

Chapter I. Introduction

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TABLE OF CONTENTS

1.1	Description of atmospheric aerosols.....	1
1.2	Climate effects of aerosols	2
1.2.1.	Direct and indirect effects	3
1.2.2.	Anthropogenic aerosol climate forcing.....	4
1.3	Reducing uncertainties in estimating aerosol climate effects.....	5
1.3.1.	Synergy between observations and model.....	5
1.3.2.	Estimates of emissions.....	6
1.3.3.	Aerosol representation in GCMs	7
1.4	Contents of this report	8
Inset 1:	Atmospheric and aerosol properties	10
Inset 2:	Molecular and aerosol light interaction	11
Inset 3:	Key properties in aerosol radiative forcing.....	12
References	13

1.1 Description of atmospheric aerosols

Earth's atmosphere consisting primarily of a mixture of gases also contains particles, such as aerosols and clouds. Aerosols are suspended liquid or solid particles whose typical diameters range over four orders of magnitude (sizes from ~ 3 nanometers, nm, to a few hundredths of millimeters, mm, generally smaller than cloud droplets) with a wide dynamic range of composition and shape, depending on their sources and atmospheric processes. It is well known that aerosols can have a variety of important impacts on the environment. Aerosols, also known as particulate matter, have long been recognized as pollutants of concern and may have detrimental effects on human health, such as impairment of pulmonary function. Sulfate and nitrate aerosols are also primarily responsible for acid deposition. Aerosols likewise strongly interact with solar and terrestrial radiation in two different ways. First, they directly scatter and absorb solar (shortwave) radiation (Insets 1 & 2). Second, by acting as cloud condensation nuclei they modify physical and chemical properties of clouds and thus can alter precipitation processes and indirectly affect cloud particle interaction with solar and terrestrial radiation. The net result of these effects is thought to be an enhancement of Earth's shortwave albedo (reflectance) affecting Earth's radiation budget and climate, and also a redistribution of the deposition of radiant and latent heat energy in the atmosphere, with possible effects on atmospheric circulations and precipitation patterns on a variety of length scales.

Major aerosol processes that influence climate are illustrated in **Figure 1.1**. Both natural and man-made processes generate aerosols. Some aerosols are emitted directly to the atmosphere (primary aerosols), while some are formed in the atmosphere from gaseous precursors through photochemical production (secondary aerosols). The amount of aerosols in the atmosphere has greatly increased over the industrial period. The nature of this particulate matter has substantially changed as a consequence of evolving emissions from industrial and residential activities, mainly combustion related. These anthropogenic aerosols are often observable as dust, smoke, haze, and in and downwind of urban environments, as smog. Important classes of natural aerosols are: sulfates - from ocean spray, volcanic emissions, and oxidized sulfides released from the ocean or decomposition of organic matter; sea salt - produced mainly from spray from breaking bubbles of ocean whitecaps; mineral dust; smoke from natural wildfires; and secondary aerosols from gas to particle conversion, mainly of natural hydrocarbons (terpenes, isoprene) emitted by vegetation that is oxidized in the atmosphere to low volatility products which condense to form aerosols. Volcanic eruptions emit large quantities of primary aerosols, which tend to be removed fairly rapidly by dry deposition, and also sulfur dioxide, a source of secondary aerosols; the latter, when injected into the stratosphere by an explosive volcano (e.g., Pinatubo, in the Philippines, in 1991) can form large amounts of sulfuric acid aerosol, which can persist, depending on altitude, for several years.

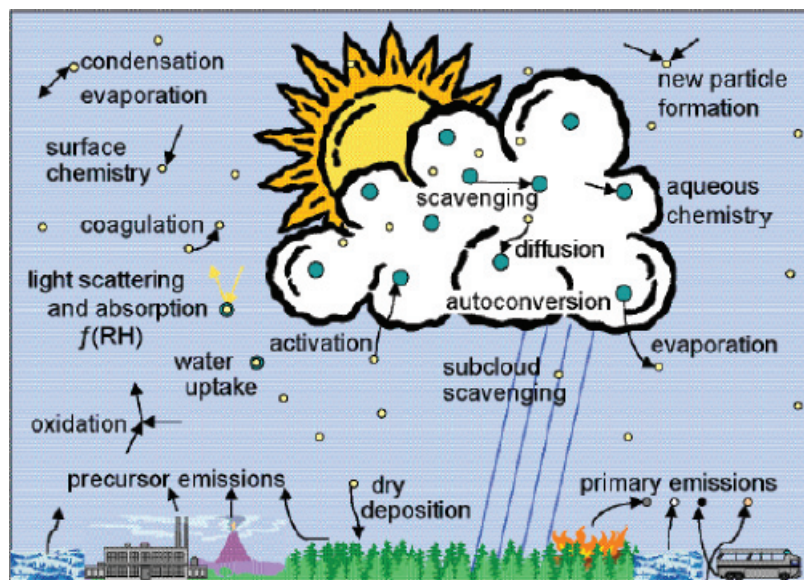


Figure 1.1 Major aerosol processes that influence climate. Aerosol particles are directly emitted as primary particles and are formed secondarily by oxidation of emitted gaseous precursors. The formation of low-volatility materials in this way results in new particle formation and condensation onto existing particles. Aqueous-phase oxidation of gas-phase precursors within cloud droplets accretes additional mass onto existing particles but does not result in new particle formation. Particles age by surface chemistry and coagulation as well as by condensation. With increasing relative humidity particles may accrete water vapor by deliquescence and further hygroscopic growth; with decreasing relative humidity water is lost and ultimately particles may effloresce to the dry state. The uptake of water increases particle size, affecting also the particle optical properties. During cloud formation some fraction of aerosol particles serve as cloud condensation nuclei, by becoming activated, that is, overcoming a free-energy barrier to form cloud droplets. Within clouds interstitial particles can become attached to cloud droplets by diffusion, and activated particles are combined when cloud droplets collide and coalesce. If cloud droplets evaporate the particles are resuspended, but if the cloud precipitates the particles are carried below the cloud and reach the surface, unless the precipitating particles completely evaporate. Aerosol particles below precipitating cloud can also be removed from the atmosphere by impaction by precipitating drops and by dry deposition to the surface. From Ghan and Schwartz (2007).

Aerosol particles are removed from the atmosphere mainly by wet deposition (uptake in cloud droplets followed by removal in precipitation) and to a lesser extent by dry deposition to vegetation, land surface, ocean water (gravitational settling of large particles; impaction of intermediate size particles, diffusion and attachment of small particles). The atmospheric residence time for tropospheric aerosols is typically about a week. As a consequence of the non-uniform distribution of sources and the short atmospheric residence time, the spatial distribution of aerosol particles in the atmosphere is quite non-uniform. For a mean atmospheric transport velocity of 5 m s^{-1} , this residence time of a week corresponds to a transport distance of 3000 km. Likewise at any given location the amount and nature of aerosols can vary substantially as a consequence of variability in atmospheric transport and in aerosol formation processes, driven largely by variability in controlling meteorology, and to some extent by intermittency of sources, e. g., wildfires, agricultural burning, or, in the extreme volcanic eruptions. For most aerosols, whose source is emissions at the surface, concentrations are greatest in the atmospheric boundary layer, decreasing with altitude in the free troposphere.

This report reviews the present state of understanding of the influences of aerosols on Earth's climate system, in particular, their direct and indirect effects for their consequences on climate change.

1.2 Climate effects of aerosols

The recognition, mainly in the past two decades, of the important influences of atmospheric aerosols on climate and climate change has generated a large amount of research. The increase in atmospheric aerosols over the industrial period is thought to have exerted a net cooling influence on Earth's climate relative to the pre-industrial period. The magnitude of this cooling influence, denoted by a negative forcing of climate change (see Inset 3 for a definition of forcing), is thought to be comparable to the warming influence (positive forcing) of enhanced atmospheric concentrations of greenhouse gases (*GHGs*) – mainly carbon dioxide, methane, nitrous oxide, chlorofluorocarbons, and ozone. Aerosol forcing is defined (Inset 3) as the difference in a quantity, such as the outgoing shortwave flux, without and with aerosols present. A negative forcing in the top-of-the-atmosphere flux for example, means that the outgoing flux is greater with aerosols present than without, and therefore represents a cooling effect. In discussions of aerosol effects a sign convention is adopted such that a positive radiative effect at the TOA indicates addition of energy to the earth-atmosphere system (i.e., a warming influence) whereas a negative effect indicates a net loss of energy (i.e., a cooling influence).

However these influences are not yet well quantified, and uncertainties associated with changes in Earth's radiation budget due to anthropogenic aerosols (radiative forcing) are considered to be the greatest contribution to uncertainty in radiative forcing of climate change over the industrial period (IPCC AR4). Much of the difficulty in quantifying aerosol influences arises from the heterogeneity of aerosol loading and properties: spatial, temporal, size, and composition. This multidimensional heterogeneity stands in marked contrast to the uniform distributions and properties of greenhouse gases and makes the characterization of aerosols and quantification of their influences on climate and climate change extremely challenging.

1.2.1. Direct and indirect effects

Aerosols participate in the Earth's energy budget (**Figure 1.2**) directly by scattering and absorbing radiation (McCormick and Ludwig, 1967; Charlson and Pilat, 1969; Atwater, 1970; Mitchell, Jr., 1971; Coakley et al., 1983) and indirectly by acting as cloud condensation nuclei (that is by serving as

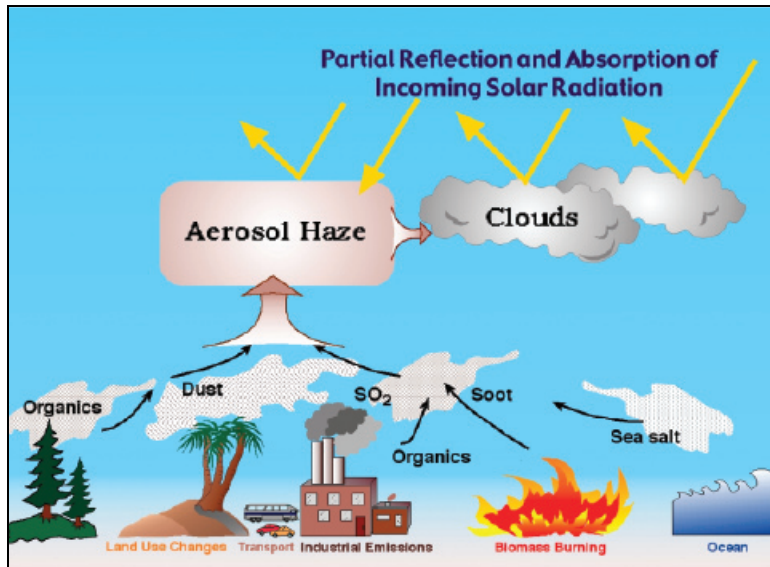


Figure 1.2. Radiative forcing by tropospheric aerosols. Tropospheric aerosols (aerosols in the lower atmosphere) scatter solar radiation; this light scattering exerts a cooling effect on climate by decreasing the absorption of solar radiation. Aerosol particles also increase the brightness and persistence of clouds, exerting a further cooling influence on climate. Increases in aerosols over the industrial period have resulted in a cooling influence on climate change that is opposite to the warming influence of increased concentrations of greenhouse gases.

the particles on which cloud droplets form and grow) and, thereby, affecting cloud microphysical and radiative properties (Gunn and Phillips, 1957; Twomey, 1977; Liou and Ou 1989; Albrecht, 1989). Other things being equal, the greater the number concentration of aerosol particles, the greater the number concentration of cloud drops, and hence the greater the probability of scattering of incident radiation, and hence the brighter the cloud; this effect is commonly referred to as the first aerosol indirect effect, or the Twomey effect. Likewise, other things being equal, the greater the number concentration of cloud drops, the less efficient the formation of precipitation, and hence the greater the persistence of the cloud, and hence the greater the time-average reflectance of solar radiation; this effect is commonly referred to as the second aerosol indirect effect, or Albrecht effect. The direct absorption of radiant energy by aerosols can influence the atmospheric temperature structure and cloud droplet evaporation rate – a phenomenon that has been labeled the “semi-direct effect” (Hansen et al., 1997; Ackerman et al., 2000; Koren et al., 2004).

The potential influences of aerosols on climate were proposed and debated at least several decades ago (Gunn and Philips, 1957; McCormick and Ludwig, 1967; Charlson and Pilat, 1969; Atwater, 1970; Mitchell, 1971; Twomey et al., 1977). However, because of the paucity of aerosol measurements, even the sign of the aerosol effect on global radiation (warming or cooling) was uncertain. Nevertheless, these pioneering studies highlighted the importance of acquiring better information concerning aerosols, and thereby inspired substantial research efforts in the intervening decades.

1.2.2. Anthropogenic aerosol climate forcing

Radiative forcing of climate change by anthropogenic aerosols regained scientific attention in the 1990s (Charlson et al., 1990; 1991; 1992; Penner et al., 1992) followed by the assessment of Intergovernmental Panel on Climate Change (IPCC, 1995 and IPCC, 1996) that first identified anthropogenic aerosol as a climate forcing agent. The Third and Fourth IPCC Assessment Reports concluded that on a global average the sum of direct and indirect TOA forcing by anthropogenic aerosols is negative (cooling) and comparable in magnitude to the positive forcing by anthropogenic GHGs of about 2.4 Wm^{-2} (IPCC, 2001; IPCC, 2007, see **Figure 1.3**). These aerosol forcing assessments have been based largely on model calculations, with scientific understanding designated as “Medium - Low” and “Low”

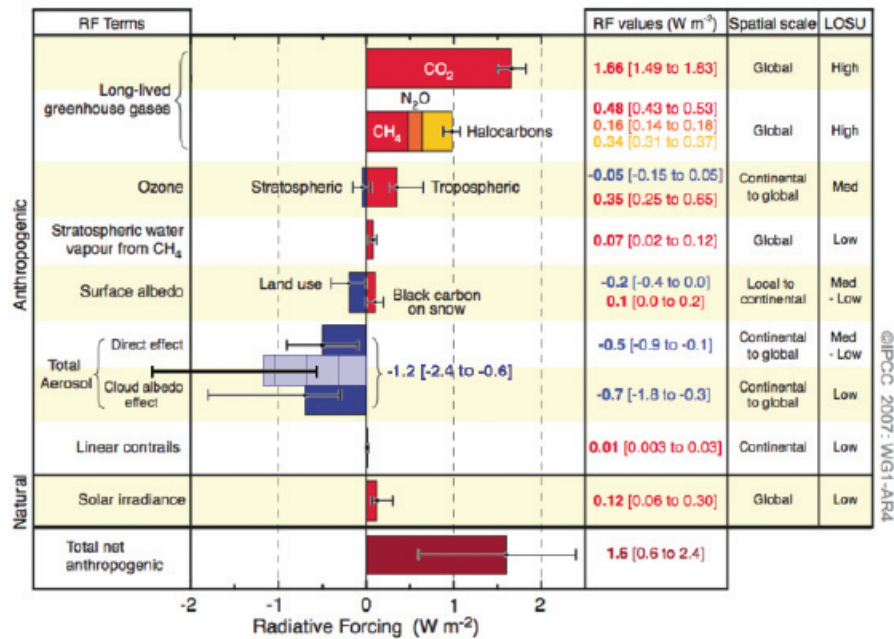


Figure 1.3. Global average radiative forcing (RF) estimates and uncertainty ranges (5-95% confidence interval) in 2005, relative to the pre-industrial climate, for anthropogenic carbon dioxide (CO₂), methane (CH₄), nitrous oxide (N₂O) and aerosols and for other important identified agents and mechanisms, together with the typical geographical extent (spatial scale) of the forcing and the assessed level of scientific understanding (LOSU). Forcings are expressed in units of watts per square meter, $W m^{-2}$. The total anthropogenic radiative forcing and its associated uncertainty are also shown. The figure is modified from IPCC (2007) by addition of a bar for total aerosol forcing (hatched blue) representing the sum of aerosol direct and indirect forcings, and associated uncertainty.

for the direct and indirect climate forcing, respectively. It is also important to recognize that the global-scale aerosol TOA forcing alone is not an adequate metric for climate change (NRC, 2005). Because of aerosol absorption mainly by soot particles, the aerosol direct radiative forcing at the surface could be much greater than the TOA forcing, and the atmospheric radiative heating rate increases. The aerosol climate forcing and radiative effect are characterized by large spatial and temporal heterogeneities due to the wide variety of aerosol sources, the spatial non-uniformity and intermittency of these sources, the short atmospheric lifetime of aerosols, and processing (chemical and microphysical) that occurs in the atmosphere. Over heavily polluted regions, the aerosol forcing can be much stronger than the global average and be far more of an offset for the GHGs warming effect. By realizing aerosol's climate significance and the challenge of charactering highly variable amount and properties of aerosols, the US Climate Change Research Initiative (*CCRI*) has specifically identified research on atmospheric concentrations and effects of aerosols as a top priority (NRC, 2001).

1.3. Reducing uncertainties in estimating aerosol climate forcing

1.3.1. Synergy between observations and models

Over the past decade, significant progress has been made on one hand in measuring aerosol distributions and properties from satellite, ground-based networks, and in-situ field experiments, and on the other hand in developing/improving chemistry transport models that simulate a suite of atmospheric aerosols. Incorporating aerosol representations in the GCM then allows assessment of aerosol climate

effects. Together, through synthesis and integration, observations can be used to improve and constrain model simulations (e.g., Bates et al., 2006; Yu et al., 2006), while the models are indispensable tools for estimating past aerosol forcing and projecting future climate due to changes in atmospheric aerosols (Schulz et al., 2006).

The key to reducing uncertainty in effects of anthropogenic aerosols on climate change is understanding of, and numerically based description of, the processes that contribute to these effects. The geographical distribution of anthropogenic aerosols and the properties of these aerosols depend on emissions of primary particles and precursor gases, on new particle formation and on gas to particle conversion processes and on aerosol dynamical processes, on removal processes, and on transport. These processes are represented in chemical transport models, which must be evaluated by in-situ measurements and by surface- and space-based remote sensing. The requirement is to accurately model the distribution of aerosol mass concentration and size and composition distribution as a function of location and time. There is a further requirement to model the optical properties (and their relative humidity dependence) and the cloud nucleating properties (CCN concentration as a function of supersaturation, and any kinetic limitations). Reduction in uncertainties in aerosol forcing thus requires a coordinated research strategy that will successfully integrate data from multiple platforms (e.g., ground-based networks, satellite, ship, and aircraft) and techniques (e.g., in-situ measurement, remote sensing, numerical modeling, and data assimilation) (Kaufman et al., 2002; Diner et al., 2004; Anderson et al., 2005). The accuracies of current measurements to describing relations between aerosol composition and optical and cloud nucleating properties are not well established; consequently, aerosol forcing has been estimated mainly using modeled mass concentrations and assumed aerosol properties. Model simulations, in turn, rely on the representation of processes of aerosol formation and evolution in the atmosphere, and in particular the estimates of emissions of primary aerosol particles and of precursor gases, which are subject to large uncertainties.

1.3.2. Estimates of emissions

Following earlier attempts to quantify man-made primary emissions of aerosols (Turco et al., 1983; Penner et al., 1993) systematic work was undertaken in the late 1990s to calculate emissions of black carbon (BC) and organic carbon (OC), using fuel-use data and measured emission factors (Liousse et al., 1996; Cooke and Wilson, 1996; Cooke et al., 1999). The work was extended in greater detail and with improved attention to source-specific emission factors in Bond et al. (2004), which provides global inventories of BC and OC for the year 1996, with regional and source-category discrimination that includes contributions from industrial, transportation, residential solid-fuel combustion, vegetation and open biomass burning (forest fires, agricultural waste burning, etc.), and diesel vehicles. Emphasis is on sub-micron sized particles, of greatest relevance to radiative forcing applications.

Emissions of primary aerosols from natural sources—which include wind-blown mineral dust, wild-fires, sea salt, and volcanic eruptions—are less well quantified, mainly because of the difficulties of measuring emission rates in the field and the unpredictable nature of the events. Often, emissions must be inferred from ambient observations at some distance from the actual source. As an example, it was concluded (Lewis and Schwartz, 2004) that available information on size dependent sea-salt production rates could only provide order-of-magnitude estimates. One conclusion from this work is that primary emissions, just like the observed aerosol concentrations, can vary dramatically over space and

time. However, again, progress has been made in modeling these inputs and observing some of them from satellite platforms.

With regard to secondary aerosol production, the emissions of their man-made precursors are in some cases quite well known, e.g., SO_2 emissions for sulfate formation and NO_x emissions for nitrate formation; however emissions of cation precursors, such as NH_3 , Ca, and Mg, are much less well known. Progress has been made at speciating the primary man-made precursors of secondary organic aerosols such as toluene and xylenes (Streets et al., 2003); however, the natural-source precursors of secondary organic aerosols, such as terpene and isoprene, are known at global scale only to within a factor of two (Guenther et al., 2006) and are poorly defined at a particular time and place. Even some of the fundamental mechanisms of secondary organic aerosol formation are not well understood; identifying these mechanisms and quantifying the aerosol production rate as a function of controlling variables is a subject of active research. Understanding of the secondary organic aerosol formation is, however, rapidly evolving. Recent studies by several groups involving field measurements, laboratory studies, and modeling are showing much greater amounts of secondary organic aerosol than were previously recognized, in some instances an order of magnitude or greater.

The difficulties encountered in quantifying present-day aerosol emissions, are magnified when attempting to develop past or future trends. Information for past years on the source types and strengths and even the world regions that dominate emissions are difficult to obtain, and the historical inventories from pre-industrial time to present had to be based on limited knowledge and database. Several studies on historical emission inventories of BC (e.g., Novakov et al., 2003; Ito and Penner 2005; Junker and Liousse, 2006; Bond et al., 2007), SO_2 , and NO_x (van Aardenne et al., 2001; Stern, 2005) have been available in the literature with some similarities and differences among them, but the emission estimates for early times do not have the rigor of the studies for present emissions. One major conclusion from all these studies is that growth of aerosol emissions in the 20th century was not nearly so rapid as the growth in CO_2 emissions. This is because in the late 19th and early 20th centuries, PM emissions were relatively high from the heavy use of biofuels and the lack of particulate controls on coal-burning facilities; however, as economic development continued, traditional biofuel use remained fairly constant and PM emissions were reduced by technological controls. Thus, PM emissions in the 20th century did not grow as fast as CO_2 emissions, as the latter are roughly proportional to total fuel use.

One pressing need is for historical open biomass burning emissions. Great strides in assembling inter-annual estimates of global biomass burning from satellite products have been made (e.g., van der Werf et al., 2003, 2006), but these obviously go back only a short time. Century-scale estimates have been attempted (van Aardenne et al., 2001; Ito and Penner, 2005; Mouillot and Field, 2005; Mouillot et al., 2006), but all researchers acknowledge the great difficulties in being certain of the historical magnitudes and trends. Nevertheless, the patterns of open biomass burning since the industrial revolution will significantly affect aerosol loadings in historical times. Tentative steps have even been taken to estimate historical trends in other natural-source emissions, e.g., Mahowald et al. (2003) on mineral dust emissions, but such work is not yet ready for use by the climate modeling community. The gas phase photochemistry that is responsible for formation of nitric acid is fairly well understood, as is the subsequent fate of HNO_3 – wet and dry deposition and uptake on aerosols, principally by neutralization by ammonia. However emissions inventories of ammonia are subject to great uncertainty historically as well as at present.

Projections of aerosol emissions into the future have been made. Faced with the need to develop future BC and OC emissions for the Third Assessment Report, the IPCC scaled present-day emissions with CO emission forecasts (IPCC, 2001). This was an unsatisfactory approach because of the different factors influencing future emissions of fine particles and CO, particularly the ability to control particle emissions at reasonable cost and the societal imperative of reducing human health effects caused by fine particle inhalation. Forecasts of future BC and OC emissions based on IPCC energy and fuel scenarios have been developed (Streets et al., 2004; Rao et al., 2005) taking care to incorporate the likely future effects of new technology deployment and environmental regulation. The expectation is that global emissions of carbonaceous aerosols will likely remain rather flat or decrease out to 2050. Aerosol emission modelers have been reluctant to venture into the 2050-2100 timeframe on account of the great difficulties of predicting the level of technology application and performance, even if energy modelers can forecast the levels of fuel use by sector. For precursor gases like SO₂, there are many forecasts available; prospective emissions depend strongly on assumptions about future emission controls.

1.3.3. Aerosol representation in GCMs

Representation of the climate influence of atmospheric aerosols has been gradually incorporated into GCMs with increasing sophistication. In the IPCC (1990) report, the few transient climate change simulations that were discussed used only increases in greenhouse gases. By the IPCC (1995) report, although most of the simulations still used only greenhouse gases, the direct effect of sulfate aerosols was added to several models (MPI and UKMO). The sulfate aerosol distribution for 1990 was derived from a sulfur cycle model in both cases (Langner and Rodhe, 1991) with estimated past aerosol emissions, and future aerosol loading followed the IS92a sulfur emission scenario (IPCC, 1992). The aerosol forcing contribution was mimicked by increasing the surface albedo. The primary purpose was to establish whether the pattern of warming was altered by including aerosol-induced cooling in regions of high emissions (such as the Eastern U.S. and eastern Asia), although even then improved agreement with the observational record of global mean temperature in the last few decades was noted.

By the time of the IPCC (2001) report, numerous groups were using aerosols in simulations of both the 20th and 21st centuries. The inclusion of the direct effect of sulfate aerosols was necessary, given the models' climate sensitivity and ocean heat uptake, to reproduce the observed global temperature change. Although most models still represented aerosol forcing by increasing the surface albedo, several groups explicitly represented sulfate aerosols in their atmospheric scattering calculations, with geographical distributions determined by off-line tracer model calculations or by separate GCM aerosol simulations. The first model calculations that included the indirect effect of aerosols were also reported.

The most recent IPCC assessment report (2007) summarized the climate change experiments from some 20 modeling groups which have now incorporated representation of a variety of aerosol species, not just sulfates but black and organic carbon, mineral dust, sea salt and in some cases nitrates as well (see Chapter 3, Table 3.3). In addition, there is a greater realization of the importance of including the indirect effect, in part because with the given model sensitivity, the (better resolved) direct effect is now thought to be insufficient to allow proper simulation of observed temperature changes. As in previous assessments, the aerosol distributions that influence both the direct and indirect effect were produced off-line, as opposed to being run in a coupled mode with the climate change simulations. This is a limitation compared with a fully interactive approach in which climate changes are allowed to change the aerosol distribution and hence the aerosol climate forcing.

The fact that models now use multiple aerosol types and often calculate both direct and indirect aerosol effects does not imply that the requisite optical characteristics of the aerosols, or the mechanisms of aerosol/cloud interactions, are well known. Much research needs to be done before the field will be able to reduce the large uncertainties associated with the modeled aerosol forcing (IPCC, 2007). Additionally, one of the major sources of error lies in estimating the emissions of natural and anthropogenic aerosol and their precursors.

1.4 Contents of this report

This report assesses current understanding of aerosol radiative effects on climate, focusing on developments of aerosol measurement and modeling subsequent to the 2001 IPCC assessment report. The Executive Summary presents an overview of the topics addressed in this report. While providing a chapter by chapter summary of topics addressed, it also summarizes the key concepts that are required for the study of aerosol effects on climate. Chapter 1 (this chapter) presents a easily understandable summary of the topics addressed and introduces the reader to the key concepts in addition to providing a framework for further discussion in these chapters.

Chapter 2 provides an assessment of in-situ and remote sensing measurements of aerosol properties, burdens, and radiative forcing. In particular, it discusses the measurement of aerosol properties and their evolution. It provides an overview of current aerosol measurement capabilities and discusses the synergy of measurements and model simulations. The measurement requirements are discussed in the context of needs for an accurate estimation of aerosol radiative effects and forcing. Inadequacies in current measurement capabilities are addressed including aerosol vertical distributions, direct forcing over land and the lack of accurate aerosol absorption measurements.

Model simulation and estimation of the global and to some extent regional aerosol direct and indirect effects are examined in Chapter 3. In particular, it examines the representations of aerosols that were used in the AR4 runs described in the IPCC (2007) report. The conclusions regarding the emissions and their effects drawn by the IPCC (2007) were based on these runs. These representations are not generally the same as those that were obtained in coupled aerosol-chemistry simulations run with “aerosol models” (in which aerosol sources are prescribed, with transport fields saved off-line from a separate run of the GCM). Similarly, the aerosols in the AR4 runs may differ from those in more recent simulations made with the same models.

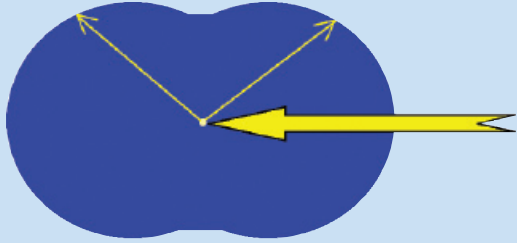

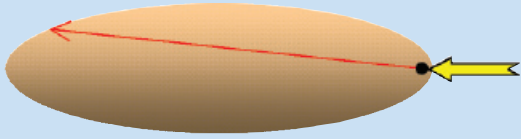

Finally, in Chapter 4 issues and procedures that need to be addressed in obtaining a comprehensive understanding of aerosol effects on climate are identified. Future representation of aerosols in climate models is considered in Chapter 4. A more detailed computation of aerosol/chemistry interactions, better calculations of size distributions, and interactions between aerosols and clouds are only a few of the processes that could be incorporated in the future. However, all of these aspects are limited by the level of understanding, and by the computer time necessary for their calculation. Future climate-change experiments will likely be performed at finer horizontal and vertical resolution in order to resolve regional effects, and this too will require increased computational expense. It is therefore not clear how rapidly improved aerosol processes will be included in climate-change experiments; consequently at least for the next set of IPCC simulations the prime mode of operation may continue to be off-line simulations, and saving of aerosol distributions.

Inset 1: Atmospheric and Aerosol properties

Earth's atmosphere is composed primarily of nitrogen and oxygen with much lesser amounts of minor gases such as carbon dioxide, argon and water vapor, and of suspended aerosol particles and cloud particles. Each of these components interacts with solar (or shortwave) and terrestrial (thermal or longwave) radiation in different ways. Gas molecules having a very small size (0.1 nm) compared to the wavelength of the solar radiation (0.3 to ~5 μm) or the terrestrial radiation (greater than 5 μm) predominantly scatter solar radiation by a process known as Rayleigh scattering (Inset 2, below) at the same frequency, thus producing secondary radiation which has well defined angular characteristics. Rayleigh scattered light intensity depends on the 4th power of the light frequency (reciprocal of the wavelength) with the result that blue light is scattered about 10 times more strongly than red light (thus giving rise to blue color of the sky). In addition, gases absorb light in discrete frequencies throughout the solar spectrum, and more so in the thermal infrared (especially near the surface where their densities are high) giving rise to the well known greenhouse effect. Increase in concentration of these molecules throughout the atmosphere, as is observed in the case of carbon dioxide and other trace gases due to human activities such as fossil fuel burning, enhances the greenhouse warming near the surface and thus provides a source of net heating of the surface and lower atmosphere (positive forcing, Table 1).

Aerosol particles on the other hand are much larger than molecules; they range in size (diameter for spherical particles) from a few nanometers (an aggregate of tens of molecules), a result of the process of nucleation, to tens of micrometers, as in the case of sea-salt and desert dust. Cloud particles are typically larger, up to hundreds of micrometers (even larger for rain clouds) a result of condensation of water vapor on the surfaces of aerosols in an atmosphere that is super-saturated (that is, the relative humidity slightly exceeds one hundred percent). Because the wavelength of light is now of the same order as the characteristic size of the particles (aerosols) or are less than the size of cloud particles, the light interaction with the particles is much more complex and is much stronger (per particle) for both scattering (for spherical particles this is known as Mie scattering) and absorption. Typically in an urban atmosphere the number concentration of aerosol particles tends to be high, with the result that the scattering by particles is of the same order (or higher) as that of the more numerous molecules. Some aerosol particles also absorb light. For example, carbonaceous (soot) particles are black and therefore have a relatively high absorption coefficient (Table 1 for definition). In such a case, when the aerosol particles are highly absorbing, the sign of the forcing attributable to aerosols tends to be the same as in the case of absorbing gases. More often though, aerosols tend to be non-absorbing (sulfates and sea-salt) and therefore reflect sunlight back into space providing cooling of the atmosphere. Indeed much attention was paid to this effect of aerosols in the mid- to late- seventies in studying the consequences of a nuclear war between nations in which studies showed that large scale injection of non-absorbing aerosols into the atmosphere (from nuclear explosions) would increase reflection of sunlight back into space thus cooling the surface – a circumstance that was termed “nuclear winter”.

Inset 2: Molecular and aerosol light scattering and absorption.

<p>Rayleigh or Molecular Scattering: The oscillating electric field of the solar photon induces an oscillating dipole in the molecule at the same frequency, thus emitting secondary radiation which has well defined angular characteristics (blue shaded area in the figure to the right). Rayleigh scattered light intensity depends on the 4th power of the frequency with the result that blue light is scattered about 10 times stronger than red light (thus giving rise to blue color of the sky).</p>	 <p>Phase Function for Rayleigh or Molecular scattering when size of molecule $d \ll \lambda$, wavelength of light, shows that forward scattering and backward scattering have equal probability.</p>
<p>Molecular Absorption: Atmospheric molecules such as carbon dioxide, water vapor and oxygen absorb solar and terrestrial radiation in the visible, near IR or thermal IR at specific wavelengths that correspond to their rotational, vibrational and/or electronic frequencies.</p>	 <p>Molecules such as CO_2 and H_2O have dipole moments and therefore strongly absorb radiation at many frequencies or wavelengths. (Figure not to scale).</p>
<p>Aerosol (Mie) Scattering: When particle sizes are large (comparable to the wavelength of light) the scattering cross-section (probability for scattering) increases dramatically. However there are many more molecules than particles in the atmosphere, with the result that the two effects are comparable.</p>	 <p>Scattering by large particles (aerosols, cloud droplets, shown here as a black dot) whose size d is approximately equal to λ, wavelength of light, is predominantly in the forward direction (red arrow).</p>
<p>Aerosol Absorption: Particles made of sulfates and sea-salt do not absorb light at solar wavelengths. Soot particles (i.e., those that are black), absorb solar and terrestrial radiation. At those wavelengths, the complex refractive index of these particles has a significant imaginary component.</p>	 <p>Particles with a large imaginary part of the refractive index have significant absorption.</p>

Inset 3: Brief description of key aerosol, cloud, and surface properties that determine the aerosol radiative forcing.

- **Aerosol extinction coefficient:** Fraction of radiant flux lost from aerosol scattering and absorption per unit thickness of aerosol, with a unit of m^{-1} .
- **Aerosol Forcing:** of a quantity such as solar irradiance or flux is defined as the difference in the quantity with and without aerosols present. Sometimes aerosol forcing just refers to the industrial period in which case the forcing is the change in quantity calculated with aerosols present during the pre-industrial and industrial periods.
- **Aerosol optical depth (AOD):** a measure of aerosol amount in optical sense. It is an e-folding length of the decrease of a direct beam due to the extinction when traveling through the aerosol layer. Changes of AOD with wavelength are usually represented by the Angstrom exponent, with high values of Angstrom exponent indicative of small particles (industrial pollution and biomass burning smoke) and low values representative of large particles (mineral dust and sea-salt).
- **Aerosol mass extinction (scattering, absorption) efficiency:** the aerosol extinction (scattering, absorption) coefficient per unit aerosol mass concentration, with a unit of $\text{m}^2 \text{g}^{-1}$.
- **Aerosol phase function:** a description of the angular distribution of scattering radiation. In practice, the phase function is parameterized with *asymmetry factor (g)*, with $g=1$ for completely forward scattering and $g=0$ for symmetric scattering. Another relevant parameter is the *hemispheric backscattered fraction (b)*, a fraction of the scattered intensity that is redirected into the backward hemisphere of the particle and can be derived from measurements made with an integrating nephelometer. The larger the particle size, the more the scattering in the forward hemisphere (i.e., larger g and smaller b).
- **Aerosol single-scattering albedo (SSA, ω_0):** a measure of relative importance of scattering and absorption. It is defined as a ratio of the scattering coefficient to the extinction coefficient. The smaller the SSA, the more absorbing the aerosol is.
- **Internal mixture vs external mixture:** Internal mixture is a chemically homogeneous mixture of particles in air, with each particle having about the same chemical composition. For external mixture, individual particles in the aerosol do not have the same chemical compositions or necessarily the same size distribution. The internal mixture has a higher absorption coefficient than the external mixture.
- **Hydrophilic aerosol vs hydrophobic aerosol:** Hydrophilic aerosols (e.g., sulfate, sea-salt) can adsorb water vapor from its surroundings and ultimately dissolve, while hydrophobic aerosols (mineral dust) do not adsorb water vapor from its surroundings and dissolve. Hydrophilic aerosols become larger and more scattered with increasing relative humidity of air.
- **Cloud condensation nuclei (CCN):** Aerosol particles that act as seeds for the formation of clouds through the condensation of water molecules onto their surfaces at low supersaturation. The activation of aerosol particles to CCN depends on the size and chemical composition of particles.
- **Cloud albedo:** Fraction of incident radiant flux reflected by cloud. The cloud albedo depends on the number and size of cloud droplets, and water path. In comparison to clean clouds, polluted clouds have more cloud droplet number and smaller droplet size and are more reflective (i.e., higher cloud albedo).
- **Surface albedo:** Fraction of incident radiant flux reflected by surface. It depends not only on surface type but also on geometry of incident light. In general, land has a larger albedo than ocean (glint-free conditions), and desert has a larger albedo than forest. The larger the surface albedo, the less negative the aerosol radiative effect at the TOA is. The TOA aerosol radiative effect can shift from negative (cooling) over ocean to positive (warming) over bright land, if aerosol is partly absorbing.

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