

Aerosol in the atmosphere

Our atmosphere consists not only of gases but also of particulate matter. The particulate matter can be either liquid droplets or solid particles, and are collectively called aerosol particles.

The important role of aerosol particles in the atmosphere early is now well established that the formation of cloud droplets also needs to have these particles, and they are called *cloud condensation nuclei* (CCN). Evidently, these particles should be viewed not just as contaminants but as an important component of the atmospheric physicochemical system.

Similar to the condensation of water droplets, the formation of ice in clouds also needs the presence of certain aerosol particles – called ice nuclei (IN) – unless the temperature is close to -40°C or colder. Both cloud condensation nuclei and ice nuclei are subsets of aerosol particles in the atmosphere.

6.1 Aerosol size categories

One of the important characteristics of aerosol particles, like clouds, is their size. Junge divided aerosol particles into three size categories:

- (1) *Aitken particles* – particles with radius $r < 0.1 \mu\text{m}$.
- (2) *Large particles* – particles with radius $0.1 \leq r \leq 1.0 \mu\text{m}$.
- (3) *Giant particles* – particles with radius $r > 1.0 \mu\text{m}$.

When aerosol particles serve as nuclei to nucleate cloud particles, they are called Aitken nuclei, large nuclei or giant nuclei according to their size. But we must keep in mind that not all aerosol particles serve as nuclei for cloud formation; hence these nuclei are only subsets of the particles of their respective size category.

Thus Whitby (1978) proposed the following naming scheme where the “size” here represents the modal size of a distribution:

- (1) *Nuclei mode* – particles with $d < 0.1 \mu\text{m}$, where d is the particle diameter; in some literature, the nuclei mode is further subdivided into nucleation mode ($d < 0.03 \mu\text{m}$) and Aitken mode ($0.03 \leq d \leq 0.1 \mu\text{m}$).
- (2) *Accumulation mode* – particles with $0.1 \leq d \leq 2.5 \mu\text{m}$.
- (3) *Coarse mode* – particles with $d > 2.5 \mu\text{m}$.

The term “diameter” here needs some explanation. Aerosol particles have a wide variety of shapes: some are approximately spherical but many are highly irregular (such as flakes or curled up fibers) But since aerosol particles are floating in air, it is useful to define an “aerodynamic diameter” to represent the size of particles having different shapes and densities. The *aerodynamic diameter* is the diameter of a spherical particle having density of 1gm^{-3} that has the same terminal settling velocity in the gas as the particle of interest. In addition to the above terminology, particles with $d < 1.0 \mu\text{m}$ are also called fine particles (FP) and those with $d < 0.1 \mu\text{m}$ are ultrafine particles (UFP).

6.2 Aerosol concentration

The concentration of aerosol particles is usually expressed in terms of either number concentration (in “number per unit volume”, for example, cm^{-3}) or mass concentration (for example, $\mu\text{g m}^{-3}$). The choice of unit usually depends on the specific application and the instrument used for the measurements. Some instruments can make number counts and some measure only the mass concentration.

Aerosol number concentration near the Earth’s surface varies widely with location. In terms of number concentration, it varies from a few hundred cm^{-3} in the air over the ocean surface to well over 10^6 cm^{-3} in highly polluted city air. In towns and smaller cities, the number concentrations are typically from a few 10^3 to 10^4 cm^{-3} . Over the mountains, the concentration is typically 10^2 to 10^3 cm^{-3} , and usually decreases with height in the mountains. It is our common experience that the air over oceans and higher up in mountains is “fresher” than that in a populated city, and the number concentration of aerosol particles is likely the main reason for this.

In terms of mass concentration, oceanic air is typically around $1 \mu\text{g m}^{-3}$ or lower. In an urban area, the mass concentration is around a few to $\sim 10 \mu\text{g m}^{-3}$. In dust storm events, the magnitude can be as high as several hundred $\mu\text{g m}^{-3}$.

6.3 Variation of aerosol concentration with height

Aerosol concentration varies not only from place to place but also with height. Although aerosol particles have sources in the atmosphere, their major source is the Earth’s surface. Hence it is not surprising that the total concentration of aerosol particles in the lower atmosphere usually has a maximum near the surface and decreases with height. But the decrease is not monotonic because there are also sources and sinks of aerosol in the atmosphere. Figs. 6.1 and 6.2 show two examples of aerosol vertical distributions. It is important to note that vertical profiles of aerosol concentration are often plotted for different size categories and hence one must read the chart carefully. Both figures here refer to Aitken particles.

Fig. 6.1 shows that the Aitken particle concentration decreases nearly exponentially with height from the surface up to about 6 km. This profile usually implies that

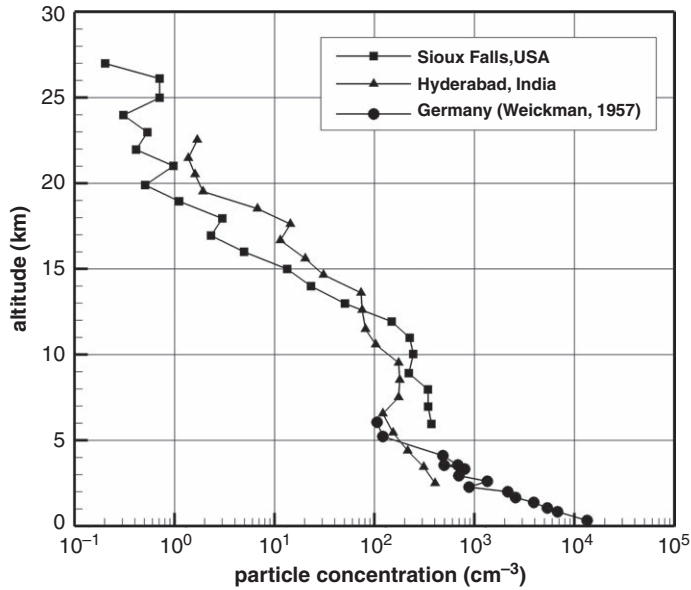


Fig 1

the surface is the source of the particles and the concentration distribution is exponential decrease ends. From $z \sim 6$ km up to about 10 km the concentration stays roughly constant at $\sim 50\text{--}500\text{ cm}^{-3}$ with 200 cm^{-3} the most frequent. Above 10 km, the Aitken particle concentration decreases again with height.

The quasi-constant phenomenon in the 6–10 km layer has been thought to indicate the existence of a “global background aerosol” of the atmosphere, which, if true, would represent an intrinsic characteristic aerosol state of the Earth’s atmosphere. Recent measurements, however, have found that the aerosol concentrations change rapidly in response to long-range transport and local sources and sinks, and hence the concept of a background aerosol is now being debated.

The case of large particles looks quite different. From the surface up to the tropopause level the concentration decreases generally, but it begins to increase above the tropopause and reaches a maximum somewhere in the middle stratosphere, the precise level depending on location and season. This stratospheric maximum has been called the Junge layer or stratospheric aerosol layer. More recent measurements confirm the worldwide existence of the Junge layer, which is located in the 20–40 km level.

Giant particles are basically confined to the lower troposphere due to their size. One notable example of giant particles is sea salt particles over the ocean. Sea salt concentration has a maximum in the lowest few hundred meters and then decreases quasi-exponentially with height. Sea salt particles are generated by the evaporation of sea spray droplets.

6.4 Aerosol size distributions

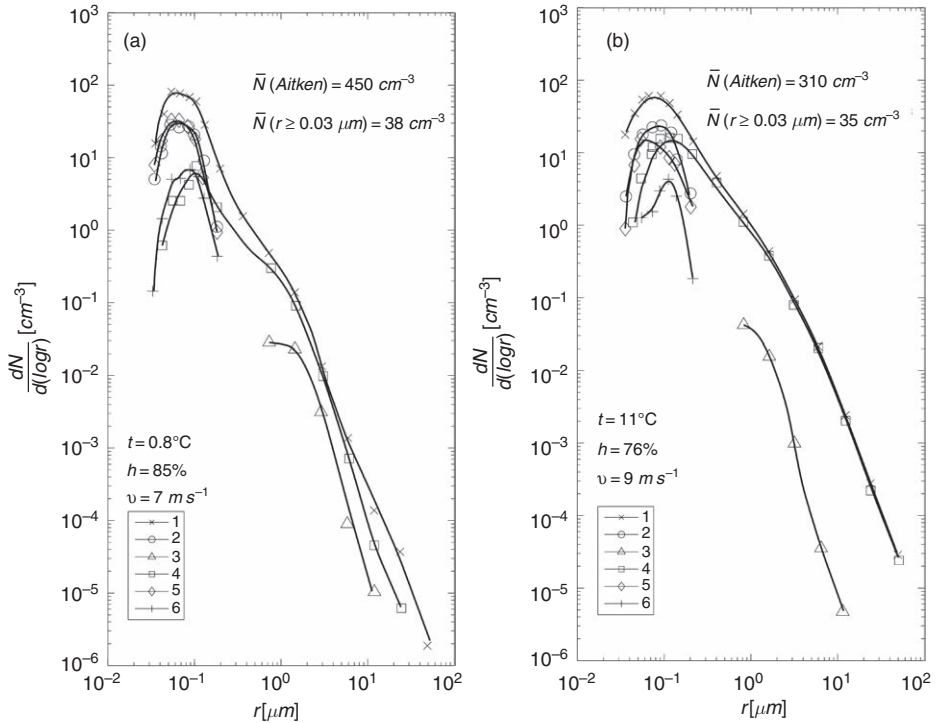


Fig. 6.3 Concentration of aerosol particles for two size ranges and size distribution of particles of different types in the range of $r \geq 0.03 \mu\text{m}$ over two different parts of the oceans of the Southern Hemisphere: (1) all particles; (2) $(\text{NH}_4)_2\text{SO}_4$; (3) long-shaped particles; (4) NaCl ; (5) non-cubic crystalline particles; and (6) H_2SO_4 . The samples were taken at 12–14 h local time.

6.5 Brownian coagulation and the aging of aerosols

Most aerosol particles are much smaller than cloud droplets and can be influenced easily by the constant bombardment of air molecules and perform random motions called Brownian motion. Brownian motion is especially pronounced for particles $<0.5 \mu\text{m}$ because of their small size, and they can collide and coagulate to form larger particles. This process is called Brownian coagulation. Particles larger than $0.5 \mu\text{m}$ are more massive and hence their Brownian coagulation is much less efficient.

Now consider a cloud of aerosol particles freshly produced that may have an initial size distribution peaking at UFP size range as indicated by curve 1 in Fig. 6.4. As time goes on, particles smaller than $0.5 \mu\text{m}$ will perform rapid Brownian coagulation and become larger and larger, whereas for particles greater than $0.5 \mu\text{m}$ the coagulation is very slow and their size will not change as quickly as smaller ones. After a certain time period, the aerosol size distributions will become quasi-steady-state as indicated by curve 4, which will change very little as time goes on. This behavior is called the *aging of aerosols*.

Because of aging, the size distribution curves on the larger particle side tend to become a quasi-straight lines when plotted on the double-log chart, as is the case in Fig. 6.4. This prompted Junge to suggest a power-law representation of aerosol size distribution (later called the Junge size distribution):

$$\frac{dN}{d \log r} = Ar^\beta, \quad (6.2)$$

where $\beta \sim -3$. Many measurements indeed substantiate this quasi-constant slope. Clearly, this only represents one side of the curve and cannot represent the complete distribution.

6.6 Physicochemical pathways of aerosol production

Aerosol particles have sources both at the Earth's surface and in the atmosphere, and that makes the tracking of aerosol sources complicated. There are generally three broad categories of physicochemical pathways of particle production.

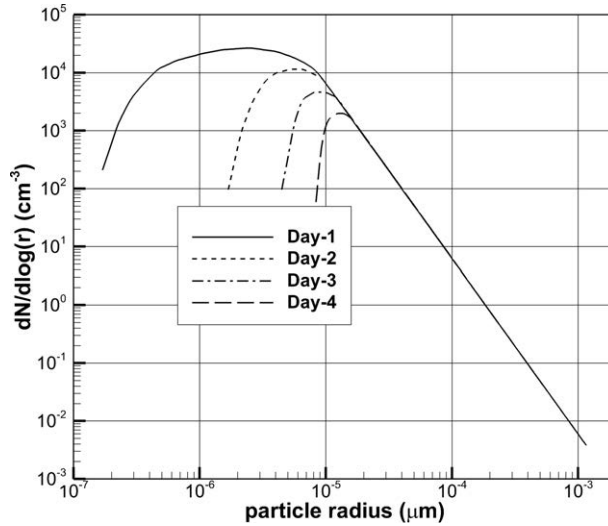
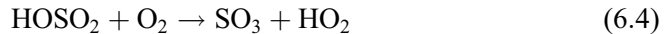


Fig. 6.4 A theoretical model interpretation of the aerosol aging phenomenon due to coagulation. Curves represent the aerosol size distribution from day 1 to day 4.

- (1) *Pulverization of bulk matter* – for example, the breaking up of rocks or dirt into finer particles due to weathering, the breaking of ocean surface to form small droplets due to winds or bubble burst, or the ejection of organic droplets from combustion engine exhaust.
- (2) *Reactions between different chemical species* – the reacting species can be either entirely in one phase (homogeneous reactions) or of different phases (heterogeneous reactions). When gases react to form aerosol particles, the process is called *gas-to-particle conversion* (GPC). One example of GPC is the formation of a sulfuric acid drop involving the hydroxyl radical OH as suggested by Warneck (1988):



Up to this stage, all molecules involved are in the gaseous phase. Then in the sulfuric acid–water vapor mixture, binary nucleation can occur to form sulfuric acid droplets.

It is also possible to have *drop-to-particle conversion* (DPC), and this occurs most commonly in clouds. Many gases and particulates are soluble in water and, if absorbed by cloud droplets, become aqueous solutions during cloud drop formation. When the cloud drop evaporates, the dissolved materials crystallize to become solid particles. If there are insoluble particles initially, they will become a part of the new particle, too. The new particle may be physically and chemically different from the original particles. This transformation of gases and particles by clouds to form new particles is often called the cloud processing of aerosol.

- (3) *Condensation* – this can be either the homogeneous condensation of gas to form liquid droplets or solid particles, or the freezing of liquid into solid, or heterogeneous condensation in which gases condense on liquids or solids, or liquids freeze on solids. Homogeneous condensation processes often occur during biomass burning. Liquids contained in the tissues of vegetation may vaporize during the combustion to become vapors which then recondense into liquid or solid form when cooled later.

6.6.1 Primary and secondary aerosol

Aerosol particles that are injected into the atmosphere directly without going through other processes in the atmosphere (such as the GPC or DPC mentioned above) are called *primary aerosol particles*. Examples are windblown dusts or pollens ejected by plants. Aerosol particles that form from further processes in the atmosphere such as GPC and DPC are called *secondary aerosol particles*.

6.7 Sources of aerosol particles

Aerosol particles come from different sources, which produce particles of different chemical compositions. Once formed, these particles may be transported by atmospheric circulations and interact with gases or with each other and coalesce to form mixed particles, making the analysis of particle chemistry complicated. The following are the major sources of aerosol particles.

6.7.1 Fragmentation of land surface

Dust storms

The land surface is full of small particles that can be carried into the air by winds or by an automobile driving on an unpaved dirt road. Most of these dust particles visible to our eyes are fairly coarse, typically 0.1–1 mm in diameter

The windblown dusts from arid regions of the world are an important source of ice nuclei. Chemically, the majority of the windblown dusts are various kinds of clay particles, such as SiO_2 , Al_2O_3 , Fe_2O_3 , and MgO .

Biomass burning

Each year, many parts of the world suffer from natural or man-made fires that destroy large areas of forests and other vegetated surfaces. These fires are

collectively called *biomass burning*. Forest or prairie fires are often said to be caused by lightning, although some must have been caused by careless handling of fire, such as smoking cigarettes or campfires. The image on the right was taken during a day with large-scale biomass burning and the air became opaque due to strong reflection of sunlight by fire-produced aerosol particles.

Recent studies also reveal that biomass burning can combine with deep convective systems to create a new kind of convection system called pyrocumulonimbus (pyroCb), which can carry combustion particles into the upper troposphere and lower stratosphere. This implies that the biomass burning particles can travel a long range, further than previously thought, and thus may have a far greater impact on the global climate.

Volcanic activity

The fact that volcanic eruptions can inject large amount of particles into the atmosphere is known from ancient times, and the story of the ash-buried city of Pompei in Italy is familiar to many. However, the amount and the height of aerosol injection vary greatly from one eruption to another. Volcanic eruptions inject directly into the atmosphere not only aerosol particles but a large amount of other gases as well. The most abundant volcanic gas turns out to be water vapor, followed by CO_2 and SO_2 . Among these gases, SO_2 is the precursor for the formation of sulfate aerosols. In the Mt. Pinatubo eruption, SO_2 injection is estimated at more than 2×10^7 tons, and much of this must have turned into sulfate particles and entered the stratosphere.

Human industrial activity

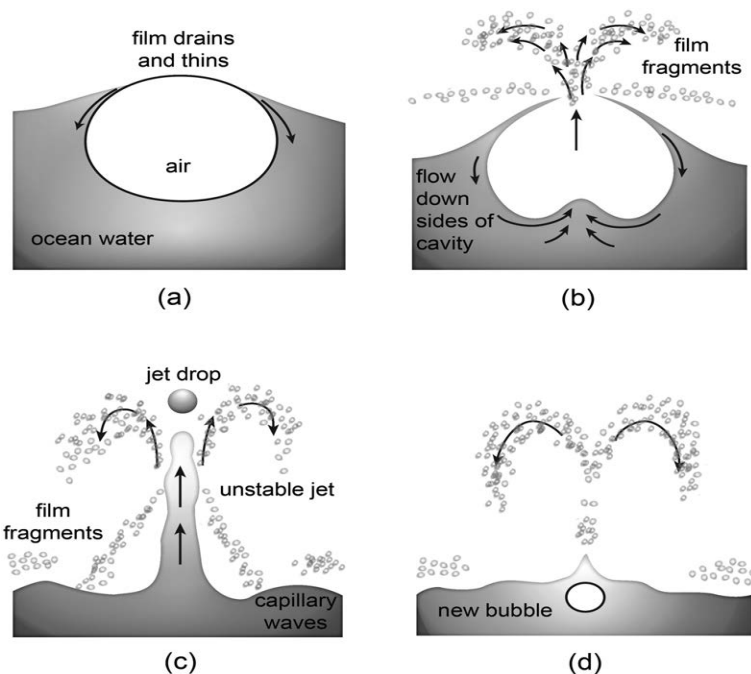
Human beings have been producing aerosol particles from ancient times, and most familiar to us is the burning of fuels (wood, coal, etc.) to obtain energy, which produces smoke.

The most conspicuous particle sources due to industry are the thick smoke coming out of numerous tall stacks in the world's industrial complexes. The most common outputs from these stacks are sulfates and black carbon. SO_2 produced by burning fossil fuels is a common precursor that usually ends up as sulfate particles through DPC or GPC. Specific factories produce specific particles; for example, downwind of paper mills one finds Na_2SO_4 , NH_4HSO_3 , $\text{Ca}(\text{SO}_3)_2$, NaOH , Na_2SO_3 , and H_2SO_4 .

6.7.2 Ocean surface process

Aerosol particles produced over the ocean surface are largely NaCl particles but there are also smaller amounts of K^+ , Mg^{2+} , CO_3^{2-} , and SO_4^{2-} . Organic and biological materials are also found in marine aerosols.

The production of sea salt particles is due to the bubble burst mechanism (also called sea spray mechanism) (Fig. 6.9). Since the sea surface is constantly subject to winds, air is trapped in the surface layer to form bubbles. Sea salt aerosol particles form in two stages of the burst process. First, when a bubble rises to the top of the surface, its cap becomes a very thin film (a), which eventually ruptures to form numerous mini-droplets that fly into the air (b). Some of these film droplets eventually evaporate to leave dry sea salt particles that may be carried up higher by winds. At the same time, the water in the remnant bubble creates a jet due to the buildup of pressure at the bottom of the bubble. As the jet rises, instability grows such that the jet breaks into a few droplets that also fly into the air (c). These jet drops are usually larger than the film drops. Some of these droplets are then carried by winds to higher layers and evaporate to become dry sea salt particles (d). Some studies found that sea salt particles produced in this way range from less than $0.1 \mu\text{m}$ (the cap droplets) to more than $10 \mu\text{m}$ (jet droplets). The number of droplets and hence the number of sea salt particles produced depends on the bubble size. The bubble size presumably depends on the winds over the sea surface and also the local conduciveness conditions for wave breaking.



6.7.3 Biogenic aerosols

Aerosol particles are produced by biological systems during their life cycle and plants are the main source of these biogenic aerosols. Pollens released by the flowers of various plants are spread in the air during pollination to become aerosol particles. They are typically about 10 μm or larger in size. Many plant fragments, spores of fungi, virus, and bacteria range from a few micrometers to less than 1 μm in size and thus can easily become airborne as aerosol particles. Some of these biogenic aerosol particles may serve as efficient nuclei for condensation and freezing.

6.7.4 Extraterrestrial source

The Earth's atmosphere is constantly bombarded by meteoroids, which are basically rocks of various sizes; some are larger than a few centimeters but most are probably just of dust particle size. These rocks are widely distributed in the interplanetary space in the Solar System and are attracted to the Earth by the gravitational force if their orbits happen to intercept that of the Earth. Because of their already high orbiting velocities around the Sun, they can have high relative velocities with respect to the Earth, which can be 50 km s^{-1} or greater when they enter the atmosphere. The extreme heating due to the ram pressure created by such high velocities causes these particles to burn and emit light – a phenomenon called meteors. Particles of sand dust to pebble size probably evaporate completely in the atmosphere, but some larger particles may break up into smaller fragments to become aerosol particles in the Earth's atmosphere. This is the extraterrestrial source of aerosol particles. The vapor of meteors can recondense to become small aerosol particles, possibly of nanometer size.

In very rare cases, meteoroids are large enough to survive the burn through the whole atmosphere and arrive at the surface with identifiable remains called meteorites. If this occurs at night, their light display against the night sky may be quite spectacular as to make the headlines in local newspapers.

Meteors usually occur in the mesosphere at an altitude somewhere between 80 and 95 km, hence the original “source” of meteoric aerosol particles must also be located at this altitude. Meteoric aerosols are known to contain elements such as Fe, Si, Mg, S, Ca, Ni, Al, Cr, Mu, Cl, K, Ti, and Co (Cameron, 1981).

6.8 Removal mechanisms of aerosol particles

The pathways via which aerosol particles are removed from the atmosphere can be divided into two broad categories – dry removal and wet removal. Dry removal mechanisms are those not involving cloud and precipitation processes, while wet removals are those related to clouds and precipitation.

6.8.1 Dry removal

Gravitational settling

Aerosol particles can stay afloat by updrafts that are strong enough to support them. If the updraft weakens or disappears, particles will go down to the surface by the pull of gravity of the Earth. The settling velocity of an aerosol particle depends on the density and size of the particle and where it is located in the atmosphere (because the air density affects the viscosity and hence the drag force on the particle).

We see that particles have higher fall velocities at higher levels because of the lower air density that results in smaller drag force. Taking a particle with radius $0.01\ \mu\text{m}$ as an example, its fall velocity is about $10^{-4}\ \text{cm s}^{-1}$ at $z = 10\ \text{km}$ but is about four times faster at $z = 20\ \text{km}$. The velocity change with height is less for larger particles. For all these particles, the fall velocities are very small. We can make a quick estimate of the time required for them to fall a certain distance. For example, a particle of a few micrometers radius has a fall velocity $\sim 1\ \text{cm s}^{-1}$ at $z = 10\ \text{km}$ and it will take this particle more than $10^6\ \text{s}$ (~ 12 days) to fall 1 km. A particle of radius $\sim 0.1\ \mu\text{m}$ has a fall velocity $\sim 10^{-3}\ \text{cm s}^{-1}$ at $z \sim 10\ \text{km}$ and hence will take $10^9\ \text{s}$ (~ 32 years) to fall 1 km! Obviously, submicrometer particles can stay in the atmosphere for a very long time.

Attachment on obstacle surfaces

Aerosol particles carried by air flow can become attached to the surface of plants, buildings or other obstacles above the ground either by their own Brownian motion, electrostatic forces, interception or inertial impaction (Fig. 6.11).

- (1) *Inertial impaction* is caused by the finite inertia of the aerosol particle. When the aerosol-laden air flows past an obstacle, for example, a cylindrical rod, the air molecules will follow the streamlines, which will not intersect the rod (except

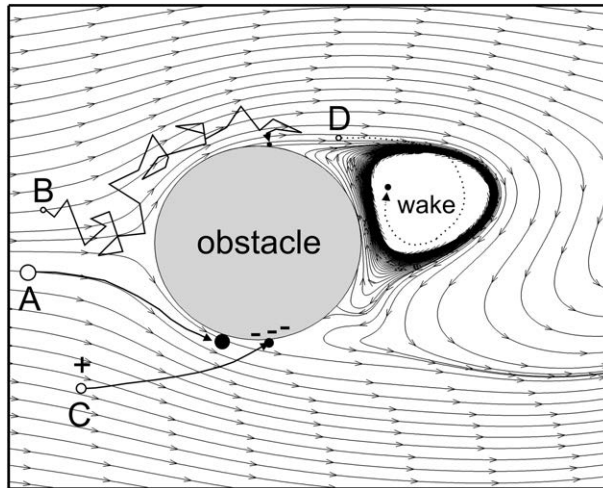


Fig. 6.11 A schematic illustrating the mechanisms involved in the dry removal of aerosol particles.

for the center streamline) but will go around it. An aerosol particle, such as particle A in Fig. 6.11, however, has finite inertia and hence its trajectory will deviate from the streamline such that it is more likely to impact on the rod and become attached. The position where the collision occurs also depends on the dimension of the particle and that effect is called interception.

Particle A collides with the obstacle in the front part. That is usually what happens to particles with larger inertia. For particles with small enough inertia, such as particle D in Fig. 6.11, it is possible that they move past the obstacle but get sucked into the wake region due to the lower pressure of the wake and become trapped there. This is called wake capture.

- (2) *Brownian motion* is due to the random bombardment of the aerosol particle by air molecules that causes the particle to perform zigzag motions and become captured by the obstacle, such as particle B in Fig. 6.11. We have discussed Brownian motion previously.
- (3) *Electrostatic forces* result because many particles and obstacles are electrically charged. If the signs of the charges on the particle and the obstacle are opposite, as is the case for particle C in Fig. 6.11, then the particles will be attracted to the obstacle by electrostatic forces and become captured.

Particle coagulation

We have already mentioned Brownian coagulation in which small particles collide and stick together to become larger particles. In accounting for the aerosol budget, coagulation causes particle concentration to decrease and also the coalesced particles are “removed” from their original size categories, so it should be considered as a sink of aerosol particles.

6.8.2 Wet removal

Cloud and precipitation processes are the most efficient way to remove aerosol particles from the atmosphere, and the process that removes aerosol particles is wet removal, also called precipitation scavenging.

Wet removal is commonly divided into two types: in-cloud scavenging and below-cloud scavenging. Sometimes they are also called rainout and washout, respectively.

In-cloud scavenging (rainout)

In-cloud scavenging, by definition, includes all mechanisms occurring in the cloud that result in aerosol removal. The most important one is nucleation scavenging, in which aerosol particles serve as either CCN or IN for the formation of cloud drops or ice particles, and thus are removed from the atmosphere. Other in-cloud scavenging mechanisms include the attachment of aerosol particles on cloud and precipitation particles due to Brownian motion, attractive forces, and inertial impaction.

Below-cloud scavenging (washout)

Once precipitation particles (raindrops, snowflakes, graupel/hail) fall from the cloud, they can remove aerosol particles either by inertial impaction or by Brownian collision of the aerosol particles. This is called below-cloud scavenging.

Problems

6.1. It is known that the terminal velocity of a very small spherical particle of radius a can be calculated by the following equation

$$u_{\infty} \approx \frac{2}{9} \eta_a a^2 \rho_p g,$$

where ρ_p is the particle density and η_a is the dynamic viscosity of air. The latter can be calculated by

$$\begin{aligned} \eta_a &= (1.718 + 0.0049T) \times 10^{-4} \quad \text{for } T \geq 0^{\circ}\text{C}, \\ \eta_a &= (1.718 + 0.0049T - 1.2 \times 10^{-5}T^2) \times 10^{-4} \quad \text{for } T < 0^{\circ}\text{C}. \end{aligned}$$

Note that η_a is in the unit of poise (1 poise = 1 P = 1 g cm⁻¹ s⁻¹) and T is in °C when using these formulas.

- Determine the fall velocities of particles of density 1 g cm⁻³ and radii 1, 10, and 100 μm falling in air of $p = 1000$ hPa and $T = 20^{\circ}\text{C}$.
- Calculate the time required for these particles to fall 1 km in air.
- Repeat the calculations of (a) and (b) but for air with $p = 200$ hPa and

Cloud Microphysics

Cloud Condensation Nuclei

A small subset of the atmospheric aerosol serves as particles upon which water vapor condenses to form droplets that are activated and grow by condensation to form cloud droplets at the supersaturations achieved in clouds (~ 0.1 – 1%). These particles are called *cloud condensation nuclei* (CCN). The larger the size of a particle, the more readily it is wetted by water, and the greater its solubility, the lower will be the supersaturation at which the particle can serve as a CCN. For example, to serve as a CCN at 1% supersaturation, completely wettable but water-insoluble particles need to be at least $\sim 0.1 \mu\text{m}$ in radius, whereas soluble particles can serve as CCN at 1% supersaturation even if they are as small as $\sim 0.01 \mu\text{m}$ in radius. Most CCN consist of a mixture of soluble and insoluble components (called *mixed nuclei*).

The concentrations of CCN active at various supersaturations can be measured with a *thermal diffusion chamber*. This device consists of a flat chamber in which the upper and lower

horizontal plates are kept wet and maintained at different temperatures, with the lower plate being several degrees colder than the upper plate. By varying the temperature difference between the plates, it is possible to produce maximum supersaturations in the chamber that range from a few tenths of 1% to a few percent, which are similar to the supersaturations that activate droplets in clouds. Small water droplets form on those particles that act as CCN at the peak supersaturation in the chamber. The concentration of these droplets can be determined by photographing a known volume of the cloud and counting the number of droplets visible in the photograph or by measuring the intensity of light scattered from the droplets. By repeating the aforementioned procedure with different temperature gradients in the chamber, the concentrations of CCN in the air at several supersaturations (called the *CCN supersaturation spectrum*) can be determined.

Worldwide measurements of CCN concentrations have not revealed any systematic latitudinal or sea-seasonal variations. However, near the Earth's surface, continental air masses generally contain larger concentrations of CCN than marine air masses (Fig. 6.5). For example, the concentration of CCN in the continental air mass over the Azores, depicted in Fig. 6.5,

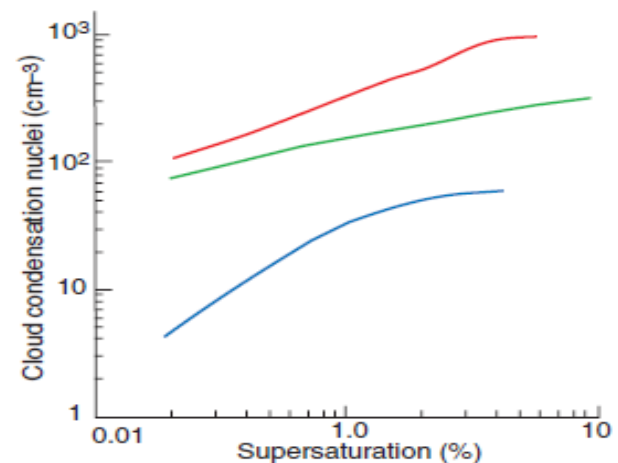


Fig. Cloud condensation nucleus spectra in the boundary layer from measurements near the Azores in a polluted continental air mass (brown line), in Florida in a marine air mass (green line), and in clean air in the Arctic (blue line).

is about $\sim 300 \text{ cm}^{-3}$ at 1% supersaturation, in the marine air mass over Florida it is $\sim 100 \text{ cm}^{-3}$, and in clean Arctic air it is only $\sim 30 \text{ cm}^{-3}$. The ratio of CCN (at 1% supersaturation) to the total number of particles in the air (CN) is $\sim 0.2\text{--}0.6$ in marine air; in continental air this ratio is generally less than ~ 0.01 but can rise to ~ 0.1 . The very low ratio of CCN to CN in continental air is attributable to the large number of very small particles, which are not activated at low supersaturations. Concentrations of CCN over land decline by about a factor of five between the planetary boundary layer and the free troposphere. Over the same height interval, concentrations of CCN over the ocean remain fairly constant or may even increase with height, reaching a maximum concentration just above the mean cloud height. Ground-based measurements indicate that there is a diurnal variation in CCN concentrations, with a minimum at about 6 a.m. and a maximum at about 6 p.m.

The observations just described provide clues as to the origins of CCN. First of all it appears that the land acts as a source of CCN because the concentrations of CCN are higher over land and decrease with altitude. Some of the soil particles and dusts that enter the atmosphere probably serve as CCN, but they do not appear to be a dominant source. The rate of production of CCN from burning vegetable matter is on the order of $10^{12}\text{--}10^{15}$ per kg of material consumed. Thus, forest fires are a source of CCN. About 80% of the particles emitted by idling diesel engines are CCN at 1% supersaturation.

There appears to be a widespread and probably a fairly uniform source of CCN over both the oceans and the land, the nature of which has not been definitely established. A likely candidate is gas-to-particle conversion, which can produce particles up to a few tenths of a micrometer in diameter that can act as CCN if they are soluble or wettable. Gas-to-particle conversion mechanisms that require solar radiation might be responsible for the observed peak in CCN concentrations at ~ 6 p.m. Many CCN consist of sulfates.

Over the oceans, organic sulfur from the ocean [in the form of the gases dimethyl sulfide (DMS) and methane sulfonic acid (MSA)] provides a source of CCN, with the DMS and MSA being converted to sulfate in the atmosphere. Evaporating clouds also release sulfate particles.