

CHAPTER 8. AEROSOLS

Aerosols in the atmosphere have several important environmental effects. They are a respiratory health hazard at the high concentrations found in urban environments. They scatter and absorb visible radiation, limiting visibility. They affect the Earth's climate both directly (by scattering and absorbing radiation) and indirectly (by serving as nuclei for cloud formation). They provide sites for surface chemistry and condensed-phase chemistry to take place in the atmosphere. We present in this chapter a general description of the processes controlling aerosol abundances and go on to discuss radiative effects in more detail. Chemical effects will be discussed in subsequent chapters.

8.1 SOURCES AND SINKS OF AEROSOLS

Atmospheric aerosols originate from the condensation of gases and from the action of the wind on the Earth's surface. *Fine* aerosol particles (less than 1 μm in radius) originate almost exclusively from condensation of precursor gases. A typical chemical composition for fine aerosol in the lower troposphere is shown in Figure 8-1. A key precursor gas is sulfuric acid (H_2SO_4), which is produced in the atmosphere by oxidation of sulfur dioxide (SO_2) emitted from fossil fuel combustion, volcanoes, and other sources. H_2SO_4 has a low vapor pressure over H_2SO_4 - H_2O solutions and condenses under all atmospheric conditions to form aqueous sulfate particles. The composition of these sulfate particles can then be modified by condensation of other gases with low vapor pressure including NH_3 , HNO_3 , and organic compounds. Organic carbon represents a major fraction of the fine aerosol (Figure 8-1) and is contributed mainly by condensation of large hydrocarbons of biogenic and anthropogenic origin. Another important component of the fine aerosol is soot produced by condensation of gases during combustion. Soot as commonly defined includes both elemental carbon and black organic aggregates.

Mechanical action of the wind on the Earth's surface emits sea salt, soil dust, and vegetation debris into the atmosphere. These aerosols consist mainly of *coarse* particles 1-10 μm in radius. Particles finer than 1 μm are difficult to generate mechanically because they have large area-to-volume ratios and hence their surface tension per unit aerosol volume is high. Particles coarser than 10 μm are not easily lifted by the wind and have short atmospheric lifetimes because of their large sedimentation velocities.

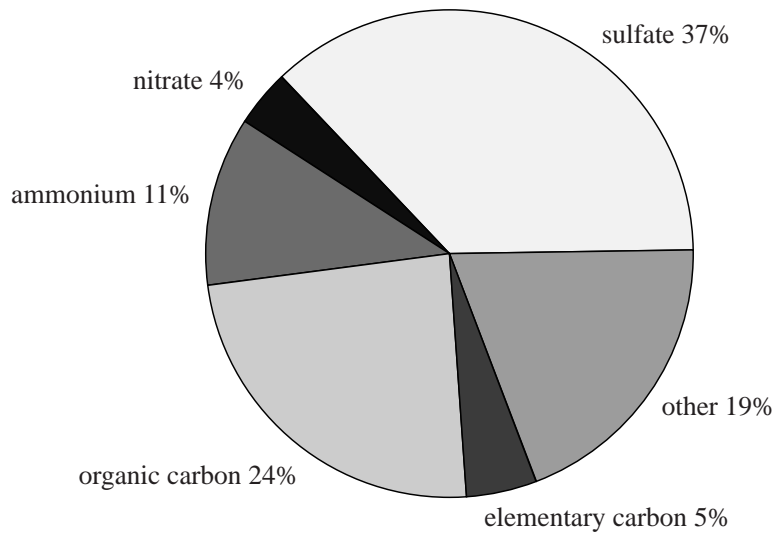


Figure 8-1 Typical composition of fine continental aerosol. Adapted from Heintzenberg, J., *Tellus*, 41B, 149-160, 1989.

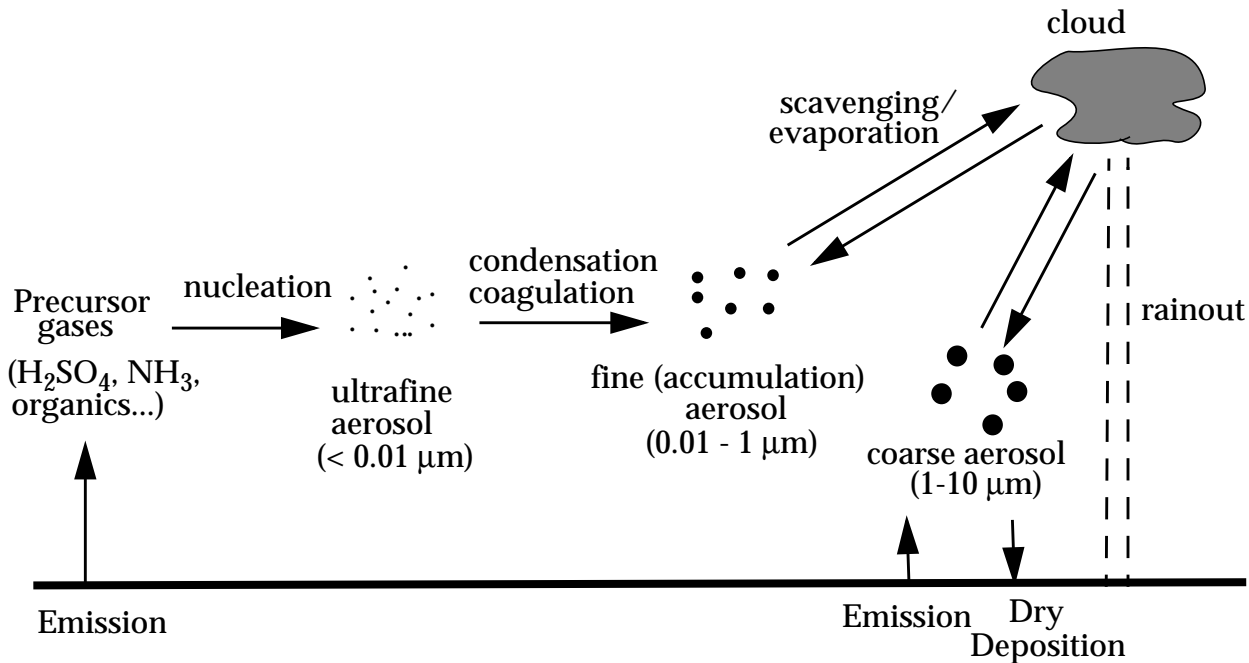


Figure 8-2 Production, growth, and removal of atmospheric aerosols

Figure 8-2 illustrates the different processes involved in the production, growth, and eventual removal of atmospheric aerosol particles. Gas molecules are typically in the 10^{-4} - $10^{-3} \mu\text{m}$ size range. Clustering of gas molecules (*nucleation*) produces *ultrafine aerosols* in the 10^{-3} - $10^{-2} \mu\text{m}$ size range. These ultrafine aerosols grow

rapidly to the 0.01-1 μm *fine aerosol* size range by condensation of gases and by *coagulation* (collisions between particles during their random motions). Growth beyond 1 μm is much slower because the particles are by then too large to grow rapidly by condensation of gases, and because the slower random motion of large particles reduces the coagulation rate. Aerosol particles originating from condensation of gases tend therefore to accumulate in the 0.01-1 μm size range, often called the *accumulation mode* (as opposed to the *ultrafine mode* or the *coarse mode*). These particles are too small to sediment at a significant rate, and are removed from the atmosphere mainly by scavenging by cloud droplets and subsequent rainout (or direct scavenging by raindrops).

Coarse particles emitted by wind action are similarly removed by rainout. In addition they sediment at a significant rate, providing another pathway for removal. The sedimentation velocity of a 10 μm radius particle at sea level is 1.2 cm s^{-1} , as compared to 0.014 cm s^{-1} for a 0.1 μm particle.

The bulk of the atmospheric aerosol mass is present in the lower troposphere, reflecting the short residence time of aerosols against deposition (~ 1 -2 weeks; see problem 8. 1). Aerosol concentrations in the upper troposphere are typically 1-2 orders of magnitude lower than in the lower troposphere. The stratosphere contains however an ubiquitous H_2SO_4 - H_2O aerosol layer at 15-25 km altitude, which plays an important role for stratospheric ozone chemistry (chapter 10). This layer arises from the oxidation of carbonyl sulfide (COS), a biogenic gas with an atmospheric lifetime sufficiently long to penetrate the stratosphere. It is augmented episodically by oxidation of SO_2 discharged in the stratosphere from large volcanic eruptions such as Mt. Pinatubo in 1991. Although the stratospheric source of H_2SO_4 from COS oxidation is less than 0.1% of the tropospheric source of H_2SO_4 , the lifetime of aerosols in the stratosphere is much longer than in the troposphere due to the lack of precipitation.

8.2 RADIATIVE EFFECTS

8.2.1 Scattering of radiation

A radiation beam is *scattered* by a particle in its path when its direction of propagation is altered without absorption taking place. Scattering may take place by reflection, refraction, or diffraction of the radiation beam (Figure). We define the *scattering efficiency* of a particle as the probability that a photon incident on the particle will

be scattered. Figure 8-4 shows the scattering efficiency of green light ($\lambda = 0.5 \mu\text{m}$) by a spherical water particle as a function of particle size. Scattering is maximum for a particle radius corresponding to the wavelength of radiation. Larger particles also scatter radiation efficiently, while smaller particles are inefficient scatterers. Atmospheric aerosols in the accumulation mode are efficient scatterers of solar radiation because their size is of the same order as the wavelength of radiation; in contrast, gases are not efficient scatterers because they are too small. Some aerosol particles, such as soot, also *absorb* radiation.

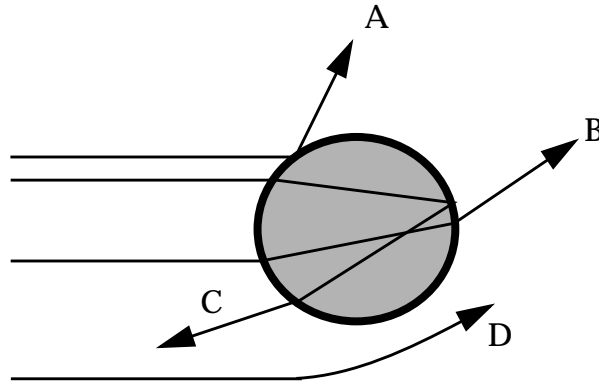


Figure 8-3 Scattering of a radiation beam: processes of reflection (A), refraction (B), refraction and internal reflection (C), and diffraction (D).

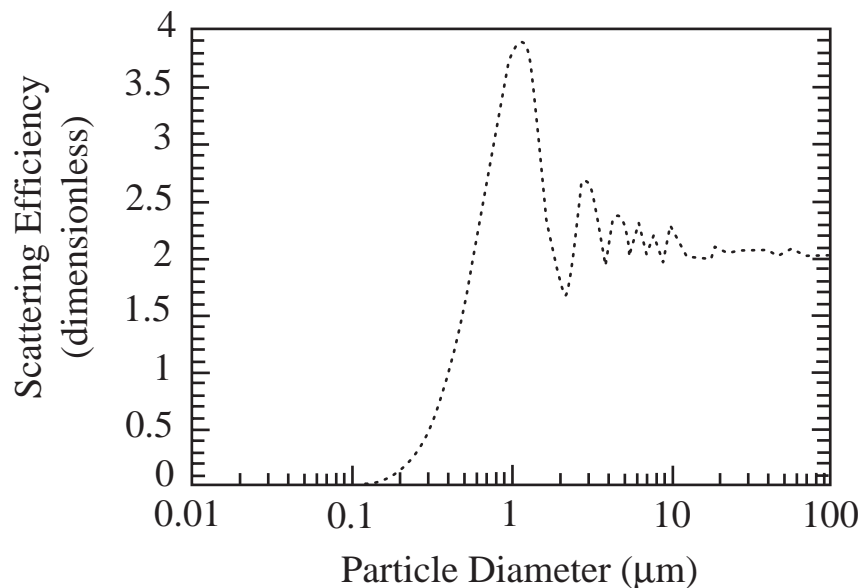


Figure 8-4 Scattering efficiency of green light ($\lambda = 0.5 \mu\text{m}$) by a liquid water sphere as a function of the diameter of the sphere. Scattering efficiencies can be larger than unity because of diffraction. Adapted from Jacobson, M.Z., *Fundamentals of Atmospheric Modeling*, Cambridge University Press, Cambridge, 1998.

8.2.2 Visibility reduction

Atmospheric visibility is defined by the ability of our eyes to distinguish an object from the surrounding background. Scattering of solar radiation by aerosols is the main process limiting visibility in the troposphere (Figure 8-5). In the absence of aerosols our visual range would be approximately 300 km, limited by scattering by air molecules. Anthropogenic aerosols in urban environments typically reduce visibility by one order of magnitude relative to unpolluted conditions. Degradation of visibility by anthropogenic aerosols is also a serious problem in U.S. national parks such as the Grand Canyon and the Great Smoky Mountains. The visibility reduction is greatest at high relative humidities when the aerosols swell by uptake of water (exercise 1-3), increasing the cross-sectional area for scattering; this is the phenomenon known as *haze*.

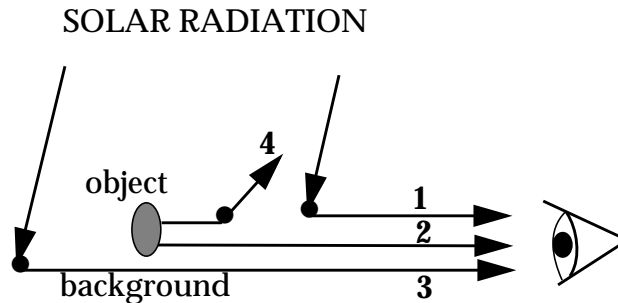


Figure 8-5 Reduction of visibility by aerosols. The visibility of an object is determined by its contrast with the background (2 vs. 3). This contrast is reduced by aerosol scattering of solar radiation into the line of sight (1) and by scattering of radiation from the object out of the line of sight (4).

8.2.3 Perturbation to climate

Scattering of solar radiation by aerosols increases the Earth's albedo because a fraction of the scattered light is reflected back to space. The resulting cooling of the Earth's surface is manifest following large volcanic eruptions, such as Mt. Pinatubo in 1991, which inject large amounts of aerosol into the stratosphere. The Pinatubo eruption was followed by a noticeable decrease in mean surface temperatures for the following 2 years (Figure 8-6) because of the long residence time of aerosols in the stratosphere. Remarkably, the optical depth of the stratospheric aerosol following a large volcanic eruption is comparable to the optical depth of the anthropogenic aerosol in the troposphere. The natural experiment offered by erupting volcanoes thus strongly implies that anthropogenic aerosols exert a significant cooling effect on the Earth's climate.

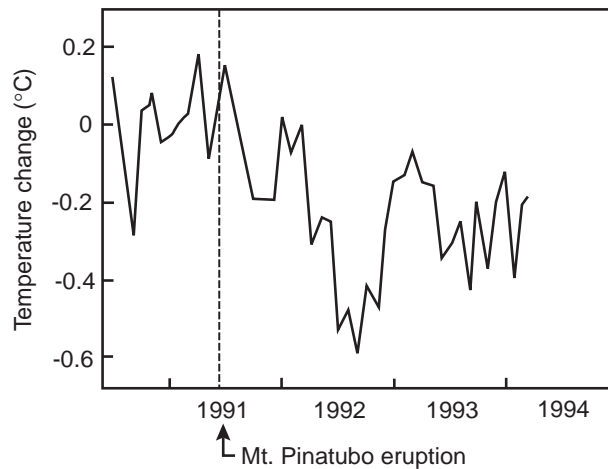


Figure 8-6 Observed change of the Earth's global mean surface temperature following the Mt. Punatubo eruption (September 1991). Adapted from *Climate Change 1994*, Cambridge University Press, New York, 1995.

We present here a simple model to estimate the climatic effect of a scattering aerosol layer of optical depth δ . It is estimated that the global average scattering optical depth of aerosols is about 0.1 and that 25% of this optical depth is contributed by anthropogenic aerosols. The radiative forcing from the anthropogenic aerosol layer is (section 7.4):

$$\Delta F = - \frac{F_S \Delta A}{4} \quad (8.1)$$

where ΔA is the associated increase in the Earth's albedo (note that ΔF is negative; the effect is one of cooling). We need to relate δ to ΔA .

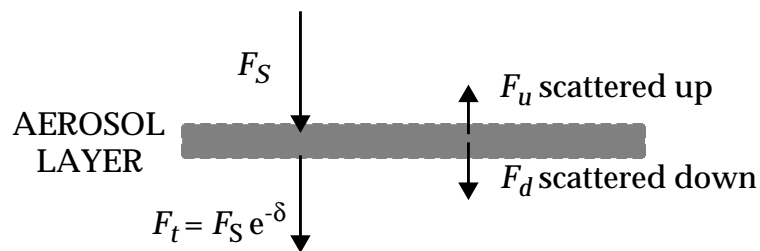


Figure 8-7 Scattering of radiation by an aerosol layer

In Figure 8-7, we decompose the solar radiation flux incident on the aerosol layer (F_S) into components transmitted through the layer ($F_t = F_S e^{-\delta}$), scattered forward (F_d), and scattered backward (F_u). Because $\delta \ll 1$, we can make the approximation $e^{-\delta} \approx 1 - \delta$. The

scattered radiation flux $F_d + F_u$ is given by

$$F_d + F_u = F_S - F_t = F_S - F_S(1 - \delta) = F_S\delta \quad (8.2)$$

The albedo A^* of the aerosol layer is defined as

$$A^* = \frac{F_u}{F_S} \quad (8.3)$$

As illustrated in Figure , an aerosol particle is more likely to scatter radiation in the forward direction (beams A, B, D) than in the backward direction (beam C). Observations and theory indicate that only a fraction $\beta \approx 0.2$ of the total radiation scattered by an aerosol particle is directed backward. By definition of β ,

$$F_u = \beta(F_d + F_u) = \beta\delta F_S \quad (8.4)$$

Replacing (8.4) into (8.3) we obtain

$$A^* = \beta\delta \quad (8.5)$$

which yields $A^* = 5 \times 10^{-3}$ for the global albedo of the anthropogenic aerosol.

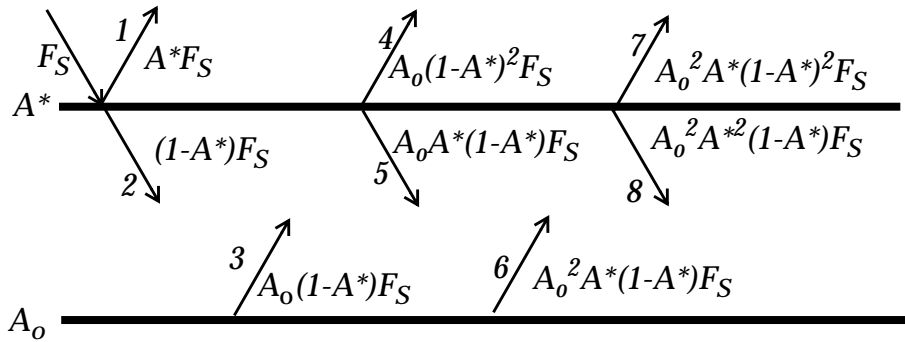


Figure 8-8 Reflection of solar radiation by two superimposed albedo layers A^* and A_o . A fraction A^* of the incoming solar radiation F_S is reflected by the top layer to space (1). The remaining fraction $1-A^*$ propagates to the bottom layer (2) where a fraction A_o is reflected upward (3). Some of that reflected radiation is propagated through the top layer (4) while the rest is reflected (5). Further reflections between the top and bottom layer add to the total radiation reflected out to space (7).

The actual albedo enhancement ΔA is less than A^* because of horizontal overlap of the aerosol layer with other reflective surfaces

such as clouds or ice. Aerosols present above or under a white surface make no contribution to the Earth's albedo. We take this effect into account in Figure 8-8 by superimposing the reflection of the incoming solar radiation F_S by the anthropogenic aerosol layer (A^*) and by natural contributors to the Earth's albedo (A_o). We assume random spatial overlap between A^* and A_o .

The total albedo A_T from the superimposed albedo layers A^* and A_o is the sum of the fluxes of all radiation beams reflected upward to space, divided by the incoming downward radiation flux F_S :

$$A_T = A^* + A_o(1 - A^*)^2 + A_o^2 A^* (1 - A^*)^2 + \sum_{n=2}^{\infty} A_o^{n+1} A^{*n} (1 - A^*)^2 \quad (8.6)$$

We sort the terms on the right-hand side by their order in A^* . Since $A^* \ll 1$, we neglect all terms higher than first-order:

$$A_T = A_o + A^*(1 - 2A_o + A_o^2) + O(A^{*2}) \approx A_o + A^*(1 - A_o)^2 \quad (8.7)$$

so that the albedo enhancement from the aerosol layer is $\Delta A = A_T - A_o = A^*(1 - A_o)^2$. Replacing into equation (8.1) we obtain the radiative forcing from the anthropogenic aerosol:

$$\Delta F = \frac{-F_S A^* (1 - A_o)^2}{4} \quad (8.8)$$

Substituting numerical values yields $\Delta F = -0.9 \text{ W m}^{-2}$, consistent with the values shown in Figure 7-15 and compensating about a third of the greenhouse radiative forcing over the past century. This *direct* forcing represents the radiative effect from scattering of solar radiation by aerosols. There is in addition an *indirect* effect shown in Figure 7-15 associated with the role of aerosols as nuclei for cloud droplet formation; a cloud forming in a polluted atmosphere distributes its liquid water over a larger number of aerosol particles than in a clean atmosphere, resulting in a larger cross-sectional area of cloud droplets and hence a larger cloud albedo. This indirect effect is considerably more uncertain than the direct effect but could make a comparable contribution to the aerosol radiative forcing.

Anthropogenic aerosols may explain at least in part why the Earth

has not been getting as warm as one would have expected from increasing concentrations of greenhouse gases. A major difficulty in assessing the radiative effect of aerosols is that aerosol concentrations are highly variable from region to region, a consequence of the short lifetime. Long-term temperature records suggest that industrial regions of the eastern United States and Europe, where aerosol concentrations are high, may have warmed less over the past century than remote regions of the world, consistent with the aerosol albedo effect. Recent observations also indicate a large optical depth from soil dust aerosol emitted by arid regions, and there is evidence that this source is increasing as a result of desertification in the tropics. Because of their large size, dust particles not only scatter solar radiation but also absorb terrestrial radiation, with complicated implications for climate (problem 8. 2).

Further reading:

Intergovernmental Panel of Climate Change, *Climate Change 1994*, Cambridge University Press, 1995. Radiative effects of aerosols.

Jacobson, M.Z., *Fundamentals of Atmospheric Modeling*, Cambridge University Press, Cambridge, 1998. Aerosol scattering and absorption.

Seinfeld, J.H., and S.N. Pandis, *Atmospheric Chemistry and Physics of Air Pollution*, Wiley, 1986. Aerosol microphysics: nucleation, condensation, coagulation, deposition.