mu\text{g m}^{-3}$. A seasonal variation in mass loading is common and is frequently dominated by local factors, including the seasonal pattern of
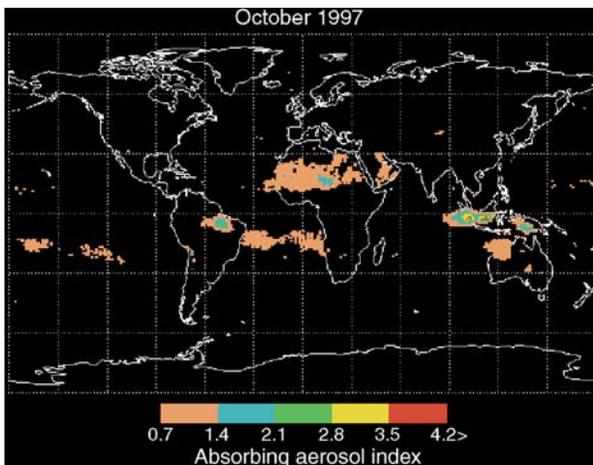

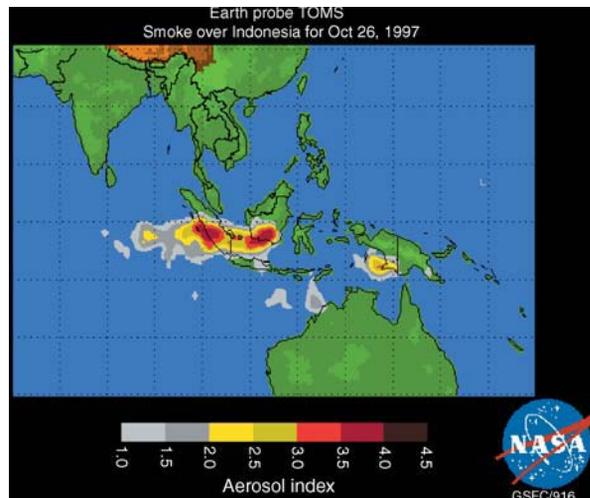**Figure 1** TOMS UV-absorbing aerosol index for October 1997. (Image courtesy NASA.)

Figure 3 TOMS UV-absorbing aerosol index for 26 October 1997, showing Indonesian fire plumes. (Image courtesy NASA.)
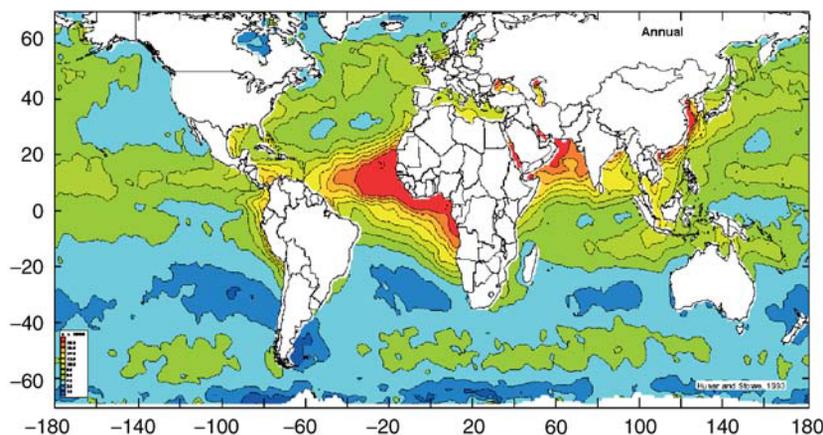
Figure 4 NOAA AVHRR equivalent aerosol optical thickness, annual composite 1989–1991. (Image courtesy NOAA.)

emissions, for example, anthropogenic sources, biomass burning, mineral dusts, changes in convection, dispersion (wind patterns and thermal structure of the atmosphere), and removal (precipitation). Other size fractions such as PM2.5 (mass of particles with $D_a < 2.5 \, \mu m$) and particles of submicrometer diameter are increasingly studied for health and climate research, but observations currently have much more limited spatial and temporal cover. Historical long-term mass data are sparse, requiring the use of proxy records. Observations of visual haze over the eastern United States from the 1950s to the 1980s, for example, show a summertime increase in light-extinction coefficient but a reduction during winter over the 1970s and 1980s. Primarily these reflect mass changes in the 0.1 to 1 μm diameter range.

Although it may be feasible to assemble a meaningful global climatology from the diverse urban and regional network measurements of aerosol mass, none exists at present. Different regions and subregions have individual patterns and many developing networks will in time allow assessment of trends.

Contributions to the mass from different sources also vary uniquely. Oceanic sea-salt aerosol production, for example, depends strongly on wind speed. Within the marine boundary layer, sea-salt typically dominates aerosol mass, although mineral, smoke, and anthropogenic aerosol can be regionally important, as shown in **Figures 1–4**. Representative sea-salt aerosol mass concentrations lie in the range 1–50 $\mu g \, m^{-3}$. Concentrations can change rapidly and strongly with synoptic events, and seasonal variation is typical with a local winter concentration maximum. Seasonal amplitude ranges vary from less than about 1.5:1 (Southern Hemisphere mid-latitudes) to greater than 8:1 (North Atlantic). Surface observations are the primary source of information on sea-salt aerosol intrinsic properties. Few, if any,

observational studies have addressed the long-term change in sea-salt aerosol.

Smoke aerosol loading is also highly variable, reflecting local seasonal patterns of biomass burning. Prescribed and uncontrolled fires occur in all vegetated ecosystems around the globe, although statistics on burning are relatively poor. Major fire regions include Africa (10°–20° S) (August–October maximum); Brazil (July–September maximum); sporadically through Indonesia (dry season, typically June–October) and northern Australia (May–October maximum); and northern boreal forests, e.g., Siberia/China (spring maximum, also autumn). Concentrations depend on the proximity to sources, and effectiveness of dispersal, with mass loading exceeding 1 $mg \, m^{-3}$ near large fires. Residual black carbon mass loading less than 5 $ng \, m^{-3}$ (comprising roughly 10% of smoke mass) is frequently observed in well-scavenged marine air. Smoke aerosol transported over long distances is often present in well-defined layers up to around 5 km. Few long-term measurements of biomass burning aerosol exist and historic records are sparse. Emissions have been estimated to have increased 2-fold to 3-fold over the past century.

Mineral aerosol is also highly regional, and seasonally dependent. Satellite remote sensing affords daily measurement and mapping of the spread of aerosol from individual events. Longer-term records are now compiled from satellite analyses (e.g., AVHRR, TOMS) and some historic context can be inferred from ships' observations from the late 19th century. Quantitative measurement of aerosol from *in situ* surface sampling has been the primary source of information on composition, mass loading, lifetime, and variability. Studies show substantial variability even during a single day, with concentrations changing up to several orders of magnitude. The spatial distribution of mineral aerosol is governed primarily

by the extent of source regions and climatological wind patterns. Thus, major dust areas include the Atlantic Ocean downwind of the Sahara, the western Pacific downwind of China, and the Arabian Sea. Northern Hemisphere mineral aerosol fluxes are an order of magnitude greater than those in the Southern Hemisphere owing to the location of active dust source regions.

As with smoke aerosol, the long-range transport of mineral aerosol frequently occurs in well-defined layers in the lower 5 km of the atmosphere. Mineral aerosol concentrations are strongly dependent on season and distance from the source regions. Aerosol from the dust areas in East Asia is observed most frequently in spring (March–April–May). Lowest concentrations occur in summer and winter when dust production and transport mechanisms are unfavorable. In spring, boundary-layer concentration of mineral aerosol ranges from several hundred $\mu g\,m^{-3}$ near the Chinese coast to values up to about $9\,\mu g\,m^{-3}$ at Mauna Loa (on Hawaii). The central Pacific has a relatively small mineral aerosol loading, whereas moderate concentrations of dust from Australian deserts are observed over the south-western Pacific in the Austral spring and summer. Saharan and sub-Saharan mineral aerosol are transported across the Atlantic, with a maximum occurrence in June–August, and observed in both North and South America. Daily average concentrations of mineral aerosol in excess of $100\,\mu g\,m^{-3}$ have been recorded in Miami and monthly mean concentrations in the dust season regularly exceed $20\,\mu g\,m^{-3}$ in Miami and at Barbados. Concentrations in the dust plumes over the Atlantic may be as high as $400\,\mu g\,m^{-3}$.

Particle Number

The integral number concentration was one of the first aerosol properties explored in the late 19th century, with the development by John Aitken of a portable 'dust counter'. Although essentially the same type of measurement is used today, systematic long-term records are a relatively recent phenomenon. Total particle number concentrations vary from less than $10\,cm^{-3}$ over the Antarctic Plateau in winter, and infrequently at mid-latitudes in polar outbreaks, to around $10^5\,cm^{-3}$ in large cities. Concentrations decrease with altitude, to around $100–200\,cm^{-3}$ in the upper troposphere. Over clean southern ocean regions, concentrations are typically several hundred cm^{-3}, varying seasonally, with a summer maximum. The amplitude of the seasonal cycle is latitude dependent. At southern mid-latitudes, monthly median concentrations of $100–600\,cm^{-3}$ are observed, and

a similar range is found on the Antarctic coast, around $50–600\,cm^{-3}$ at Mawson, for example, but the range is much smaller at the South Pole with a seasonal cycle around $10–150\,cm^{-3}$. A smaller range is also found in the Tropics; for example, at Samoa the monthly mean particle number ranges from about 200 to $300\,cm^{-3}$. Photoxidation of DMS, emitted by marine phytoplankton, is considered the major natural source of the climatically important submicrometer aerosol particles over the global oceans.

In north-eastern Atlantic air (e.g., Mace Head, Ireland) the number concentration in clean air ranges from about 100 to $700\,cm^{-3}$ with no seasonal cycle, although a summer peak is found in methyl sulfonate, an oxidation product of DMS, the principal natural precursor. Long-range transport from the United States may inhibit a winter number concentration minimum. Closer to major continental source regions, for example, over the north-western Atlantic and north Pacific, concentrations are more typically $4000–6000\,cm^{-3}$.

The most widely quoted particle concentrations for continental sites date to the first half of the 20th century and may be geographically biased; concentration values around $10^4\,cm^{-3}$ are typical of areas away from population centers. In truly clean continental areas (for example, across Australia), particle concentrations are more typically around $700\,cm^{-3}$ in the boundary layer and around $200\,cm^{-3}$ above the boundary layer.

Arctic aerosol and its associated haze have been studied intensively. Typically a phenomenon of the lowest 3 km, occasionally up to 5 km, haze events result from the long-range transport of anthropogenic aerosol from Eurasia and North America. Particle numbers (monthly mean) at the surface are typically $50–400\,cm^{-3}$, occasionally up to $800\,cm^{-3}$, with spring and summer maxima, whereas light scattering (and absorption) peak in spring and winter. Recent analyses for Barrow, Alaska, indicate a significant positive trend in particle number (around 8% per year since 1976) and a decreasing trend in spring light-scattering coefficients. Natural sulfur sources are important in summer.

Above the boundary layer, number concentrations are lower. For example, in Hawaii (Mauna Loa, 3.4 km) monthly mean concentration is typically $260\,cm^{-3}$; there is a weak seasonal cycle (concentration range around 1.5:1) with a September maximum. A trend of $+1.7\%$ per year was seen in the 1970s and 1980s. Similarly, in free tropospheric air over the European Alps, (JungFrauJoch, 3.6 km), typical summer concentrations are around $800\,cm^{-3}$ and about half of this concentration in winter. Across eastern Europe, average particle concentrations of

$100–500\,cm^{-3}$ are typical at 3 km (mid 1900s). Over the continental United States during the 1980s, at altitudes of 2.5–5 km, number concentrations were typically $1800\,cm^{-3}$ in summer, and $700\,cm^{-3}$ in winter, with an overall negative trend around 5% per year.

Longer-term measurements usually show interannual variations in concentration. In Southern Hemisphere oceanic regions there is evidence of variations with decadal time scales, although records are relatively short (**Figure** 5). At Cape Grim (41° S 145° E) annual-average particle concentration increased by an average $1.4 \pm 0.3\%$ per year from 1976 to 1998. Historic records comprising full seasonal data at regionally representative sites do not exist. Proxy data, specifically small-ion conductivity, suggest that number concentrations in the northern Atlantic may have approximately doubled in the 60 years to 1970. Similar (sparse) records from the south Pacific suggest little or no change in this period.

Upper Troposphere (>5 km)

Compared to the lower 5 km, surprisingly few systematic long-term studies have addressed the upper-tropospheric aerosol. Methodologies that access this region include satellite-based light-extinction measurement, *in situ* balloon-borne samplers, high-altitude aircraft, and various lidar (laser radar) systems. The widest geographic cover is from satellites such as SAGE-2 (Stratospheric Aerosol and Gas Experiment), looking at long-path light extinction (limb-scanning) during sunrise and sunset. These provide information on extinction coefficients at $1\,\mu m$ and submicrometer wavelengths and indirect data on particle sizes. Balloon-borne measurements over more than two decades have produced consistent *in situ* particle information in both hemispheres.
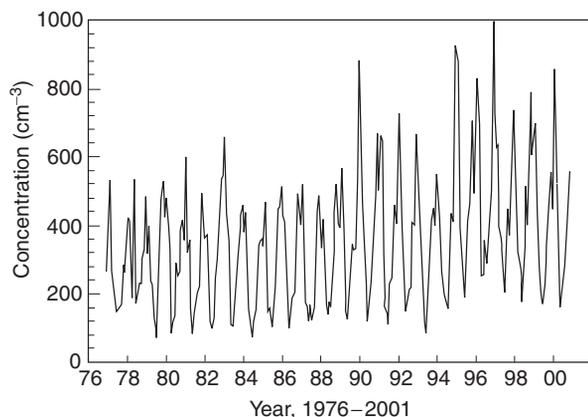

Figure 5 Monthly median concentration of particles in clean Southern Ocean air at Cape Grim Tasmania, 1976–2001.

Limb-scanning satellite data (from 1984 to 1991) give an extensive depiction of the upper-tropospheric aerosol via the light extinction coefficient. The data have also been used to infer aerosol mass concentration and effective size in the submicrometer size range. These extinction data and *in situ* determinations show a relatively uniform aerosol between about 5 km altitude to near the tropopause. In both hemispheres (20°–40°) a seasonal cycle is seen in light extinction, with a maximum in the spring–summer period when particle size is also maximum. In the Northern Hemisphere the spring period is characterized by a maximum in mineral aerosol production and transport. At mid-latitudes in Australia, balloon-borne measurements show marked increases in particle concentrations in the entire troposphere during spring, probably linked to long-range transport of biomass burning emissions. Enhanced concentrations are also found over the south-western Pacific during this season. At southern mid-latitudes in spring, mixing ratios of submicrometer aerosol are comparable to those at northern mid-latitudes in summer, representing an enhancement of more than an order of magnitude over other periods.

In general, significantly more light extinction and mass are observed in the Northern Hemisphere, particularly at mid-latitudes and polewards, and the seasonal cycle in extinction is about five times greater in the Northern Hemisphere. AVHRR data indicate around 50% more column light extinction in the Northern Hemisphere than the Southern Hemisphere. Multidecadal measurements of particle size and number at northern mid-latitudes, made from balloon-borne samplers, also show a spring–summer peak in an annual cycle of optically active particles, where concentrations vary by close to an order of magnitude. For total particle number, the seasonal variation is smaller, around a factor of 3, with a summer peak. All of these observations include quiescent and enhanced conditions; *in situ* aerosol backscatter measurements at mid-latitudes from only quiescent periods indicate relatively little interhemispheric difference.

See also

Aerosols: Observations and Measurements; Physics and Chemistry of Aerosols; Role in Cloud Physics; Role in Radiative Transfer. **Biogeochemical Cycles:** Sulfur Cycle. **Deposition**. **Dust**. **Global Change:** Biospheric Impacts and Feedbacks. **Lidar:** Backscatter. **Observation Platforms:** Balloons. **Observations for Chemistry (*In Situ*):** Particles. **Satellite Remote Sensing:** Aerosol Measurements. **Soot**. **Tropospheric Chemistry and Composition:** Aerosols/Particles. **Volcanoes:** Role in Climate.

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Observations and Measurements

P H McMurry, University of Minnesota, Minneapolis, MN, USA

Introduction

Atmospheric aerosols consist of a suspension of solid or liquid particles in air. These particles range from molecular clusters less than one nanometer in diameter to pollens, wind-blown soil dust and sea salt of 10 μm or larger. **Figure 1** shows an idealized atmospheric aerosol size distribution with the characteristic modes in which particles tend to be found. Homogeneous nucleation leads to the clustering of molecules that grow to particles several nanometers in diameter at the small end of the spectrum. Aitken or nuclei mode particles in the 10 to 100 nm diameter range often contain 'primary' particles that are directly emitted into the atmosphere by combustion processes. Accumulation mode particles contain most of the submicron mass. Most of this mass is typically 'secondary' in origin (i.e., formed in the atmosphere by chemical transformations). Sulfates, nitrates, and organics typically comprise most of the accumulation mode mass, and this mass can accumulate in different submodes, depending on the chemical mechanisms by which they are formed. Coarse-mode particles tend to be produced by mechanical processes.

Figures 2A–C show photographs of atmospheric particles obtained by electron microscopy. The chain-agglomerate soot particle in **Figure 2A** is typical of particles emitted by diesel engines or other combustion sources. **Figures 2B** and **C** illustrate particles of mixed composition. The salt in **Figure 2B** was prob-

ably emitted directly into the atmosphere as sea spray, while the sulfate crystals attached to it may have been produced in the atmosphere by chemical transformations. The rutile in **Figure 2C** probably originated as wind-blown dust and may have accumulated the sulfate coating as a result of atmospheric processing. The variabilities in shape and composition illustrated by the photographs in **Figure 2A–C** introduce ambiguities in aerosol measurements since the responses of particle measuring instruments depend on such properties. In addition, particles often contain volatile or
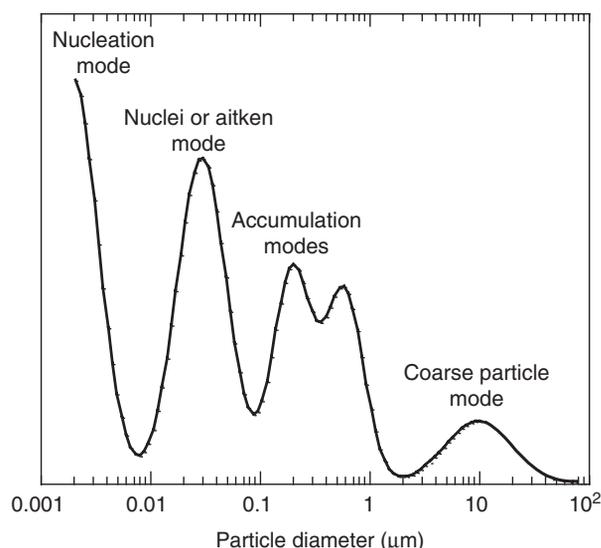

Figure 1 Idealized atmospheric aerosol size distribution. Particles tend to be found in modes that reflect the different processes by which they are formed in the atmosphere by chemical transformations or are emitted directly into the atmosphere from sources.