Hans-Christen Hansson and Jonas Bhend

Abstract

This chapter starts by introducing the complex nature of atmospheric aerosols, their sources, formation and properties and describes how they interact with clouds. This is important background information for discussing how aerosols affect climate, both directly and indirectly by affecting the radiative properties of clouds. The complexity of the aerosol–cloud–climate interaction causes large uncertainty in the projections of future climate. Results from different modelling studies on the European region are presented, and these show that the large spatial and temporal variations in atmospheric aerosol concentrations and properties have large regional differences in their effect on climate. This chapter concludes with an example of a co-beneficial global air quality and climate change mitigation scenario.

24.1 Introduction

This chapter starts by introducing the complex nature of atmospheric aerosols, their sources, formation and properties and describes how they interact with clouds. This is important background information for discussing how aerosols affect climate, both directly and indirectly by affecting the radiative properties of clouds. The complexity of the aerosol–cloud–climate interaction causes large uncertainty in the projections of future climate. Results from different modelling studies on the European region are presented, and these show that the large spatial and temporal variations in atmospheric aerosol concentrations and

properties have large regional differences in their effect on climate. This chapter concludes with an example of a co-beneficial global air quality and climate change mitigation scenario.

24.2 The Basics About Aerosols and Climate

24.2.1 Influence of Aerosols on Climate

Atmospheric aerosols, particles and droplets in the atmosphere influence the Earth's radiation balance directly and indirectly. Aerosols interact directly with incoming solar radiation by scattering and absorbing the sunlight. Aerosols can be observed on satellite images. Saharan dust, for example, is often easily observed in the trade wind belt when transported over the Atlantic towards South America, so are anthropogenic aerosols in the Po Valley (Italy) and in the outflow of air masses from the Chinese mainland. Changes in aerosol concentration or changes in its composition affecting its optical properties influence the radiation balance. The total direct effect of aerosols has been investigated in many studies starting with that by Charlson et al. (1991). Estimates of direct radiative forcing due to anthropogenic

H.-C. Hansson (⊠)
Department of Applied Environmental Science,
Stockholm University, Stockholm, Sweden
e-mail: hc@itm.su.se

J. Bhend Federal Office of Meteorology and Climatology, MeteoSwiss, Zurich, Switzerland 442 H.-C. Hansson and J. Bhend

aerosols range up to about $-1~W~m^{-2}$. The Intergovernmental Panel on Climate Change (IPCC) gave $-0.5 \pm 0.4~W~m^{-2}$ as a best estimate (IPCC 2007).

Clouds have a large effect on the radiation balance. Thus, changes in their albedo or lifetime can have a substantial influence on climate. There are several process chains through which aerosols are suggested to affect cloud albedo and lifetime. The first indirect aerosol effect, also called the Twomey effect, is caused by an increasing number of cloud droplets due to an increasing number of particles resulting in a higher cloud albedo. Marine stratus clouds are the most susceptible to changing particle number (Andreae and Rosenfeld 2008). This effect is well known and is observed by satellite. However, there is large uncertainty in the estimates of the global Twomey effect with a best estimate of -0.7 W m^{-2} (range $-0.3 \text{ to } -1.8 \text{ W m}^{-2}$) (IPCC 2007).

Other indirect effects mainly concern cloud lifetime often as a consequence of the change in droplet size and number described in the context of the Twomey effect. They concern the influence of aerosols on the coalescence processes initiating rain, as for example increased amount of absorbing material evaporating the droplets, effects on ice nuclei formation and other processes in ice or mixed clouds affecting the formation of precipitation. Even though large positive and negative climate forcing caused by these effects has been reported, they are not well known and there is no scientific consensus on their climate effect (Lohmann and Hoose 2009; Stevens and Feingold 2009).

The total aerosol effect, including direct and indirect effects, yields an estimate of $-1.2~W~m^{-2}$ (5–95 % range -0.6 to $-2.4~W~m^{-2}$) since preindustrial times (IPCC 2007). This can be compared to the forcing by carbon dioxide (CO₂) of $1.66 \pm 0.17~W~m^{-2}$ and total greenhouse gas forcing of $2.9 \pm 0.3~W~m^{-2}$ (IPCC 2007). The estimated total aerosol effect is considerable; aerosols counteract almost half of the greenhouse gas-induced warming since preindustrial times. As the aerosols have a limited atmospheric lifetime, their influence is mainly limited to the source region. The regional patterns of climate change can thus be expected to vary widely and in places to be considerably greater than the global average climate change.

The magnitude of the total aerosol climate effect has been shown to be crucial in making better projections of future climate. The present total aerosol effect, estimated at about $-1.2~\rm W~m^{-2}$, is considerable compared to the present greenhouse gas effect, estimated at about $+2.9~\rm W~m^{-2}$; that is, aerosols currently mask more than a third of the greenhouse gas-induced warming. Schwartz et al. (2010) showed that uncertainty in the total anthropogenic aerosol effect dominates uncertainty in projecting future warming, giving a range of $1.5-4.5~\rm ^{\circ}C$ for the projected temperature increase at double the preindustrial $\rm CO_2$ concentration. Considering the

possible regional climate effects at a global temperature increase of 4.5 °C compared to a temperature increase of 1.5 °C, the uncertainty must be seen as very large which would seriously affect the confidence in the projections as well causing a large uncertainty in climate change mitigation strategies.

Box 24.1 Sources of aerosols

Atmospheric aerosols originate from a variety of natural and anthropogenic sources. These can be classified as primary sources that emit aerosols directly and secondary sources that emit precursor gases that form particles in the atmosphere through physical and chemical reactions.

The total mass of the global natural aerosol emissions is dominated by primary sources emitting particles larger than 1 μm (such as desert dust and sea spray). Secondary sources contribute less than 10 % to the global natural aerosol mass with mainly fine particles less than 1 μm in aerodynamic diameter (Kiehl and Rodhe 1995). Although contributing much less in terms of mass, secondary particles contribute a much greater number of particles and hence dominate the aerosol effect on climate. This is because radiation and cloud formation depend more on the number of particles than on the mass.

In contrast to natural emissions, anthropogenic emissions mainly contribute to the fine particle fraction and have increased the global fine particle mass loading considerably since preindustrial times (Table 24.1). It should be noted that although anthropogenic activities are responsible for a large global emission of aerosols, their effect is mostly regional as their atmospheric lifetime is fairly short; only a few days to a week giving a transport range of typically 1000–2000 km (Tunved et al. 2003). This means that the southern Baltic Sea area aerosol is dominated by particles from anthropogenic sources, while natural aerosols dominate the northern Baltic Sea area.

Owing to the multitude of sources and processes involved, measuring natural aerosol emissions is difficult and estimates vary widely (e.g. Kiehl and Rodhe 1995; Andreae and Rosenfeld 2008). As a result, natural emission estimates used in climate models differ considerably, whereas estimates for anthropogenic sources are generally similar across models (Textor et al. 2006). The combined natural and anthropogenic aerosol burden determines the aerosol optical depth, which is measured globally and with high spatial and temporal resolution by satellites.

Table 24.1 Global emission estimates of the major aerosol sources (based on Andreae and Rosenfeld 2008)

Global aerosol	emissions (T	g year ⁻¹)	Total particles	Coarse particles	Fine particles	Natural fine particles	Anthropogenic fine particles
Carbonaceous d	aerosols						
Pri	imary organic	0–2 µm	95		95		
		Biomass burning					54
		Fossil fuel					4
		Biogenic				35	
Bla	ack carbon	0–2 μm	10		10		
		Open burning, bi	ofuel				6
		Fossil fuel					4.5
Sec	condary orga	anic	28		28		
		Biogenic				25	
		Anthropogenic					3.5
Sulphates			200		200		
Bio	ogenic sulph	ates				57	
Vo	olcanic sulph	ates				21	
An	nthropogenic	sulphates					122
Nitrates			18		18	9	9
Industrial dust			100	70	30		30
Sea salt			10,130				
<1	μm				180	180	
1-	16 μm			9,940			
Mineral dust			1,600				
<1	μm				165	165	
1	2.5 μm			496			
2.5	5–10 μm			992			
Total			12,181	11,498	726	492	233

Climate models generally show fair agreement with these measurements (Kinne et al. 2006). European sources affecting the Baltic Sea area are discussed in detail in Chap. 15. The past and projected future emissions in Europe and their effect on regional climate are discussed in Sect. 24.6.

24.2.2 Atmospheric Particle Size Distribution

Atmospheric particles range in size over about five orders of magnitude. Observations of the particle number size distributions show that generally four modes can be identified (see Fig. 24.1). The fine mode particles include nuclei mode particles in the 1–10 nm range, Aitken mode particles from

20-80 nm and accumulation mode particles from 100 to 200 nm. Coarse mode particles are found in the range 1-100 µm. The nuclei mode emerges when new particles form, that is nucleation. Nuclei-mode-sized particles also emerge as primary particles from high-temperature combustion (such as engines), and 10-nm sea spray particles have even been observed. The Aitken mode consists of particles grown out of nuclei mode due to coagulation and condensation of condensable atmospheric gases originating from gaseous emissions and gas phase reactions in the atmosphere. Further growth into accumulation-mode-sized particles requires other processes such as liquid phase processes in clouds. Liquid phase chemistry occurs when the absorption of atmospheric gases into cloud droplets forms a very reactive environment, for example sulphur dioxide (SO₂) is mainly oxidised to sulphate (SO₄) in cloud droplets. Primary particles are observed in all size fractions, but in the sub-micron

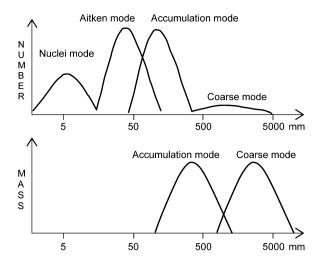


Fig. 24.1 Schematic illustration of particle number and mass distribution with particle size for atmospheric aerosol. Number is dominated by sub-micron particles in the size range 10–200 nm, observed mainly in three modes depending on source and formation process. Particle mass is mainly found in larger sub-micron and super-micron particles

size range are mainly from high-temperature processes and in the coarse mode mainly from mechanical processes.

Not only do emissions influence the number of particles in different size fractions, but sink processes are also important. Although diffusion losses are an important sink process, mainly occurring through collision with other particles or droplets, precipitation (rain or snow) is the main sink for atmospheric particles larger than about 50-70 nm. Such processes affect the fine mode particles mainly through in-cloud scavenging where particles act as cloud condensation nuclei and eventually get scavenged in a precipitating cloud. Whether a particle acts as a cloud condensation nucleus depends on its size as well as its chemical composition. Generally, particles in the upper Aitken size range and larger with a major fraction of hygroscopic compounds such as inorganic salts are good condensation nuclei in warm clouds. In ice or mixed clouds, the ice and snow formation processes are still not well known and information on which particles act as good ice nuclei is poor. Formation of ice crystals in a cloud strongly influences the onset of precipitation. For coarse particles, losses due to sedimentation start to become significant at about 5 µm.

The atmospheric particle size distribution reflects the dynamic interaction of sources, atmospheric formation processes and sinks mixing natural and anthropogenic components. Different air masses can thus have very different particle size distributions depending on the sources over which they had passed and the meteorology they had experienced. Therefore, it is not possible to identify a specific European size distribution, but rather a more or less scavenged pollution aerosol mixed with natural emissions.

24.2.3 Formation of Aerosols in the Atmosphere

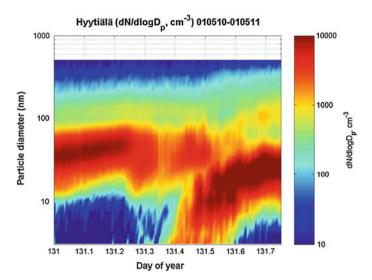
Aerosols are formed in the atmosphere either through new particle formation, condensation onto already existing particles or in-cloud liquid processes. New particle formation, often called nucleation, is the process by which molecules form unstable clusters that grow over a threshold size to form a stable particle. The threshold size is about 1–2 nm. Nucleation is followed by strong condensation causing the newly formed particles to grow rapidly in diameter by several nanometres per hour (Vehkamäki and Riipinen 2012). Nucleation has been studied extensively in the boreal forest. During a 15-year period of measurements, major nucleation events occurred about 100 times a year (Kulmala et al. 2004). The boreal forest that almost completely surrounds the Baltic Sea is thus one of the most important sources of natural aerosol in the area.

A nucleation event, as seen in Fig. 24.2, is clearly visible in the particle size distribution measurements. In the morning, the aerosol is dominated by 50–70 nm particles, which slowly disappear over the course of the morning. Particles of 3 nm in diameter start to form around 10 a.m. and begin to grow. By 3–4 p.m., the particles have grown to about 30–40 nm.

Much attention has been paid to the influence of nucleation on aerosol particle size distribution and its implication for the number of cloud condensation nuclei, especially within the European framework (see the EUCAARI project; Kulmala et al. 2011). The main results of the studies were as follows:

- Nucleation is a major source of aerosol particle number concentrations, and usually, several tens of per cent of sub-micron aerosol particles have originated from condensation of atmospheric vapours—thus being of secondary origin. Roughly, half of the number of aerosol particles in the European boundary layer has originated from nucleation.
- Secondary aerosol formation is due to both natural and anthropogenic influence: while anthropogenic sulphate emissions are a major factor governing formation of new particles, natural emissions of biogenic organic vapours play an important role in defining aerosol size distributions and the climatic impact of aerosols. The results suggest that the anthropogenic contribution (both primary and secondary) dominates in most parts of Europe, the biogenic component being of less importance. However, halving SO₂ and anthropogenic primary particle emissions would only result in reductions of the order of 20 % on the total particle number concentrations—which might suggest that natural aerosol production could compensate somewhat for the reduction in anthropogenic aerosol production.

Fig. 24.2 A nucleation event occurring just before noon 10 May 2001 at Hyytiälä, Finland (Kulmala et al. 2004)



It is clear that reductions in anthropogenic primary (e.g. black carbon, also called 'soot') and secondary precursors (e.g. SO₂ and ammonia, NH₃) will not result in proportional changes in the number and size of the particles. The natural production and formation of particles might increase to reach some balance defined by atmospheric chemical and physical conditions.

24.2.4 Aerosol-Cloud Interactions

Atmospheric particles are needed as condensation sites for water vapour to form clouds. Particle size and chemistry influence the number of cloud droplets and thus the development of a cloud. Particles forming cloud droplets are strongly affected by the cloud either by being precipitated or by being transformed by fast liquid phase chemistry with absorbed gases changing the chemistry and increasing the mass of particles emerging from the dissipating cloud.

Not all particles are effective as cloud condensation nuclei, which require a certain amount of soluble hygroscopic compounds to grow into a cloud droplet (Pruppacher and Klett 1997). Such compounds are typically inorganic salts such as sulphates, nitrates and chlorides. Organic compounds are typically less hygroscopic and thus less efficient in providing good cloud condensation nuclei (Laaksonen et al. 1998; Swietlicki et al. 2008). In non-precipitating clouds, particles form droplets, pass through the cloud and exit the cloud, mainly at the top of the cloud, where the water evaporates.

Clouds even at low latitudes often contain ice crystals. The mix of droplets and ice crystals in clouds complicates the physical and chemical processes considerably and affects the formation of precipitation. Research on mixed phase or pure ice clouds is quite complicated, and understanding is

not well developed. At present, dust seems to be the most important ice nucleus, while there is disagreement concerning black carbon. Biological particles such as bacteria are good ice nuclei but are often present in low numbers and so are minor ice nuclei contributors (Stratmann et al. 2010).

In conclusion, clouds affect the aerosol both in terms of formation, adding more mass to cloud condensation nuclei through liquid phase chemistry, and as the major sink of submicron particles. Particles affect cloud albedo and possibly cloud lifetime. However, information on the specific processes and their general effect on lifetime in, for example, mixed clouds is still limited. A better understanding of these processes and their incorporation in climate models is crucial for advancing climate projection (and hindcasting) capability.

24.3 Aerosol Processes Affecting Climate

24.3.1 Direct Effects

The direct particle effect on climate forcing is caused by scattering and absorption of sunlight. Both scattering and absorption are strongly dependent on particle size but also on the chemical composition of the particles. The total forcing effect is estimated at -0.5 ± 0.4 W m⁻² (IPCC 2007); scattering accounting for -0.7 W m⁻² and absorption for +0.25 W m⁻². However, there has been strong debate on the absorption estimate. Ramanathan and Carmichael (2008) argued that absorption is underestimated and a more accurate global estimate could be as large as +0.9 W m⁻² due to black carbon aerosol alone. While Quaas et al. (2009) using several global models estimated the direct effect at -0.4 ± 0.2 W m⁻², the EUCAARI project claimed to have narrowed the range

further to $-0.2 \pm 0.1~\rm W~m^{-2}$ (Kulmala et al. 2011). In an AEROCOM exercise involving nine global climate models, Schulz et al. (2006) found a total direct forcing in the range $-0.2 \pm 0.2~\rm W~m^{-2}$ including a warming by black carbon in the range $+0.2 \pm 0.15~\rm W~m^{-2}$. These findings indicate that the direct effect is relatively small, but Quaas et al. (2008) derived an estimate of the direct effect of $-0.9 \pm 0.4~\rm W~m^{-2}$ from satellite measurements. However, they recognised the discrepancy and suggested that it may be partly that satellite retrievals of aerosols are not available over bright surfaces such as deserts, snow- or ice-covered surfaces and low-level clouds where the direct forcing may even be positive. They suggested applying a reduction of 30–60 % to the satellite-based estimate, which brings the observations and model results quite close.

An important source of uncertainty in the total direct aerosol effect is the uncertainty in relative humidity, spatially and temporally, as atmospheric particles are generally hygroscopic. In measurements of how the atmospheric aerosol increases in size with increasing humidity, it is usually found that a dominating number of the particles is growing by about 30–50 % in diameter; that is, a factor of 2–3 in volume at 90 % relative humidity while scattering increases by a factor of about 3 compared to dry particles (Swietlicki et al. 2008). Growth is increasingly sensitive to relative humidity, for example, increasing humidity from 90 to 95 % results in an additional increase in scattering of roughly 30 % (Zieger et al. 2010).

When air pollution spreads to the top of the boundary layer, relative humidity usually increases and particles grow and scatter more light back to space. This strongly affects the aerosol optical depth, which is measured by sun photometers from satellites and on the ground generating an extensive database of observations across the globe that is used to control climate models. Errors in estimates of relative

humidity thus strongly affect estimates of the direct effect of particles on climate. To ensure good model performance, comparisons with observation of relative humidity and aerosol optical parameters are necessary.

24.3.2 Indirect Effects

At least six aerosol-cloud interactions have been identified that indirectly affect climate. Table 24.2 summarises processes discussed in the literature (Lohmann and Feichter 2005), with newer estimates from Lohmann et al. (2010).

The first indirect aerosol effect affects cloud albedo through the increase in cloud condensation nuclei due to anthropogenic emissions. The most obvious evidence of the Twomey effect (see Table 24.2) is ship tracks easily observable from space. The ship tracks are white narrow cloud streaks resulting from ship emissions.

According to the IPCC (2007), the best estimate of a global climate effect due to the first indirect effect is about -0.7 W m^{-2} with uncertainty giving a range of -0.3 to -1.8 W m⁻². Lohmann et al. (2010) suggested $-0.9 \pm 0.4 \text{ W m}^{-2}$ which is close to the $-0.7 \pm 0.5 \text{ W m}^{-2}$ proposed by Kulmala et al. (2011). Quaas et al. (2008) found that satellite measurement showed considerably less effect, giving an estimate of -0.2 ± 0.1 W m⁻² for the first indirect effect. They recognised this to be considerably lower than most models but consistent with estimates from models constrained by satellite observations of clouds in terms of cloud droplet number, cloud liquid water path and cloud top temperature (Lohmann and Lesins 2002; Quaas et al. 2006). However, Penner et al. (2011) argued that satellite measurements underestimate the first indirect effect by a factor of 3-6 because they typically use the present-day relationship between observed cloud drop number concentrations and

Table 24.2 Summary of different indirect aerosol effects on climate (based on Lohmann and Feichter 2005; Lohmann et al. 2010)

Effects	Cloud type	Description	Forcing, W m ⁻²
First indirect aerosol effect ('Twomey effect')	All clouds	The more numerous smaller cloud particles reflect more solar radiation	-0.9 ± 0.4
Second indirect aerosol effect ('Albrecht affect')	All clouds	Smaller cloud particles decrease precipitation efficiency, prolonging cloud lifetime	Uncertain
Semi-direct effect	All clouds	Absorption of solar radiation by black carbon may cause evaporation of cloud particles	Uncertain
Glaciation indirect effect	Mixed ice and liquid clouds	More ice nuclei increase precipitation efficiency	Uncertain
Thermodynamic effect	Mixed ice and liquid clouds	Smaller cloud droplets delay the onset of freezing	Uncertain
Riming indirect effect	Mixed ice and liquid clouds	Smaller cloud droplets decrease riming efficiency	Uncertain
Total anthropogenic aerosol effect	All cloud types	Includes the above-mentioned indirect effects plus the direct aerosol effect	0 to -1.8

aerosol optical depths, and these are not valid for the preindustrial values of droplet numbers.

Other effects mentioned in Table 24.2 are suggested to affect cloud lifetime. As clouds generally cool the climate by having a higher albedo than the Earth's surface, shorter cloud lifetime will warm the climate, while longer cloud lifetime will cool the climate. The second indirect aerosol effect (also referred to as the Albrecht effect) is, like the Twomey effect, based on the premise that the number of cloud droplets will increase with an increasing number of available cloud condensation nuclei. In contrast to the first indirect effect, the second indirect effect concerns the processes initiating precipitation. The onset of precipitation is sensitive to the formation of a few big droplets, also referred to as precipitation embryos (Albrecht 1989). Ice nuclei are similar precipitation embryos considered to be crucial for the onset of precipitation.

Black carbon is a strong light absorber and when enclosed in cloud droplets might cause evaporation and thus dissipate the cloud prematurely. This process is often referred to as the semi-direct effect. However, studies show that partial evaporation causes multiple effects as higher albedo due to smaller droplets and fewer giant droplets suppress precipitation both with a negative forcing on climate. A review by Koch and Del Genio (2010) indicated that the semi-direct effect is most likely to produce/account for a slight negative forcing that could be large enough to eliminate the direct warming of black carbon.

The glaciation effect refers to the formation of ice nuclei in cold clouds, that is clouds at least partially containing ice crystals or frozen droplets. In cold clouds, formation of ice crystals is important for the formation of precipitation. Anthropogenic emissions enhancing the number of good ice nuclei might then increase the precipitation probability and thus decrease cloud lifetime resulting in a positive radiative forcing. Black carbon particles have been suggested to be good ice nuclei, but other reports have concluded differently (Stratmann et al. 2010). Dust particles, on the other hand, are found to be important ice nuclei, while the fraction of anthropogenic dust is very difficult to estimate. Hoose et al. (2008) investigated the effect of assuming black carbon to have favourable ice nuclei properties and found that the positive increase in ice nuclei from black carbon particles was counteracted by dust particles losing their ice nuclei capability due to a coating of anthropogenic inorganic salts. These findings highlight the complexity of the processes and the need for a better understanding of ice nuclei properties and the key processes controlling cloud lifetime.

Other processes taking place within mixed ice and liquid water clouds are the thermodynamic effect and the riming effect, both connected to the competition between ice crystals and water droplets for condensing water in the cloud. The World Meteorological Organization International Aerosol Precipitation Science Assessment Group (Levin and Cotton 2007) concluded that observations and understanding of how precipitation is affected by pollution are still lacking. Rosenfeld et al. (2008) stated that large concentrations of man-made aerosols have been shown to affect precipitation and suggested that through different radiative and cloud-mediated processes, aerosols affect the thermodynamics driving the formation of clouds and precipitation. Stevens and Feingold (2009), however, recognised the inability to detect the specific effects of aerosols on precipitation and proposed that the difficulty in disentangling relationships among the aerosol, clouds and precipitation reflects the inadequacy of existing tools and methodologies as well as a failure to account for processes that buffer cloud and precipitation responses to aerosol perturbations.

In conclusion, it is clear that the indirect climate effects of anthropogenic atmospheric aerosols are not well known. Uncertainty in describing the indirect climate effect therefore dominates the uncertainty in total aerosol forcing.

24.4 Aerosol Influence on Regional Climate

Aerosols, including black carbon particles, and ozone are air pollutants that influence climate forcing and possibly also the hydrological cycle. Particles have a short lifetime in the atmosphere, typically two to four days, giving them a transport range of 1000-2000 km (Tunved et al. 2003). This means that the southern Baltic Sea area is dominated by anthropogenic emissions from the central European continent, while the northern Baltic Sea area is dominated by natural aerosol sources (such as the boreal forests). Particle concentration decreases with distance from the source, initially due to dilution and later due to sinks such as precipitation. However, secondary processes such as condensation of organic components and cloud chemistry add mass to the aerosols, increasing their influence on climate relevant processes and so extending the range of climate influence. Tropospheric ozone has a longer atmospheric lifetime of about a month thus giving it time to spread through the whole hemisphere.

The radiation effects of air pollution were measured directly in the Asian Brown Cloud Study and showed strong heating effects due to light-absorbing black carbon (Ramanathan et al. 2005). The simulated annual mean surface heat budget from 10 S to 30 N and 60 E to 100 E, that is over southern India and the Indian Ocean, shows progressive dimming since the 1930s. The dimming is about –15 W m⁻², meaning that 15 W m⁻² less heat is reaching the ground and is instead trapped within the atmosphere by heat-absorbing aerosols. Besides redistributing heat within the atmosphere (affecting the surface and atmospheric

448 H.-C. Hansson and J. Bhend

temperature), evaporation of water and thus atmospheric water vapour concentration are also affected.

The heating and cooling effects of aerosols imply a complicated mix of interacting atmospheric processes that affect radiation and heat transfer which in turn affect regional temperature and the hydrological cycle.

European anthropogenic emissions of air pollutants have varied strongly over time, changing with population growth, major changes in land use and industrialisation. Emissions grew rapidly through the mid-twentieth century (Fig. 24.3) and in London were responsible for the Great Smog of December 1952 that resulted in many thousands of premature deaths. Banning the use of coal for residential heating and introducing high stacks for major industrial sources, both elements of the UK 1956 Clean Air Act helped to reduce local air pollution but in doing so caused problems elsewhere, particularly 'acid rain'. Acid rain—a potent mix of acidifying gases and particles—travelled long distances and when it eventually came down damaged surface water, groundwater and forests soils.

The adoption of the Convention on Long-range Transboundary Air Pollution (CLRTAP) in 1979 has resulted in a strong reduction in sulphur emissions such that present emissions are now back to pre-war levels (Fig. 24.3). Present

emissions are now about 25 % of those in 1990. European emissions of black carbon decreased by about 40 % between 1960 and 2000 (Fig. 24.4).

Clearly, there have been very large variations in European emissions over the past 50 years, especially for sulphur, and this should be reflected in the regional climate if anthropogenic aerosols are indeed strong climate forcers, as has been suggested by the IPCC (2007) for example.

Using a coupled atmosphere-ocean climate model (GISS-ER) to investigate the response to forcing imposed over different latitude bands, Shindell and Faluvegi (2009) showed that the mid-latitudes and polar regions are sensitive to the source location. Although mid-latitude emissions mainly affect this region, they also affect the tropics. Furthermore, the authors also concluded that decreasing sulphur emissions and increasing black carbon have substantially contributed to the rapid warming observed in the Arctic.

Booth et al. (2012) using a state-of-the-art Earth system climate model (HadGEM2-ES) showed that aerosol emissions and periods of volcanic activity explain 76 % of the simulated multi-decadal variance in detrended 1860–2005 North Atlantic sea surface temperatures. The variation in sea surface temperature mainly depends on the indirect aerosol-cloud climate effects. The authors concluded with a strong

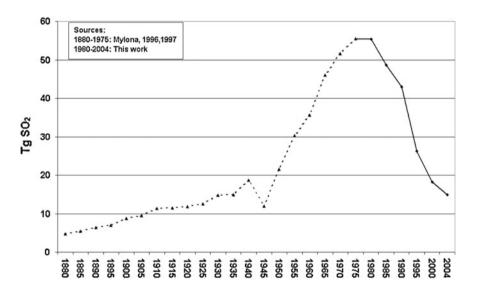


Fig. 24.3 Historical development of sulphur dioxide (SO₂) emissions in Europe (Vestreng et al. 2007)

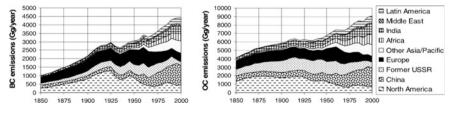


Fig. 24.4 Emissions of (left) black carbon and (right) organic carbon. Emissions are segregated by region (Bond et al. 2007)

statement based on their findings: 'Our findings suggest that anthropogenic aerosol emissions influenced a range of societally important historical climate events such as peaks in hurricane activity and Sahel drought. Decadal-scale model predictions of regional Atlantic climate will probably be improved by incorporating aerosol—cloud microphysical interactions and estimates of future concentrations of aerosols, emissions of which are directly addressable by policy actions'.

Regional models allow higher spatial resolution than global models and a more detailed description of processes but need set boundary conditions specifying the influence of climate, meteorology and air components outside the model domain. Zubler et al. (2011) used a regional climate model that included interactive aerosol and cloud physics to investigate surface shortwave radiation in Europe over the past 50 years. This period is characterised by 'global dimming' until the mid-1980s and then 'brightening' and was fairly well described by the model in terms of the variation in aerosol emissions. The main cause of the brightening/dimming appeared to be the varying cloud fraction, that is the aerosol effect on cloud lifetime. Huszar et al. (2012) used an interactive coupling of a regional model (RegCM3) and a chemistry transport model (CAMx) to study the interaction between regional climate and air pollutants and found a perturbation of temperature in the range −1.5 to +1.5 °C due to aerosols and ozone. Interestingly, they did not find any correlation between forcing and induced temperature changes, which indicates the complexity of the climate system. However, they did identify methodological difficulties with regional models in applying realistic boundary conditions.

It seems possible that climate forcing by air pollutants strongly affects regional climate and thus global climate not only increasing global temperature but also causing changes in large-scale circulation. Even though the total anthropogenic forcing implies a warmer future climate in Europe, changing meteorology will also cause spatial variability. The main uncertainty in global climate models seems due to poor understanding of aerosol-cloud interactions. This uncertainty strongly affects the ability to make climate projections. Furthermore, there are still difficulties in achieving the necessary spatial resolution. Although the resolution of current regional climate models should be good enough, according to Huszar et al. (2012) and Zubler et al. (2011), there are methodological problems in adjusting the boundary conditions for changes in regional forcing on the general meteorology that in turn affect the boundary conditions.

24.4.1 Baltic Sea Area

The annual mean temperature in Sweden increased by about 0.9 °C between 1990 and 2005 (see climate records at

www.smhi.se), which coincided with a major decrease in European sulphur emissions (see Fig. 24.3). This increase in temperature is about double that observed in the northern hemisphere over this period. The modelling studies discussed in Sect. 24.4 show that it is still difficult to determine to what extent the observed warming in Scandinavia is caused by global climate change and to what extent it is caused by the strong regional decline in aerosol concentrations. However, the Baltic Sea area is on one of the major outflow paths of air pollutants from the European landmass, and so it is feasible that a substantial fraction of the observed warming is due to the strong decline in sulphur emissions over the last 20 years. Black carbon emissions have also decreased over this period although substantially less than for sulphur. Some of the warming might have been offset by the decreasing black carbon.

Precipitation in Sweden seems to have increased between 1990 and 2005, but it is not statistically significant compared to 1960–1990 (see climate records at www.smhi.se). Current climate models have considerable uncertainties in describing precipitation and its dependence on aerosols, which complicates discussions on the influence of air quality on precipitation. A better understanding of aerosol–cloud interactions is needed for climate models to better simulate observations, not only for precipitation but also for aerosols, gases, clouds and other parameters affecting precipitation (see Chap. 10).

24.5 Climate Change Mitigation by Air Quality Mitigation

Air pollution contains components that both heat the climate (black carbon and ozone) and cool the climate (particulates). Emission abatement measures to mitigate air pollution, thus risk counteracting climate change mitigation. The UNEP Integrated Assessment of Black Carbon and Tropospheric Ozone (UNEP 2011) was performed on a mixture of available abatement measures (see Table 24.3) considered to be commonly available and possible to implement. The measures focus on black carbon and methane emissions, but coemitted compounds such as organic carbon and ozone precursors are also considered in the analysis.

The impact of air pollutants on global climate and air quality was investigated using two different and well-established global climate models—ECHAM and GISS (Shindell et al. 2012). Besides the measures mentioned in Table 24.3, CO₂ abatement was assumed such that a maximum atmospheric concentration of 450 ppm of CO₂ would be reached, which assumes CO₂ emissions to stabilise by 2020 and then decrease to zero by 2080. The investigation was performed to evaluate the impact of black carbon and methane abatement measures in the context of a very

Table 24.3 Measures that improve climate change mitigation and air quality and that have a large emission reduction potential (Shindell et al. 2012; UNEP 2011)

Measure	Sector	
Methane measures		
Extended premine degasification and recovery and oxidation of methane from ventilation air from coal mines	Extraction and transport of	
Extended recovery and utilisation, rather than venting, of associated gas and improved control of unintended fugitive emissions from the production of oil and natural gas	fossil fuels	
Reduced gas leakage from long-distance transmission pipelines		
Separation and treatment of biodegradable municipal waste through recycling, composting and anaerobic digestion as well as landfill gas collection with combustion utilisation	Waste management	
Upgrading primary wastewater treatment to secondary/tertiary treatment with gas recovery and overflow control	_	
Control of methane emissions from livestock, mainly through farm-scale anaerobic digestion of manure from cattle and pigs	Agriculture	
Intermittent aeration of continuously flooded rice paddies		
Black carbon technological measures (affecting BC and other co-emitted compounds)		
Diesel particle filters for road and off-road vehicles as part of a move to worldwide adoption of Euro 6/VI standards	Transport	
Introduction of clean-burning biomass stoves for cooking and heating in developing countries ^{a,b}	Residential	
Replacing traditional brick kilns with vertical shaft and Hoffman kilns	Industry	
Replacing traditional coke ovens with modern recovery ovens, including the improvement of end-of-pipe abatement measures in developing countries	_	
Black carbon regulatory measures (affecting BC and other co-emitted compounds)		
Elimination of high-emitting vehicles in road and off-road transport	Transport	
Ban on open burning of agricultural waste ^a	Agriculture	
Substitution of clean-burning cook stoves using modern fuels (LPG or biogas) for traditional biomass cook stoves in developing countries ^{a, b}	Residential	

^a Partly motivated by its effect on health and regional climate including areas of ice and snow

^b For cooking stoves, given their importance for black carbon emissions, two alternative measures are included

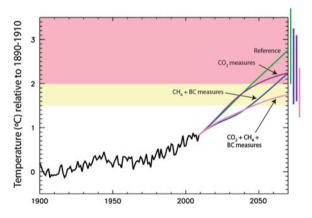


Fig. 24.5 Observed change in mean global temperature to 2009 and projected change in mean global temperature under various future scenarios, all relative to the 1890–1910 mean. *Shaded areas* show zones beyond 1.5 °C (*yellow*) and 2 °C (*pink*) (UNEP 2011)

ambitious CO₂ abatement programme. Figure 24.5 shows that abatement measures for black carbon and methane could reduce the projected global warming by 2070 by 0.5 °C. However, abatement of these compounds, often referred to

as short-lived climate forcers, will only delay the rise in global warming if CO₂ emissions continue unchecked.

Abatement of CO_2 emissions is critical for limiting the rise in global temperature, but the implementation of abatement measures for black carbon and methane could also make a significant contribution. As the climate response to changes in the emissions of these short-lived climate forcers is relatively fast, reductions in the emission of black carbon and methane can be used to slow the immediate rise in global temperature and limit the level at which it would stabilise due to CO_2 emissions alone.

24.6 Conclusion

The recently revised Gothenburg Protocol to the Convention on Long-range Transboundary Air Pollution (CLRTAP) should ensure a decline in emissions of sulphur and nitrogen compounds over the next few years (see www.unece.org/env/lrtap). By 2020, emissions from the EU are required to have decreased by 59 % for SO₂, 42 % for nitrogen oxides (NO_X),

6 % for NH_3 and 28 % for volatile organic compounds (VOC) relative to 2005. Simultaneously, particle emissions ($PM_{2.5}$) must decrease by 22 %, with a voluntary emphasis on black carbon. Black carbon emissions will decrease considerably with the introduction of the new EU emission standards for vehicle and truck diesel engines (EURO 5 and EURO 6) that will take effect during the next 10–15 years as the transport fleet is gradually renewed. However, the major black carbon emission is from furnaces and stoves, for which abatement plans have not been introduced.

The major changes in emissions of sulphur and nitrogen compounds are thus already complete, while black carbon will probably decrease far more, as will organic compounds emitted from the same combustion sources. As a result, the total climate effect from the expected changes in aerosol emissions will probably be minor (Kulmala et al. 2011), while emission reductions strongly affecting ozone and methane concentrations may reduce climate warming significantly. However, ozone and methane concentrations over Europe and the Baltic Sea area are strongly dependent on emissions over the northern hemisphere and the world as a whole, respectively.

In conclusion, analyses on regional aerosol effects in northern Europe are rare and the commonly used regional climate models are mostly unable to simulate aerosol-climate interactions. However, recent modelling efforts investigating the influence of European aerosol emissions indicate an effect on large-scale circulation over Europe that is very likely to have affected the climate in the Baltic Sea region. To what extent is still not known. Development of the modelling capability and targeted analyses is urgently needed to reduce uncertainties related to the effect of changes in aerosol concentration on regional climate.

Open Access This chapter is distributed under the terms of the Creative Commons Attribution Noncommercial License, which permits any noncommercial use, distribution, and reproduction in any medium, provided the original author(s) and source are credited.

References

- Albrecht BA (1989) Aerosols, cloud microphysics, and fractional cloudiness. Science 245:1227-1230
- Andreae MO, Rosenfeld D (2008) Aerosol-cloud-precipitation interactions. Part 1: The nature and sources of cloud-active aerosols. Earth-Sci Rev 89:13-41
- Bond TC, Bhardwaj E, Dong R, Jogani R, Jung S, Roden C, Streets DG, Trautmann NM (2007) Historical emissions of black and organic carbon aerosol from energy-related combustion, 1850-2000. Global Biogeochem Cy 21:GB2018 doi: 10.1029/2006GB002840
- Booth BBB, Dunstone NJ, Halloran PR, Andrews T, Bellouin N (2012) Aerosols implicated as a prime driver of twentieth-century North Atlantic climate variability. Nature 484:228-232

- Charlson RJ, Langner J, Rodhe H, Leovy CB (1991) Perturbation of the Northern Hemisphere radiative balance by backscattering from anthropogenic sulfate aerosols. Tellus A 43:152-163
- Hoose C, Lohmann U, Erdin R, Tegen I (2008) The global influence of dust mineralogical composition on heterogeneous ice nucleation in mixed-phase clouds. Environ Res Lett 3:025003 doi:10.1088/1748-9326/3/2/025003
- Huszar P, Miksovsky J, Pisoft P, Belda M, Halenka T (2012) Interactive coupling of a regional climate model and a chemical transport model: evaluation and preliminary results on ozone and aerosol feedback. Clim Res 51:59–88
- IPCC (2007) The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, Cambridge, UK and New York, USA
- Kiehl JT, Rodhe H (1995). Modelling geographical and seasonal forcing due to aerosols. In: Charlson RJ, Heintzenberg J (eds), Aerosol Forcing of Climate. J. Wiley and Sons Ltd, p 281-296
- Kinne S, Schulz M, Textor C et al (2006) An AeroCom initial assessment - optical properties in aerosol component modules of global models. Atmos Chem Phys 6:1815-1834
- Koch D, Del Genio AD (2010) Black carbon semi-direct effects on cloud cover: review and synthesis. Atmos Chem Phys 10:7685–7696
- Kulmala M, Vehkamaki H, Petaja T, Dal Maso M, Lauri A, Kerminen VM, Birmili W, McMurry PH (2004) Formation and growth rates of ultrafine atmospheric particles: a review of observations. J Aerosol Sci 35:143-176
- Kulmala M, Asmi A, Lappalainen HK et al (2011) General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales. Atmos Chem Phys 11:13061-13143
- Laaksonen A, Korhonen P, Kulmala M, Charlson RJ (1998) Modification of the Kuhler equation to include soluble trace gases and slightly soluble substances. J Atmos Sci 55:853-862
- Levin Z, Cotton W (2007) Aerosol Pollution Impact on Precipitation: A Scientific Review. Report from the WMO/IUGG International Aerosol Precipitation Science Assessment Group (IAPSAG) (World Meteorological Organization, Geneva, Switzerland
- Lohmann U, Feichter J (2005) Global indirect aerosol effects: A review. Atmos Chem Phys 5:715-737
- Lohmann U, Lesins G (2002) Stronger constraints on the anthropogenic indirect aerosol effect. Science 298:1012-1015
- Lohmann U, Hoose C (2009) Sensitivity studies of different aerosol indirect effects in mixed-phase clouds. Atmos Chem Phys 9:8917-8934
- Lohmann U, Rotstayn L, Storelvmo T et al. (2010) Total aerosol effect: radiative forcing or radiative flux perturbation? Atmos Chem Phys 10:3235-3246
- Quaas J, Boucher O, Lohmann U (2006) Constraining the total aerosol indirect effect in the LMDZ and ECHAM4 GCMs using MODIS satellite data. Atmos Chem Phys 6:947-955
- Quaas J, Boucher O, Bellouin N, Kinne S (2008) Satellite-based estimate of the direct and indirect aerosol climate forcing. J Geophys Res 113:D05204 doi: 10.1029/2007JD008962
- Quaas J, Ming Y, Menon S et al (2009) Aerosol indirect effects general circulation model intercomparison and evaluation with satellite data. Atmos Chem Phys 9:8697–8717
- Penner JE, Xu L, Wang M (2011) Satellite methods underestimate indirect climate forcing by aerosols. PNAS 108:13404–13408
- Pruppacher HR, Klett JD (1997) Microphysics of Clouds and Precipitation. Kluwer Academic Publishers, Dordrecht and Boston
- Ramanathan V, Carmichael G (2008) Global and regional climate changes due to black carbon. Nat Geosci 1:221-227
- Ramanathan V, Chung C, Kim D, Bettge T, Buja L, Kiehl JT, Washington WM, Fu Q, Sikka DR, Wild M (2005) Atmospheric

- brown clouds: Impacts on South Asian climate and hydrological cycle. PNAS 102:5326-5333
- Rosenfeld D, Lohmann U, Raga GB, O'Dowd C, Kulmala M, Fuzzi S, Reissell A, Andreae MO (2008) Flood or drought: how do aerosols affect precipitation? Science, 321:1309-1313
- Schulz M, Textor C, Kinne S et al (2006) Radiative forcing by aerosols as derived from the AeroCom present-day and pre-industrial simulations. Atmos Chem Phys 6:5225-5246
- Schwartz SE, Charlson RJ, Kahn RA, Ogren JA, Rodhe H (2010) Why hasn't Earth warmed as much as expected? J Clim 23:2453-2464
- Shindell D, Faluvegi G (2009) Climate response to regional radiative forcing during the twentieth century. Nat Geosci 2:294-300
- Shindell D, Kuylenstierna JCI, Vignati E et al (2012) Simultaneously mitigating near-term climate change and improving human health and food security. Science 335:183-189
- Stevens B, Feingold G (2009) Untangling aerosol effects on clouds and precipitation in a buffered system. Nature 461:607-613
- Stratmann F, Bilde M, Dusek U et al (2010) Examination of laboratorygenerated coated soot particles: An overview of the LACIS Experiment in November (LExNo) campaign. J Geophys Res 115:D11203 doi: 10.1029/2009JD012628
- Swietlicki E, Hansson H-C, Hämeri K et al (2008) Hygroscopic properties of submicrometer atmospheric aerosol particles measured with H-TDMA instruments in various environments - a review. Tellus B 60:432-469

- Textor C, Schulz M, Guibert S et al (2006) Analysis and quantification of the diversities of aerosol life cycles within AeroCom. Atmos Chem Phys 6:1777–1813
- Tunved P, Hansson H-C, Kulmala M, Aalto P, Viisanen Y, Karlsson H, Kristensson, Swietlicki E, Dal Maso M, Ström J, Komppula M (2003) One year boundary layer aerosol size distribution data from five Nordic background stations. Atmos Chem Phys 3:2183-2205
- UNEP (2011) Integrated Assessment of Black Carbon and Tropospheric Ozone: Summary for Decision Makers. www.unep.org/dewa/Portals/67/pdf/Black_Carbon.pdf
- Vehkamäki H, Riipinen I (2012) Thermodynamics and kinetics of atmospheric aerosol particle formation and growth. Chem Soc Rev 41:5160-5173
- Vestreng V, Myhre G, Fagerli H, Reis S, Tarrason L (2007) Twenty-five years of continuous sulphur dioxide emission reduction in Europe. Atmos Chem Phys 7:3663–3681
- Zieger P, Fierz-Schmidhauser R, Gysel M, Ström J, Henne S, Yttri KE, Baltensperger U, Weingartner E (2010) Effects of relative humidity on aerosol light scattering in the Arctic. Atmos Chem Phys 10:3875–3890
- Zubler EM, Folini D, Lohmann U, Lüthi D, Schär C, Wild M, (2011) Simulation of dimming and brightening in Europe from 1958 to 2001 using a regional climate model. J Geophys Res 116:D18205 doi: 10.1029/2010JD015396