



Energy research Centre of the Netherlands

Climate change and the impact of aerosol

A literature review

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Preface

A substantial part of this research study is financed by the Ministry of Housing, Spatial Planning and the Environment (project no. 5.0092). The work in this project has been executed after close consultation with the sponsor.

The information in this report is based on a wide reading of the relevant literature and existing expertise in the topic. Not all topics have an equally strong base, as they differ in the extent to which it has been researched (e.g. aerosol emissions from biofuel use are only scarcely known).

Abstract

Some of the main uncertainties in understanding climate change concern the role of aerosols.

Aerosol particles vary widely in size and chemical composition, and both are important in governing their effects on climate and human health. For climate effects, the number of particles (of intermediate size) is more important than the mass (in contrast to e.g. the ecological effects, which are solely influenced by the mass concentration). Aerosols have both natural and anthropogenic sources. They usually consist of a mixture of components: Sulfates, nitrates, ammonium, organic carbon, black carbon, sea salt, mineral dust, trace metals and water.

Aerosol particles affect the climate directly by scattering and absorbing of solar radiation and indirectly by modifying the properties of clouds. Black carbon causes warming, whereas all other aerosol causes cooling. The short lifetime of aerosols (days/weeks) means that their effects are more regional and less persistent into the future than those of the long lived greenhouse gases. Aerosol concentrations are highly variable in space and time due to their short lifetime. This variability is an important factor in the relatively large uncertainty surrounding aerosol effects on climate.

Climate can change as a consequence of many factors, some of which are natural, and some of which are anthropogenic. The global warming as the globe has experienced over the past ~150 years is due to a combination of factors, the most important one being man-made greenhouse gases, which cause the earth to trap more heat. Aerosol cooling has masked some of this warming. The influence of natural factors, such as the sun and volcanoes, has been minor and episodic, respectively. Over the longer term (decades to centuries), CO₂ will be the main driver of climate change because of its very long presence in the atmosphere (in sharp contrast to aerosols).

The strong increase in man-made aerosols during the middle of the 20th century has counteracted the increase in greenhouse gases during that same period, causing the global temperature to remain relatively steady. After the late 1970's however, greenhouse forcing became dominant, and the globe warmed again.

Aerosol concentrations are expected to be reduced globally, but at a different pace in different regions. Reductions are achieved first in developed countries, whereas in developing countries the aerosol burden may first increase before it starts to decline at a later stage. A consequence of reductions in aerosol concentrations will be a more pronounced warming, as the underlying greenhouse warming trend will be revealed. Trends in aerosol concentrations continue to have an important influence on the climate, especially regionally.

Many combustion processes result in the simultaneous emission of (warming) greenhouse gases and (cooling and warming) aerosols. In devising policies to curb health effects, simultaneous climate impacts should be considered, and vice versa. It is becoming more and more clear that regional air quality and global climate issues are intricately linked with one another.

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1. Introduction

The global climate has significantly warmed since the beginning of the 20th century. Several factors have contributed to this warming. The dominant factor is the emission of greenhouse gases resulting from human activity. The amount of aerosols has also increased, and since they exert a cooling effect on average, part of the greenhouse warming has been compensated. Natural factors, such as an increase in solar activity in the first half of the 20th century, also contributed to the warming over that time period. Large volcanic eruptions cause episodic cooling (from stratospheric aerosol). In isolation however, these natural factors fall far short in explaining the observed increase in global temperature (especially since the solar output has not changed over the past 50 years besides the 11 year solar cycle).

Even though large uncertainties remain, the picture of the global climate system, of how and why it is changing, has gradually become clearer over the past decades. The scientific basis has grown ever stronger (Oreskes, 2007), and on this steadily growing basis –with its inherent uncertainties- policies will have to be devised. Since the major thrust and cause of the expected changes are clear, so is the direction of the needed policies.

However, subtleties remain in many areas, with ramifications for the optimal policy portfolio. Many of these involve the behavior of aerosols, whose role in climate change is credited with the largest uncertainty. This report aims to highlight their role in the climate system, and how it is expected to change in the future. For example, the reduction of aerosol sources (needed because of their adverse health and ecological effects) will lead to a net warming, because the full greenhouse warming will then be unmasked. The current knowledge about aerosols will be put in the context of climate change as a whole, to maximize the policy relevance of this document.

The main thrust of current climate change is clear: The globe is warming, predominantly due to emissions from human activity. The main uncertainties concern the role of aerosols.

2. Aerosols and their role in the climate system

2.1 Background

Origin

Aerosols are liquid or solid particles suspended in the atmosphere. They can either be directly emitted into the atmosphere (primary aerosol like dust, sea salt and black carbon), or they can be formed in the atmosphere by condensation (secondary inorganic aerosol such as sulfates, nitrates and ammonium). Organic aerosol can be both of primary or secondary origin. Organic aerosol and black carbon are sometimes called carbonaceous (carbon-based) aerosol. Condensation of vapour can lead to the growth of existing particles, or to the formation of new particles (called nucleation).

Size

Almost all of the aerosol properties, and thus effects, are size dependent: The particle size governs the rate at which they deposit to the ground (and thus atmospheric lifetime), their interaction with radiation, their impact on clouds, and their health effects. Their size can range from a few nanometers (1 nm = one millionth of a millimeter) to tens of micrometers (1 μm = one thousandth of a millimeter). Cloud droplets and raindrops can grow even larger, but are usually classified separately. Though not all particles are spherical, they are usually characterized according to their equivalent spherical diameter.

Number and mass

The tropospheric number concentration of aerosol particles ranges from several tens or hundreds per cubic centimeter of air in remote locations, to more than hundred thousand or a million per cubic centimeter in polluted environments or after a strong nucleation event. The number or volume concentration as a function of particle size is described by a size distribution. An important feature of atmospheric aerosol size distributions is their multimodal character, an example of which is given in Figure 2.1. The number of particles is generally dominated by the smaller aerosols, whereas the volume (and thus mass) is dominated by the larger ones. The exact shape of the size distribution depends on the environment, as it reflects the sources, sinks, and transformations of the particles.

Besides the total number and mass of particles, the effects of an aerosol population also depend on the shape of the size distribution (an example of which is shown in Figure 2.1), and is often dominated by a certain size range (dependent on the effect of interest).

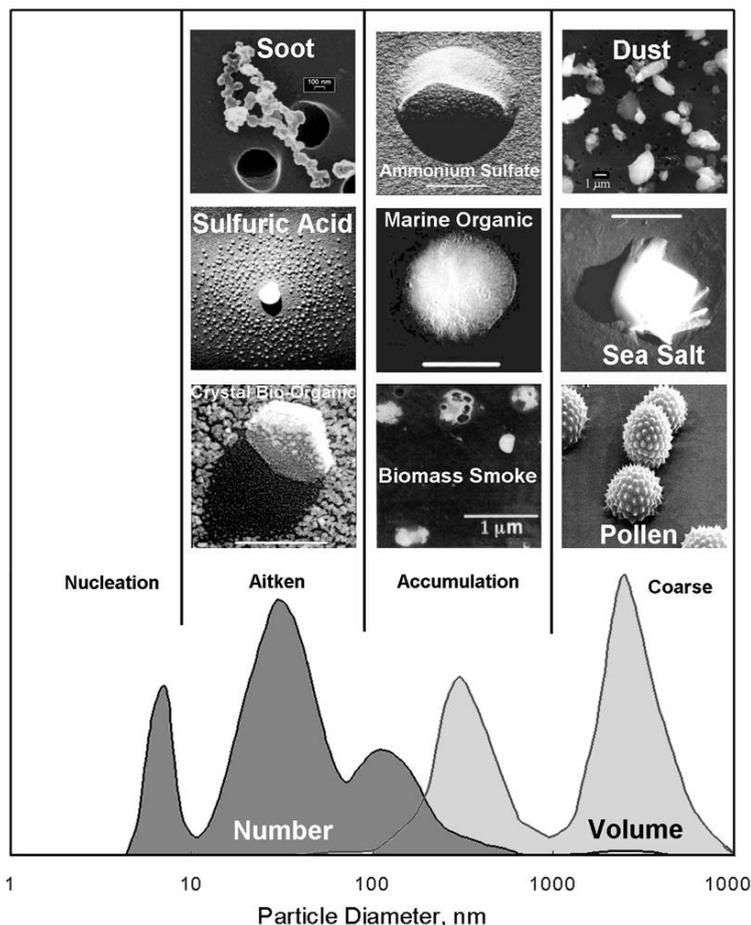


Figure 2.1 *Typical shape of the aerosol number and volume size distribution, showing the different size classes ('modes') as well as example pictures of some archetypical aerosol particles. 1 nanometer (nm) = 0.001 micrometer; upper right value should read 10000 nm = 10 micrometer. (From http://www.esf.edu/chemistry/dibble/presentations/IX_Aerosol.ppt)*

Formation

New particle formation occurs when different vapor molecules collide together to form a new particle. This process is called nucleation. The most abundant trace gases in the atmosphere do not generally nucleate new aerosols (or even condense on existing ones), because they are too volatile. They first have to be oxidized (usually under the influence of sunlight) to produce a compound with a lower vapor pressure. The prime example of this is the oxidation of SO_2 into sulfuric acid, H_2SO_4 , which has a very low vapor pressure. The H_2SO_4 vapor can then condense together with water molecules to form a stable cluster of molecules: a new particle is born of typically 1-2 nanometers (0.001 to 0.002 micrometer) in diameter. Besides sulfuric acid, organic compounds and ammonia are considered to be potentially important in the nucleation process (Metzger et al., 2010). Ions (charged molecules) can also play a role.

A freshly nucleated particle will quickly grow larger by condensation of organic and inorganic vapors. Aerosol particles can also collide together, in a process called coagulation. After a nucleation event, during which hundreds of thousands of new particles can be produced, coagulation is particularly efficient. Only a small fraction of the small particles will reach large enough sizes to influence climate. The majority of small particles (with diameters less than 0.1 micrometer) are created in the atmosphere by nucleation. Favorable conditions for nucleation to occur include a strong source of condensable vapor; high solar radiation intensity; low pre-

existing aerosol concentration; high relative humidity; low temperature; presence of ions; and atmospheric mixing processes (Verheggen, 2009).

Transformation

In the atmosphere, particles are subject to transformation processes (e.g. oxidation, polymerization, evaporation, condensation, coagulation) and they will not persist in their original state (as shown in Figure 2.1) for very long. For example, a black carbon or soot particle will start to be oxidized at the edges, after which more water retaining species condense on its surface, and it will become a liquid mixed particle, with traces of black carbon, inorganic compounds and organic species. Dependent on their size, aerosol particles only reside in the atmosphere for several days to weeks before being deposited to the ground. Particles of intermediate size (of a few hundred nanometers, aptly named the accumulation mode) have the longest atmospheric lifetime.

Table 2.1 *Particle emission/production (for primary and secondary aerosol, respectively, in teragrams per year; 1 Tg = 1 million tonnes) and burden for both mass and number of aerosols, estimated for the year 2000. Note that sea salt and dust are mainly present in the coarse mode (large sizes). For climate effects the aerosol number is more important than the mass. (From Andreae and Rosenfeld, 2008)*

	Mass emission			Mass Burden	Number Prod.	Number Burden
	"Best guess"	Min	Max			
	Tg a ⁻¹			Tg	a ⁻¹	
Carbonaceous aerosols						
Primary organic (0–2 µm)	95	40	150	1.2	–	310 · 10 ²⁴
Biomass burning	54	26	70	–	7 · 10 ²⁷	–
Fossil fuel	4	3	9	–	–	–
Biogenic	35	15	70	0.2	–	–
Black carbon (0–2 µm)	10	8	14	0.1	–	270 · 10 ²⁴
Open burning and biofuel	6	5	7	–	–	–
Fossil fuel	4.5	3	6	–	–	–
Secondary organic	28	2.5	83	0.8	–	–
Biogenic	25	2.5	79	0.7	–	–
Anthropogenic	3.5	0.05	4.0	0.08	–	–
Sulfates	200	107	374	2.8	2 · 10 ²⁸	–
Biogenic	57	28	118	1.2	–	–
Volcanic	21	9	48	0.2	–	–
Anthropogenic	122	69	214	1.4	–	–
Nitrates	18	12	27	0.49	–	–
Industrial dust, etc.	100	40	130	1.1	–	–
Sea salt						
d < 1 µm	180	60	500	3.5	7.4 · 10 ²⁶	–
d = 1–16 µm	9940	3000	20,000	12	4.6 · 10 ²⁶	–
Total	10,130	3000	20,000	15	1.2 · 10 ²⁷	27 · 10 ²⁴
Mineral (soil) dust						
< 1 µm	165	–	–	4.7	4.1 · 10 ²⁵	–
1–2.5 µm	496	–	–	12.5	9.6 · 10 ²⁵	–
2.5–10 µm	992	–	–	6	–	–
Total	1600	1000	2150	18 ± 5	1.4 · 10 ²⁶	11 · 10 ²⁴

Range reflects estimates reported in the literature. The actual range of uncertainty may encompass values both larger and smaller than those reported here. Values are based on the following publications: (Andreae, 1995; Andreae and Merlet, 2001; Guelle et al., 2001; Penner et al., 2001; Gong et al., 2002; Luo et al., 2003; Bond et al., 2004; Liousse et al., 2004; Zender et al., 2004; Stier et al., 2005; Ito and Penner, 2005; Junker and Liousse, 2006; Bauer et al., 2007; Henze et al., 2007).

Sources

Atmospheric particles have both biogenic (natural) and anthropogenic (man-made) sources. Examples of natural (primary) particle emissions are the shattering of sea-spray into tiny droplets, particles caused by natural forest fires, soil dust, pollen, etc. Examples of natural emissions of precursors to (secondary) aerosol are SO₂ and H₂S from volcanoes, dimethylsulfide (DMS) from phytoplankton, volatile organic compounds such as monoterpenes from vegetation, etc.

Examples of anthropogenic (primary) particle sources are vehicular emissions, industrial emissions, and some natural sources that are significantly enhanced by human activities, such as biomass burning and soil dust (due to e.g. erosion). Anthropogenic precursors to (secondary) aerosol are emitted by industry, burning of fossil fuels, usage of solvents, agriculture, etc. Table 2.1 gives the estimated particle emissions and burdens, distinguished by source.

Aerosol particles vary widely in size and chemical composition, and both are important in governing their effects on climate and human health. They have both natural and anthropogenic sources. Atmospheric aerosol usually consists of a mixture of components, e.g. sulfates, nitrates, ammonium, organic carbon, black carbon, sea salt, mineral dust, trace metals and water.

2.2 Climate effects of aerosols

Factors that can impact on our climate are often expressed as ‘radiative forcing’. This refers to a change in the radiation budget of the planet, to which it responds by either cooling or warming. Aerosol particles can influence climate in many different ways, as schematically represented in Figure 2.2.

Direct effects

Direct effects refer to the climate effects of the aerosol particles themselves (Yu et al., 2006): They can scatter and absorb (in the case of black carbon) solar radiation. Since a portion of the scattered radiation will be reflected back into space, this leads to a cooling of the atmosphere. Intermediate sized particles (from 0.1 to 2 micrometer diameter) are the most effective at backscattering of solar radiation. The number of these intermediate sized particles is the primary factor governing the strength of the direct effect.

Black carbon absorbs solar radiation, which leads to a warming of the atmosphere. If black carbon rich particles are incorporated into cloud droplets, the lens effect of the water increases the absorption, and the cloud droplet may evaporate due to the local heating. This is called the semi-direct effect. Black carbon deposition on snow and ice could decrease the surface albedo, leading to additional warming (not included in Figure 2.2). Aerosol compounds other than black carbon predominantly cause cooling, since they do not absorb solar radiation. Mineral dust can absorb solar radiation to a small extent.

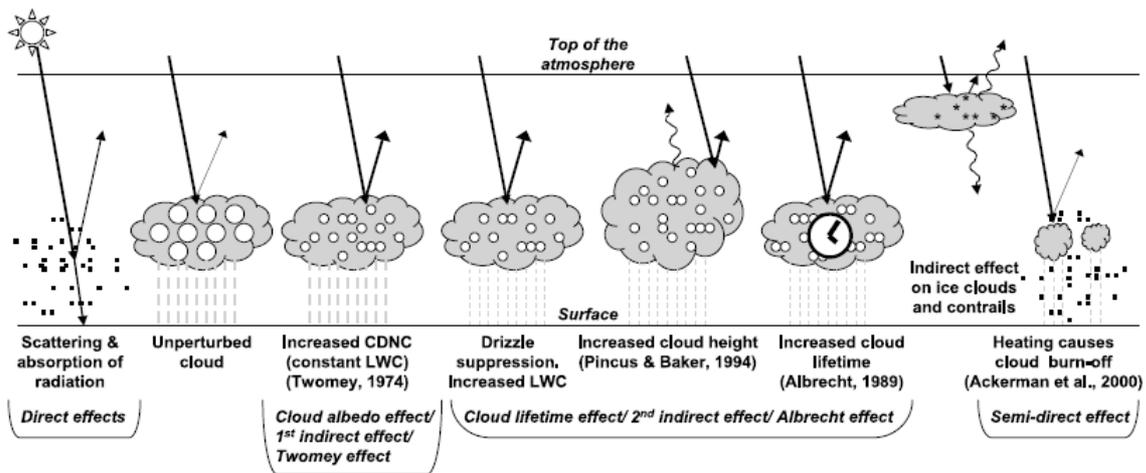


Figure 2.2 Different ways in which aerosol particles affect climate. The small black dots represent aerosol particles; the larger open circles cloud droplets, and the asterisks ice crystals. Perturbed clouds have more (and thus smaller) droplets because of the availability of more aerosol particles on which to condense. CDNC = cloud droplet number concentration; LWC = liquid water content. (From IPCC, 2007)

Indirect effects

Indirect effects refer to the climate effects of aerosols via their role in cloud formation (Lohmann and Feichter, 2005): Aerosol particles act as cloud condensation nuclei (CCN) around which cloud droplets can form. The larger the aerosol particle, the more effective it is a CCN. Under most conditions relevant for Western Europe, particles have to surpass a diameter of around 0.1 micrometer to be engaged in cloud formation.

If more aerosol particles (CCN) are available, more cloud droplets will be formed. Since the available water has to be divided over more droplets, their average size will be smaller. However, the total surface area of all the droplets together is larger, and the resulting cloud is more reflective (in other words, it has a higher albedo; this is the *first indirect effect*, or cloud albedo effect). Such a ‘perturbed’ cloud, existing of more but smaller droplets, will not rain out as quickly. As a result, the cloud persists longer (this is the *second indirect effect*, or cloud lifetime effect). The particle chemical composition also affects the CCN activity, though to a lesser extent than the particle size (Dusek et al., 2006). The primary parameter influencing the strength of the indirect effect is the number concentration of particles larger than ~0.1 micrometer (as long as they are not hydrophobic / water repellent).



Figure 2.3 The indirect effect is demonstrated by aerosol emissions from shipping. The elevated aerosol concentration in the wake of a ship causes the stratus clouds to be more reflective (i.e. whiter).

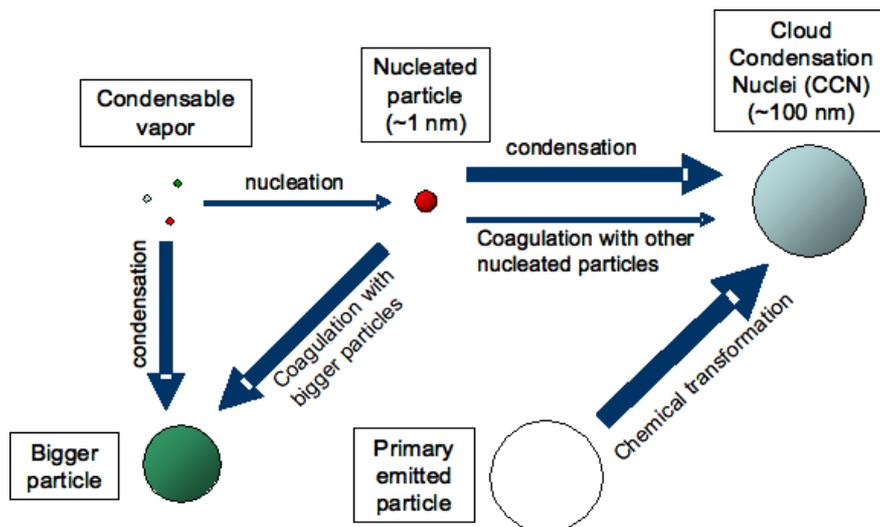


Figure 2.4 *Different factors influence the extent to which nucleation contributes to the number of cloud condensation nuclei (CCN). (From Verheggen, 2009)*

A small subset of particles can also act as ice nuclei (IN), on which ice crystals can form. The formation of an ice crystal usually occurs via freezing of a liquid droplet, and is called ice nucleation. Mineral dust is typically regarded as a good ice nucleus. There are also indications that black carbon can participate in ice nucleation, though the evidence is not yet conclusive (Cozic et al., 2007). If indeed it does, then the increased burden of black carbon due to human activities could have caused an increase in the number of ice crystals, which are effective at initiating precipitation. This would result in a reduced cloud lifetime and cloud cover, causing a warming of the climate. However, this is still a speculative hypothesis and has not been either confirmed or quantified.

The exact effect of having more CCN in the atmosphere depends on a lot of factors: Seen from below, clouds absorb and re-emit infrared radiation back to the surface (much as greenhouse gases do), and so exert a warming effect. Seen from above, clouds reflect sunlight and emit infrared radiation to space, and so exert a cooling effect. The global net effect is a cooling, but dependent on conditions, this can vary with location and time: Low clouds generally cool, though high clouds generally warm the surface. This is due to high cirrus clouds usually being optically thin, so reflecting only a small portion of the incoming solar radiation. In addition, due to their low temperature, they re-emit radiation of low energy content, so less energy is lost to space and more remains in the atmosphere. At night time clouds cause warming, since there is no solar radiation to reflect, but there is infrared radiation from the surface to absorb and re-emit. During the day cooling generally dominates (dependent on altitude).

Uncertainty

The impacts of aerosols on climate are significant, but also very uncertain. There are several reasons why the uncertainty in aerosol climate effects is so large compared to that of long-lived greenhouse gases:

- The lifetime of the major greenhouse gases (with the exception of tropospheric ozone) is counted in years (e.g. methane) or even centuries (e.g. carbon dioxide), whereas aerosol particles only reside in the atmosphere for several days to at most a few weeks, before they are deposited onto the ground.
- As a consequence, the aerosol concentration is highly variable in space and time, and estimates of (changes in) its global concentration field are very uncertain.
- The atmospheric aerosol is a conglomerate of particles of widely varying size and chemical composition. The climate (as well as the health) effects depend on particle size and composition.

- The formation of aerosols in the atmosphere (by nucleation processes) is poorly understood.
- The influence of aerosols on clouds is uncertain, because cloud dynamics and microphysics have many different dependencies and are poorly understood.

This means that even though our knowledge about the role of aerosols in the climate system is increasing with small increments, it will likely continue to dominate the uncertainty of expected climate changes into the near future (few decades). However, the climate change to be expected in the farther future (multiple decades to centuries) is much more dependent on greenhouse gases than on aerosols, since the plateau on the former is deemed higher than on the latter.

Aerosol particles affect the climate directly by scattering and absorbing of solar radiation and indirectly by modifying the properties of clouds. Black carbon causes warming, whereas all other aerosol causes cooling. The short lifetime of aerosols (days/weeks) means that their effects are more regional and less persistent into the future than those of the long lived greenhouse gases. Large uncertainties regarding their precise effects will remain a steady feature of climate science for some time to come.

3. Climate change

3.1 Background

Greenhouse warming versus aerosol cooling

Even though greenhouse gases and aerosols have an opposite effect on global average temperature, they do not perfectly cancel each other.

First off, the spatial and time scales of their effects are very different: Because of their short lifetime (1-2 weeks), the effects of aerosols are much more regionally concentrated than those of greenhouse gases, and they don't persist nearly as long. In contrast, even though the majority of emitted CO₂ will be drawn down into the oceans after a few centuries, a significant fraction (20%) will stay in the atmosphere for millennia (Archer and Brovkin, 2008).

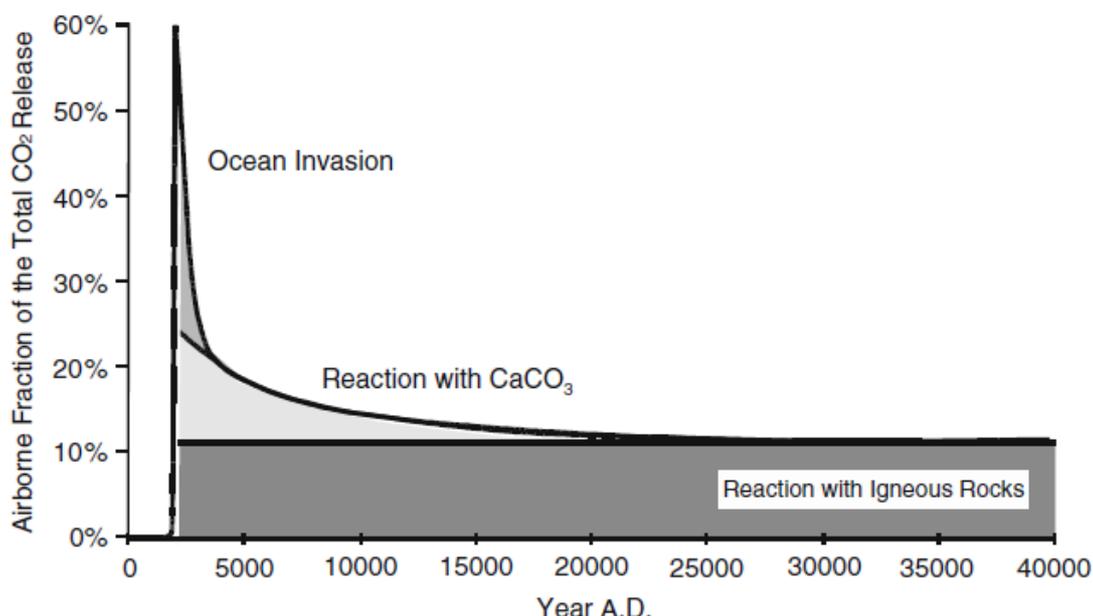


Figure 3.1 *The fraction of CO₂ remaining in the atmosphere after emission. Two thirds of the initial peak is absorbed by the oceans within a century. However, the remainder takes much longer to be removed, as different –much slower– processes are responsible for its further removal. (From Archer and Brovkin, 2008)*

Secondly, greenhouse forcing is based on intercepting part of the infrared radiation that the earth emits. Since a part is reflected back, this leads to a warming of both the atmosphere and the surface. Aerosols on the other hand reflect and absorb solar ultraviolet radiation. This leads to a cooling of the surface (because of less incident UV radiation), but to a warming of the atmosphere and a change in the atmospheric thermal structure (Ramanathan and Feng, 2009). Both absorbing and reflecting aerosols reduce the amount of surface solar radiation (“global dimming”), which in turn makes the planet drier via the suppression of evapotranspiration. Greenhouse forcing on the other hand makes the planet wetter (due to the positive water vapour feedback on temperature). These effects don't occur in the same places, so they do not necessarily cancel each other on a regional scale.

Climate effects of different compounds

Figure 3.2 gives the emission specific radiative forcing of climate from pre-industrial until the present (2005).

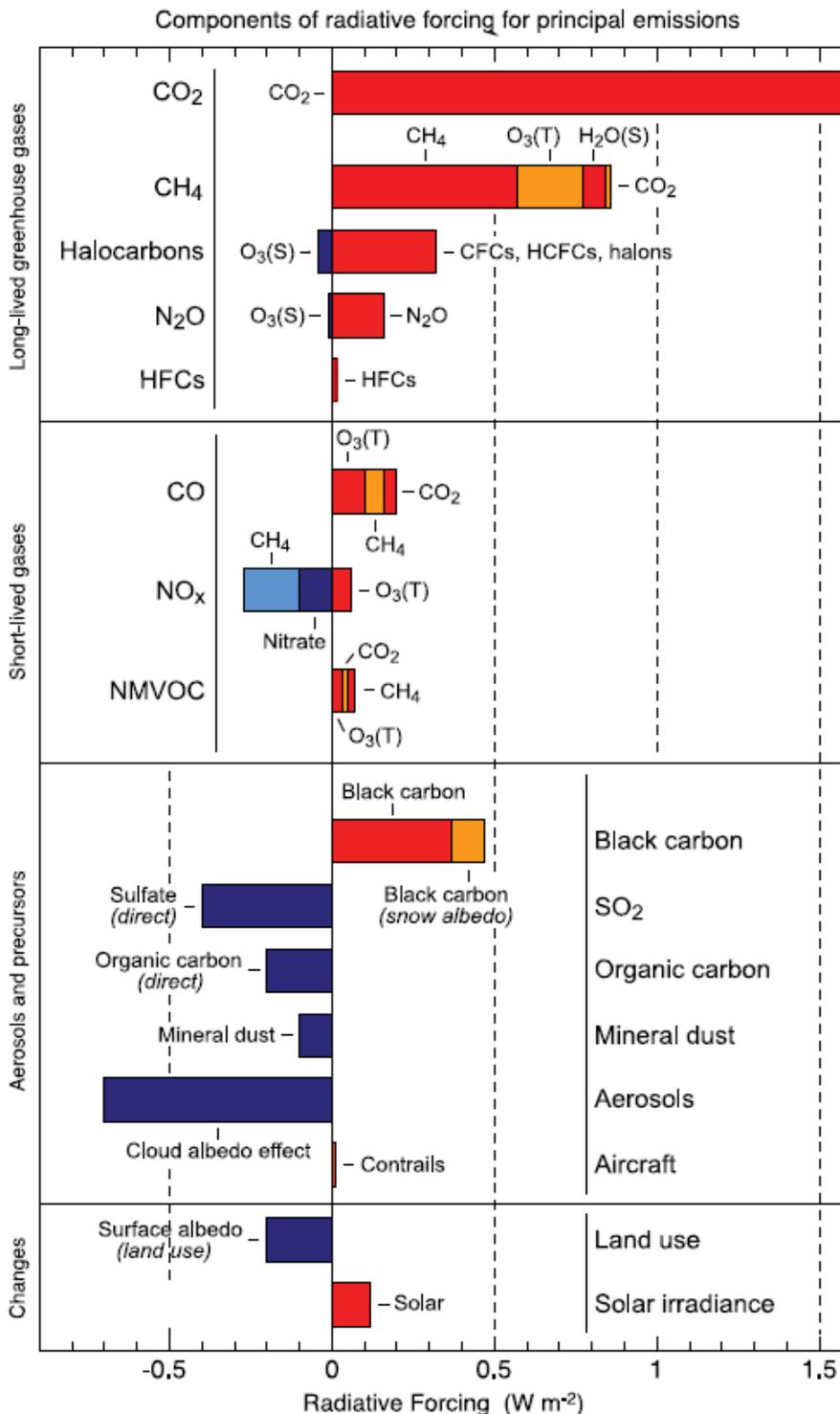


Figure 3.2 Radiative forcing of climate in 2005 due to emissions and changes since 1750. Indirect effects attributable to the emission of a primary compound are classed with the latter (e.g. more CH₄ leads to more O₃). Note that nitrate aerosol is thus classed as being dependent on NO_x emissions. (From IPCC, 2007)

Climate sensitivity

The amount of warming that the globe has experienced so far depends on three main factors:

- The net radiative forcing of climate (both natural and anthropogenic)
- The equilibrium climate sensitivity (the increase in global mean temperature at equilibrium, in response to a radiative forcing equaling a doubling of the CO₂ concentration)
- The climate response time (mainly due to ocean inertia); this governs how far removed the climate is from equilibrium.

Due to the large uncertainty in the climate effects of aerosols, the net radiative forcing since pre-industrial times is not accurately known: Figure 3.3 shows that its value is most likely within the range 0.5 – 2.5 W/m². This does not directly translate into uncertainty in climate sensitivity (the equilibrium warming resulting from a specified amount of radiative forcing), as has been postulated by Andreae et al. (2005). The climate is currently not in equilibrium, and we do not know exactly how much warming is still ‘in the pipeline’. This climate response time is linked to e.g. the rate at which the accumulated heat mixes down to the deep ocean. Hansen (2008) has postulated that the equilibrium climate sensitivity is much better constrained by paleoclimate data and the response to volcanic eruptions (at 3 degrees per doubling of CO₂, see Figure 3.4), and that the uncertainty in current radiative forcing is rather the mirror image of uncertainty in the response time of the climate system.

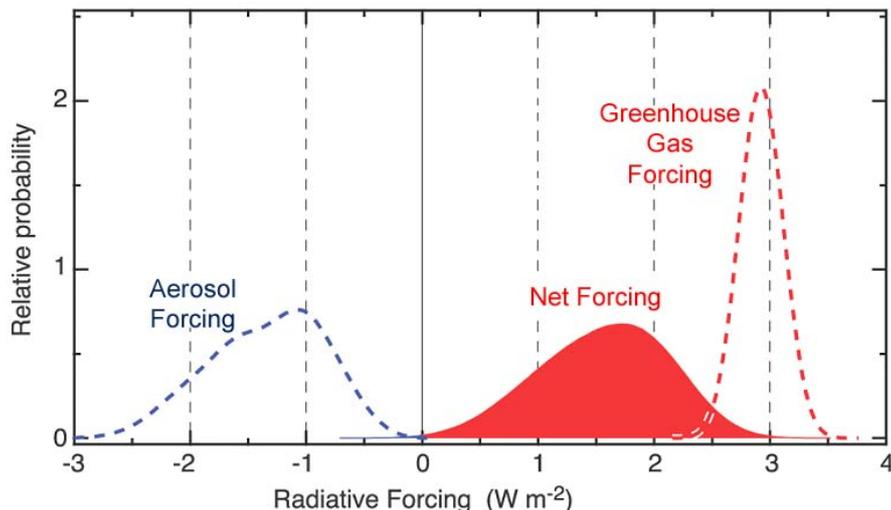


Figure 3.3 *The greenhouse gas forcing (dashed red curve) is relatively well known, but the aerosol forcing (dashed blue curve) is not. The resulting net anthropogenic forcing (red solid curve) is not well constrained. The height of the curve gives the relative probability of the associated value, i.e. the net climate forcing is likely between 1 and 2 W/m², but could be anywhere between 0 and 3 W/m². (From IPCC, 2007)*

Roe and Baker (2007) have argued that the large and skewed uncertainty¹ in climate sensitivity is an inevitable consequence of the nature of the climate system. Others have pointed out that this uncertainty is not inherent to the climate system, and that Roe and Baker’s result is “caused by a peculiar definition of uncertainty used by these authors” (Hannart et al., 2009). Annan and Hargreaves (2006) arrive at a much narrower uncertainty bound than would be expected if Roe and Baker’s analysis were correct (see also Figure 3.4).

¹ “Skewed” uncertainty refers to the fact that the upper bound is less well constrained than the lower bound: The probability density function of climate sensitivity has a “fat tail” towards high values.

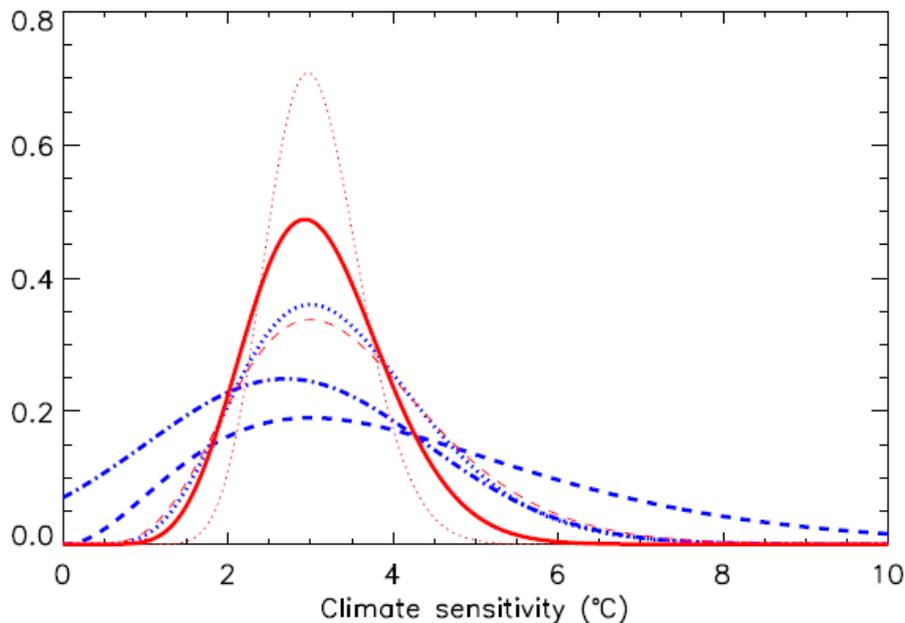


Figure 3.4 *Likelihood functions for climate sensitivity (defined as the equilibrium warming following a doubling of CO₂ concentrations) based on various observational constraints. Blue dashed line: 20th century warming. Blue dotted line: volcanic cooling. Blue dot-dashed line: Last Glacial Maximum (ice age) cooling. Red solid line: combination of the three constraints. Thin red dashed line: combination of three copies of widest constraints. Thin red dotted line: five constraints. (From Annan and Hargreaves, 2006)*

Observed warming since pre-industrial times

Global average surface temperatures have increased by about 0.75 degrees Celsius since the beginning of the industrial revolution, of which ~0.6 °C is attributable to human activities. The total radiative forcing by greenhouse gases is around 3 W/m², with which we have ‘committed’ the planet to warm up by 2.4 °C (1.6-3.6 °C), according to a climate sensitivity of 3 °C (2-4.5 °C) for a doubling of CO₂. The observed amount of warming thus far has been less than this, because part of the excess energy is stored in the oceans (amounting to ~0.5 °C), and the remainder (~1.3 °C) has been masked by the cooling effect of anthropogenic aerosols (Ramanathan and Feng, 2009). This simple analysis shows that the ‘2 degree target’ of maximum acceptable warming is looming on the horizon, as the climate equilibrates and aerosol pollution is cleaned up. It highlights the need for strong and immediate reduction of greenhouse gas emissions, if we are to prevent global warming to exceed 2 degrees above pre-industrial levels.

Modeled warming since pre-industrial times

Global climate models can reproduce the global pattern of the observed temperature change rather well, but only if both natural and anthropogenic climate forcings are taken into account. The good match between observed and simulated warming instills some confidence that the models include the most important physical processes of roughly the correct magnitude. See Figure 3.5 for an example of how the global average temperature anomaly as observed over the past 130 years is matched by a global climate model (GCM).

The same climate forcings as mentioned in Figure 3.2 are provided here (though grouped differently), and their change in time is shown. As mentioned above, the greenhouse gases (warming) and aerosols (cooling) are the dominant drivers of climate change on this timescale. The relatively strong aerosol cooling between the 1950’s and 1970’s is thought to be responsible for the relatively stable temperatures during the middle of the 20th century.

The increase in solar output had a clear, though minor influence on the increasing temperatures during the first half of the 20th century. Since the 1950's however, solar output has not changed significantly (besides the 11 year solar cycle), and consequently, it can not be the cause of the large upswing in global temperature that occurred since 1975. Large volcanic eruptions, of which the plume extends all the way into the stratosphere (higher than ~10 km altitude), exert a cooling effect for a few years. This is due to the enhanced amount of sulfate aerosol they put in the stratosphere, where its lifetime is much longer than in the troposphere (1-2 years).

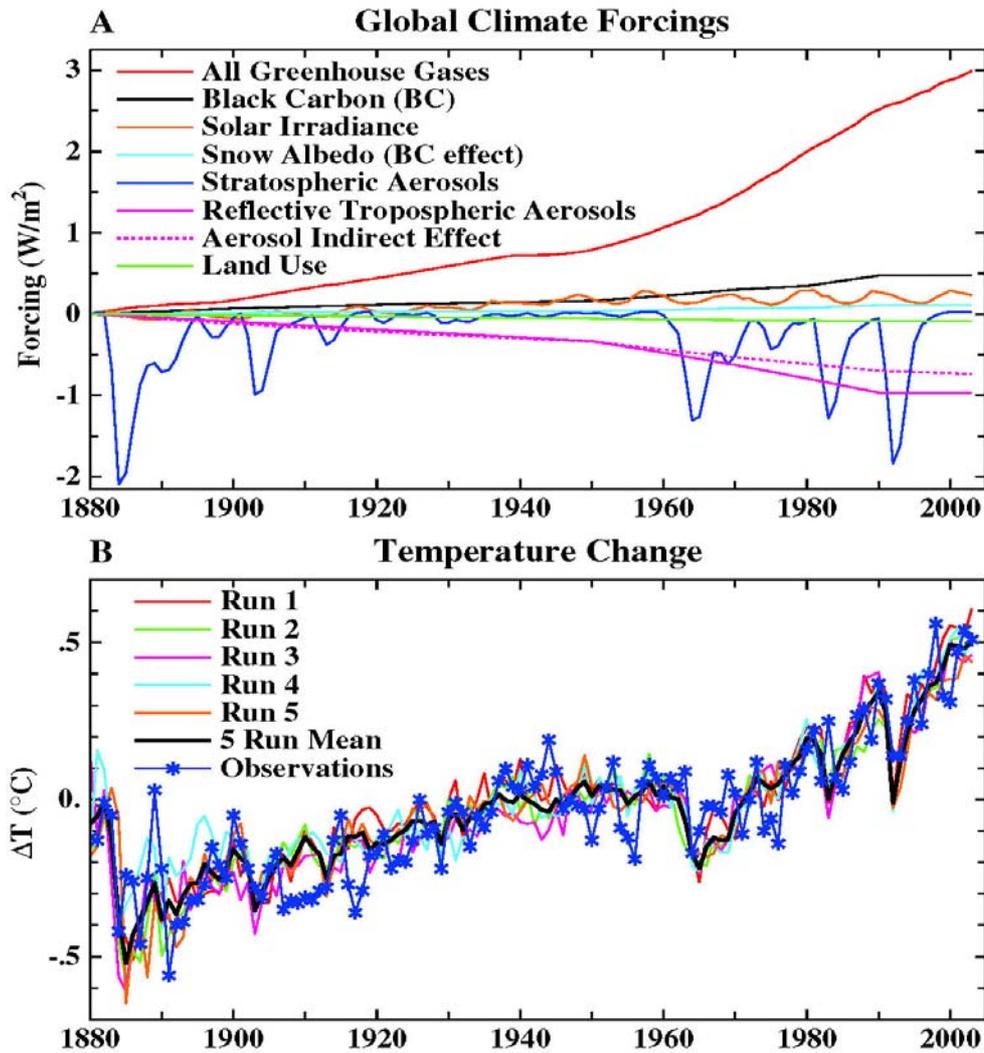


Figure 3.5 A: The different types of radiative forcing of climate since pre-industrial times (stratospheric aerosols originate from major volcanic eruptions). B: Simulated (lines) and observed (blue asterisks) surface temperature change. (From Hansen et al, 2005.)

Climate can change as a consequence of many factors, some of which are natural, and some of which are anthropogenic. The global warming as the globe has experienced over the past ~150 years is due a combination of factors, the most important one being man-made greenhouse gases, which cause the earth to trap more heat. Aerosol cooling has masked some of this warming. The influence of natural factors, such as the sun and volcanoes, has been minor and episodic, respectively. Over the longer term (decades to centuries), CO₂ will be the main driver of climate change because of its very long presence in the atmosphere (in sharp contrast to aerosols). Each doubling of CO₂ concentrations is expected to cause a global warming of ~3°C (2 to 4.5) after equilibration of the climate system (barring any surprises such as the passing of ‘tipping points’). This is referred to as the “climate sensitivity”.

3.2 Trends in aerosol climate forcing

Due to the high spatial variability of aerosols and the incomplete spatial coverage of in-situ monitoring, satellite retrievals play an important role in assessing the global distribution of aerosols and any trends therein (Isaksen et al., 2009). Indirect measures can also be used to elucidate trends in aerosol concentrations.

Aerosol trends: From global dimming to global brightening

In several locations, the amount of solar radiation reaching the earth surface decreased between ~1950 and ~1980. This was not due to changes in the output of the sun, which has remained relatively constant from the 1950’s onwards (apart from the 11 year solar cycle). This phenomenon was named ‘global dimming’, and it has been attributed to increases in the amount of aerosols, which cause more sunlight to be reflected or absorbed before it can reach the earth surface. From the 1980’s onwards, the trend in some regions (predominantly Europe) started to reverse: The amount of surface solar radiation started to increase again. This was called ‘global brightening’, and was attributed to emission reductions of aerosols and their precursors, notably sulfur dioxide. This change in trend is also expected to occur in other parts of the world, as air pollution control starts to be implemented more widely. A thorough review of global dimming and brightening is given by Wild (2009). Trends in aerosols and aerosol related quantities are most likely caused by anthropogenic rather than natural sources. Natural production of aerosols has not likely experienced strong trends. The effect of episodic volcanic aerosols is evident in many records nevertheless. A selection of findings relating to the period of ‘global brightening’ will be discussed below.

The amount of haze, mist and fog over Europe has decreased over the past 30 years. This was deduced from visibility data, which correlate with sulfur dioxide emissions (see Figure 3.6). This correlation with an important aerosol precursor suggests that air quality improvements, and subsequent decreases in aerosol concentrations, contributed to the decline of low-visibility conditions (Vautard et al., 2009).

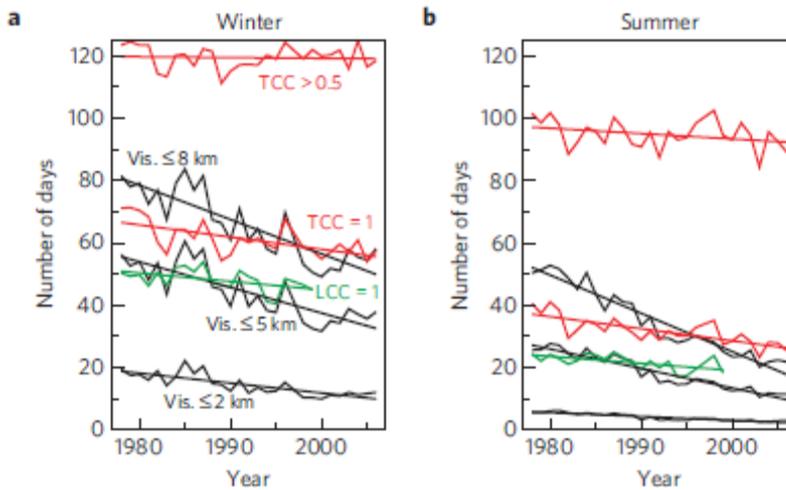


Figure 3.6 *Time evolution of the number of low-visibility days. A: Time evolution of the mean number of low-visibility and overcast (total cloud cover) weather days in the autumn–winter season (Jan.–March and Oct.–Dec. of each year). B: The same as A for the spring–summer period (April–Oct.). ‘Vis’ means visibility, ‘TCC’ means ‘total cloud cover’ and ‘LCC’ means ‘low cloud cover’. (From Vautard et al, 2009)*

Extended records of direct measurements of atmospheric aerosol concentrations are scarce. In the Canadian Arctic, atmospheric concentrations of sulfate and black carbon aerosols have been monitored since 1989. An evaluation of these records showed a substantial decrease of 29% in sulfate aerosols and 56% in black carbon aerosols between 1989 and 2002 (Sharma et al., 2004), again in line with ‘global brightening’ over the same period. However, Sharma et al. (2006) report an increase during winter (January to April) beginning in 2000. Quinn et al. (2007) report a decreasing trend for both sulfate and black carbon at several Arctic stations in the 1990’s, whereas after 2000 some station showed a slight reversal in trend.

Sulfur trapped in ice cores in Greenland is indicative of the presence of sulfate aerosols downwind of the United States and Canada (McConnell et al., 2007). These data show an increase of sulfur in Greenland ice cores from the 1940’s to the 1980’s and a decrease thereafter, in line with northern American emission inventories.

Satellite based measurements of the Aerosol Optical Depth (AOD, a measure of the amount of light scattered by aerosols) confirm that over Europe, aerosols have been in decline since the 1980’s. Over South Asia aerosols are still increasing in concentration, over North America there is no appreciable trend over the past 20 years. Globally, as well as over North America, the AOD appears to be slightly increasing again after having reached a plateau in the 1980’s - 1990’s (Wang et al., 2009). This seems to be consistent with the observations in the Arctic described above. The satellite measurements are shown in Figure 3.7.

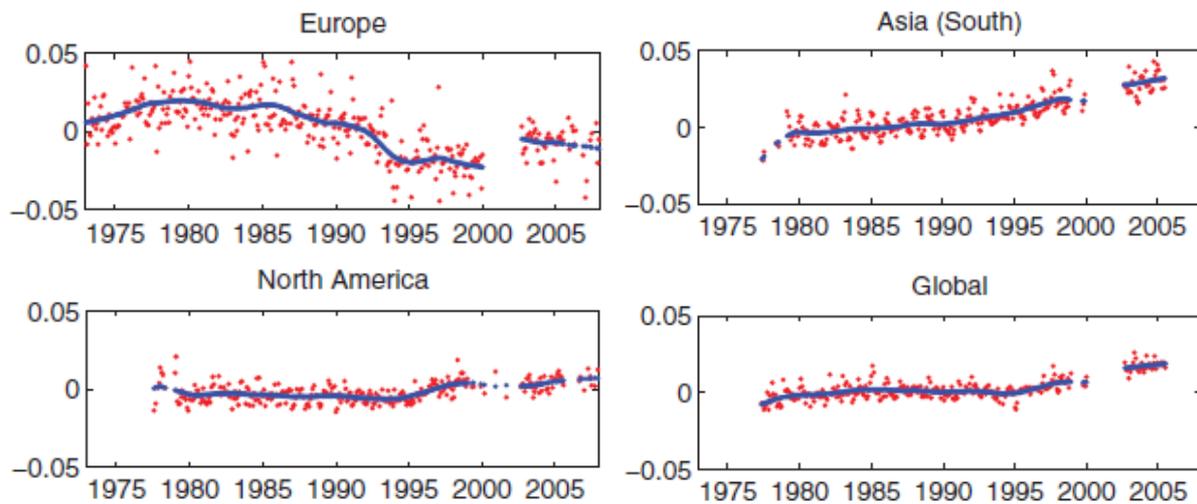


Figure 3.7 *Aerosol Optical depth (AOD) anomaly over land as measured from satellite. AOD is a measure of the amount of light attenuated by the presence of aerosols. Red dots are monthly values; blue lines give the smoothed 5 year average. (from Wang et al., 2009)*

Rising aerosol concentrations from the 1950's to the 1980's, and subsequent decreases due to emission reductions in Europe likely contributed to the observed temperature evolution over mainland Europe since 1950 (Figure 3.8). Slightly decreasing temperatures since the 1950's (green) are in line with rising aerosol concentrations and a subsequent solar dimming, which masked anthropogenic greenhouse warming. The rapid temperature rise since the 1980's (blue) is partly due to a decline in aerosol concentrations over Europe, which led to so-called solar brightening. The decline in aerosol cooling caused the previously masked greenhouse warming to be revealed in a relatively short period of time. This probably contributed to mainland Europe having warmed up about twice as fast as the global average (0.38 and 0.18 degree per decade, respectively). The exact attribution of the rate of warming is complicated by the large amount of variability in regional temperature trends, as well as the fact that land surfaces have warmed more than the oceans (0.18 degree per decade is the global average, including oceans).

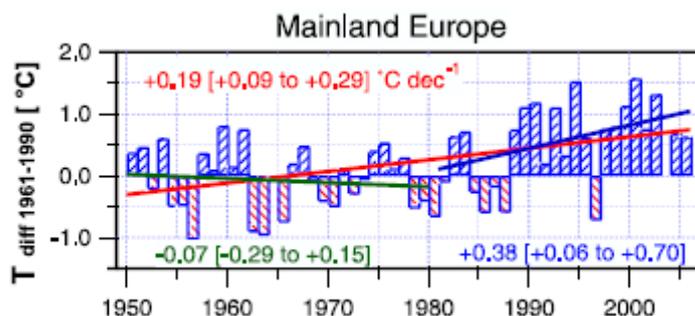


Figure 3.8 *Temperature anomaly with respect to the 1961-1990 mean over mainland Europe since 1950. Linear regression lines and decadal trends show the temperature decline from 1950 to 1980 (green), and the temperature rise to be twice as large for the period 1981 to 2005 (blue) than for the whole period 1950 to 2005 (red). Data from CRU/UEA. (from Philipona et al., 2009)*

Philipona et al. (2009) have attempted to explain the fast European warming by using measurements of the solar radiation received at the Earth's surface in several Swiss and German locations (see Figure 3.9).

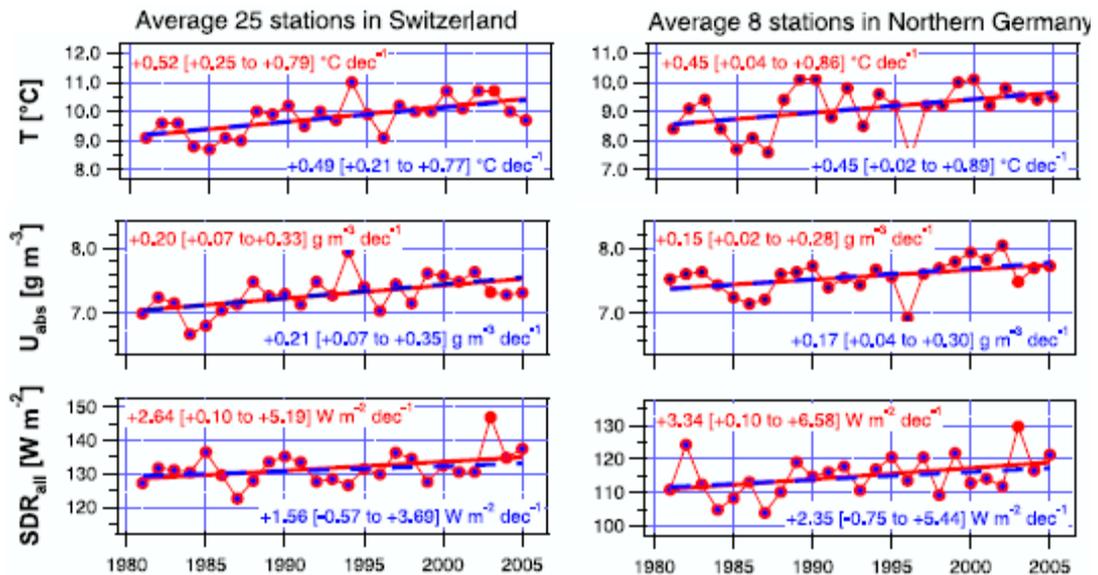


Figure 3.9 Temperature, humidity and radiation increase since 1981. Annual mean values of temperature (T), absolute humidity (U_{abs}) and shortwave downward radiation (SDR) averaged over (left) 25 stations in Switzerland and (right) eight stations in Northern Germany. Linear regression lines and decadal trends with 95% confidence interval in brackets (red) are shown for the period 1981 to 2005. Linear regressions and trends (blue) are also given for the same period but without the year 2003. The rapid temperature and humidity increase seems to be influenced by a gradual solar radiation increase ('brightening' in response to an aerosol decline) but not specifically by the extreme summer 2003. (From Philipona et al., 2009)

The strong increase in man-made aerosols during the middle of the 20th century has counteracted the increase in greenhouse gases during that same period, causing the global temperature to remain relatively steady. After the late 1970's however, greenhouse forcing became dominant, and the globe warmed again. The period of increasing aerosol concentrations is often referred to as 'global dimming' (since it causes a decrease in sunlight reaching the earth's surface). Some regions of the globe are now experiencing 'global brightening', caused by a reduction in the amount of aerosols. This is notably the case in Europe.

3.3 Expected future trends

The global burden of different aerosol types, as simulated by global transport models for the year 2000, is shown in Figure 3.10. Notably absent in this figure is nitrate. Sulfate has received much more attention than nitrate as a dominant aerosol component, though awareness of its importance is slowly increasing.

Ammonium nitrate aerosol forms if sulfate aerosol is fully neutralized and there is excess ammonia. The amount of nitrate aerosol (and its climate impact) therefore depends on both ammonia as well as NO_x emissions. Because of high emissions of ammonia, nitrate aerosol is relatively more prevalent in the Netherlands compared with elsewhere.

Because of the indirect dependence on sulfate neutralization, a decrease in sulfate aerosol may lead to an increase in nitrate aerosol (if ammonia and not NO_x is the limiting factor for ammonium nitrate formation), which could partially balance the climate effects.

Table 3.1 provides model estimates of the global burden of several human influenced aerosol compounds, and how they are projected to change under the assumption of the SRES A2 scenario. (The SRES A2 could be described as one of the business as usual scenario's with the largest expected temperature increase). Nitrate levels are expected to become larger than those of sulfate. Note that table 3.1 and Figure 3.10 both give the mass burden of different aerosol components; the number of particles that this relates to is not necessarily the same for each component. E.g. sea salt, dust and (to a lesser extent) nitrate are usually present in larger size fractions, so the same amount of mass corresponds to much fewer particles than the same amount of mass of e.g. sulphate, organic or black carbon would.

Table 3.1 *Model simulated estimates for global burden of the major human influenced aerosol components, including a projection for 2100 under the SRES A2 scenario (From Adams, 2001).*

Global burden (Teragrams)	Sulfate	Nitrate	Black carbon	Organic carbon
Pre-industrial	0.32	0.18	0.01	0.33
Present day	1.40	0.48	0.23	1.60
Projection for 2100	1.37	1.97	0.54	3.31

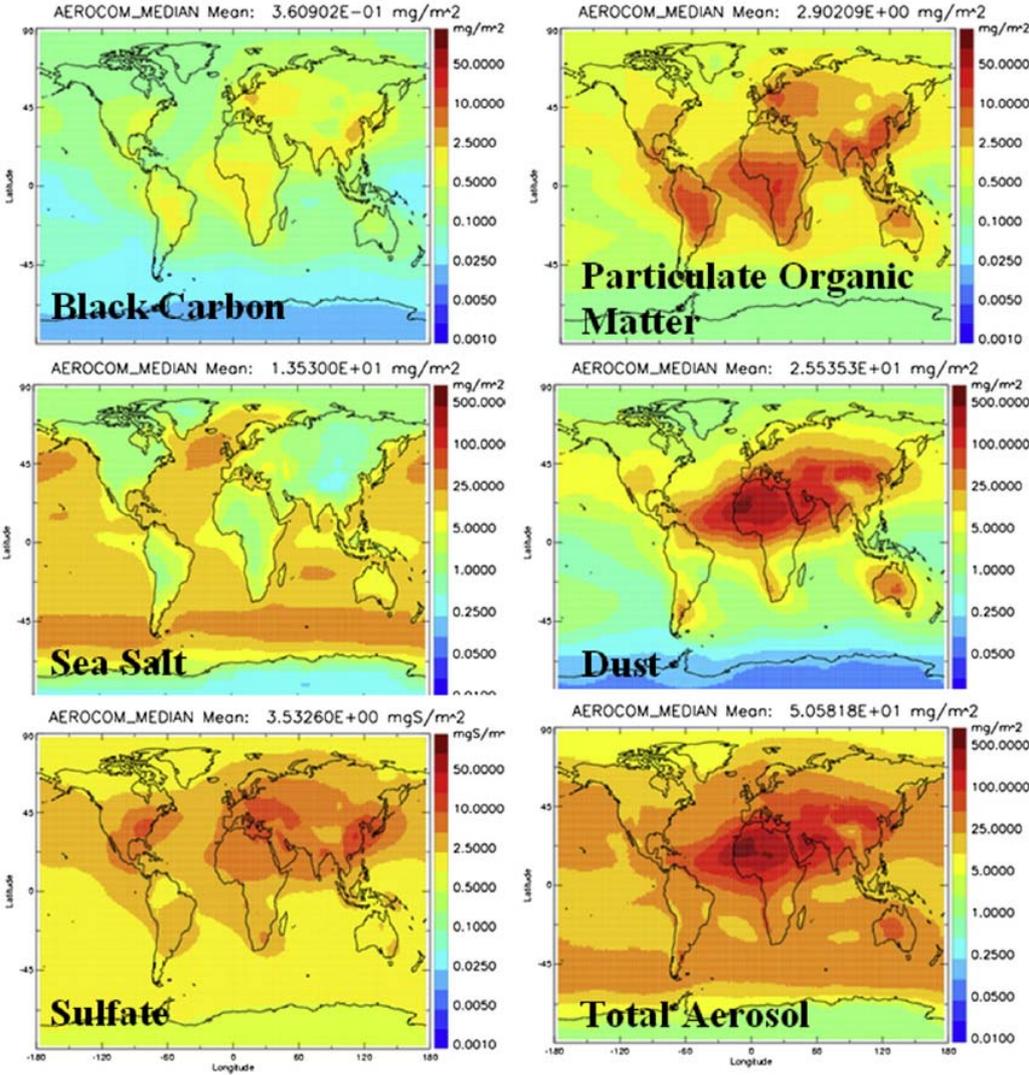


Figure 3.10 *Aerosol column burden of major components derived from the AeroCom B model simulations (median of 9 models) for the year 2000. (Isaksen et al, 2009).*

The aerosol burden depends to a large extent on the emissions of gaseous precursors, such as sulfur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH₃) and volatile organic compounds (VOC's). Emission reductions of these pollutants have been mostly achieved in developed nations, though eventually developing countries are expected to follow suit (primarily for public health reasons).

For the coming decades, SO₂ emissions are predicted to continue their decline over North America and Europe, but to increase in China and India. The emissions in other parts of the world are not predicted to change strongly. The same pattern of predicted change is valid for NO_x (Isaksen et al., 2009). Most VOC's are emitted by the biosphere, and its future development is uncertain. Some hypothesize that increased temperatures will lead to increased biogenic emissions of e.g. terpenes, which contribute to secondary organic aerosol (SOA) formation (e.g. Carslaw et al., 2009). Direct aerosol emissions, e.g. from transport, are also expected to be further curbed.

Two considerations are important for estimates of future climate impact from man-made emissions: The short chemical lifetime of most climatically active compounds as compared to CO₂, and the possibility for mitigation options to reduce emissions. In light of these considerations, it is likely that in the future (timescale of decades to centuries) the relative contribution of CO₂ to climate change will increase even further (Isaksen et al., 2009).

Raes and Seinfeld (2009) recently published a short article outlining the complex relation between greenhouse gas warming and aerosol cooling. Figure 3.11 below shows their schematic of the temperature evolution, how it would have been in the absence of aerosols (i.e. only greenhouse gas warming, dotted lines) and how it has been (and is expected to be) in the presence of aerosols. The same considerations as mentioned above are central to this schematic: Aerosol sources are expected to be cleaned up because of their immediate health hazard, and aerosols (and their precursors) are much shorter lived in the atmosphere than CO₂ is.

The flattening of the temperatures during the middle of the 20th century, as a consequence of aerosol cooling masking part of the greenhouse warming, can be clearly discerned (i.e. the solid red line running below the red dotted line from the 1940's to the 1980's). As aerosol sources are cleaned up, the temperature 'quickly' climbs up to almost the same level as the greenhouse gas only trend (i.e. the solid lines rising rapidly the approach the dotted lines between now and the 2030's).

For long term temperature stabilization, it is imperative that CO₂ emissions are eventually reduced to near zero. In the short term however (i.e. decades), emission reduction measures could marginally increase the rate of warming, because such reductions are often accompanied by simultaneous air pollution (including aerosol) reductions. This is e.g. the case for many energy related mitigation measures, since fossil fuel combustion leads to the simultaneous emission of air pollutants (including aerosol precursors). This is the reason that initially, the blue solid line runs slightly above the red solid line in Figure 3.11b.

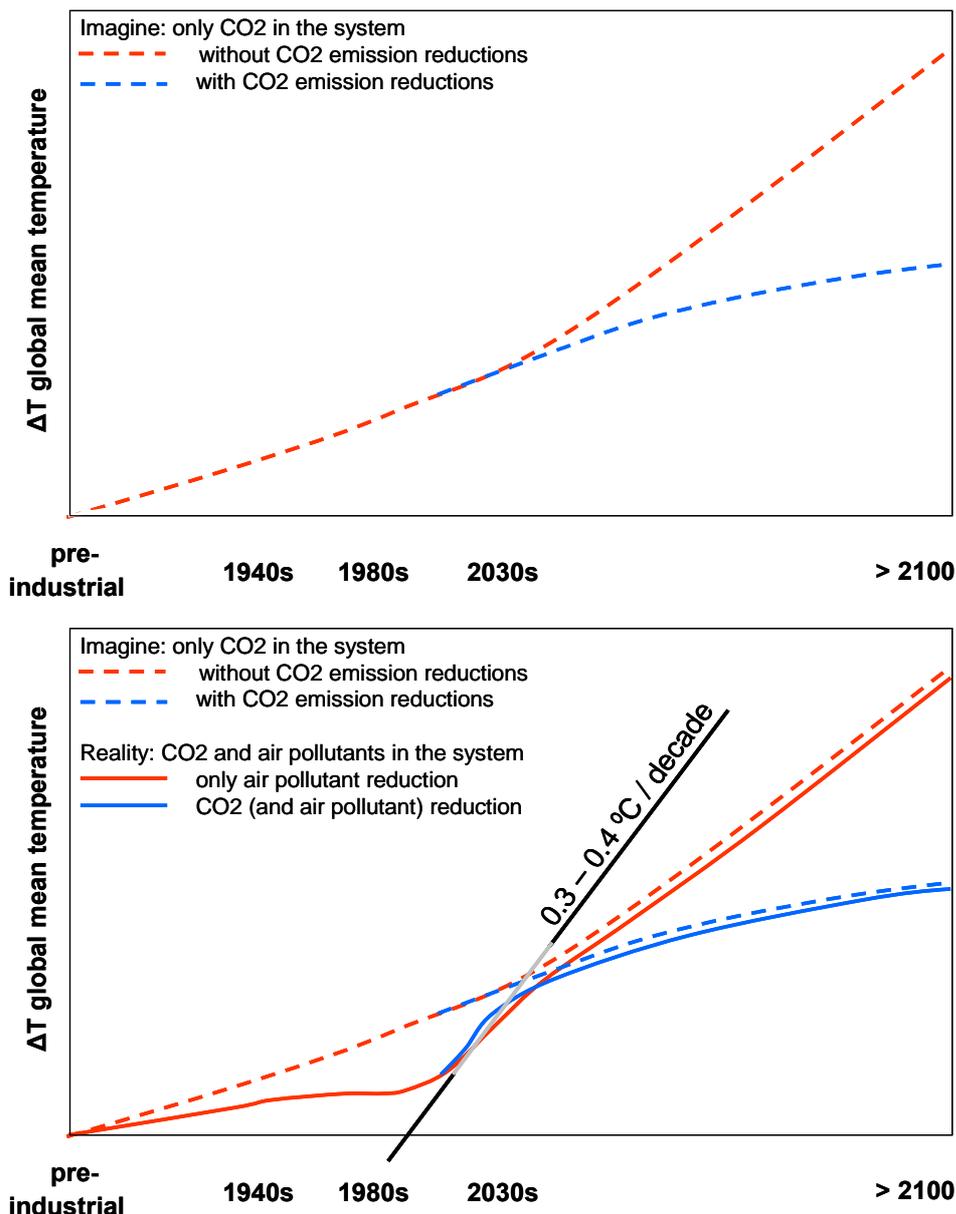


Figure 3.11 *The figure shows schematically the climate consequences of climate change and air pollution (particulate matter) abatement strategies. Not considering aerosols, greenhouse gas reductions would slowly lead to a stabilization of temperatures. The intended stabilization is only reached slowly, because greenhouse gases are long-lived in the atmosphere and because of the slow ocean response time (dotted lines, top panel). In reality, cooling aerosols have masked part of the greenhouse warming in the 20th century (as is evident from the solid red line running below the dotted line; lower panel). Air pollutant reductions are expected to accelerate the increase in global mean temperature in the short term (< 30 years; the black line denotes the hypothetical rate of warming), on the way to eventual long-term (> 100 years) climate stabilization under the influence of CO₂ reductions. (From Raes and Seinfeld, 2009)*

While the schematic above is probably valid for the globe as a whole, different regions follow different trajectories in terms of their emissions, as discussed in the previous section. It is therefore expected that different regions are on a different point of the warming trend, i.e. Europe is expected to reach the 2030's inflection point the earliest, followed by North America.

Philipona et al. (2009) hypothesize that with anthropogenic aerosols now reaching low and stable values over Europe, solar brightening will subside and European temperature trends for the next decades will likely reduce to about $+0.2^{\circ}\text{C}$ per decade. In other words, they argue that (Western) Europe is already at the point marked 2030's in Figure 3.11: We have experienced the extra fast warming due to aerosol reductions over the past 2 decades (0.38 degrees per decade, see Figure 3.8), and soon the warming trend will resume to the underlying greenhouse trend. No strong corroboration of this hypothesis is presently available, and it should be noted that the dataset they used is limited both in temporal as well as in spatial scale. The attribution of regional temperature trends is more complicated than that of global trends, because the lower 'signal-to-noise' ratio when averaging temperatures over a smaller area. Other factors can also influence regional trends, e.g. land surfaces have warmed more than ocean surfaces.

It should be emphasized that the expected warming further into the future (i.e. after 2040) is mainly dependent on the development of atmospheric greenhouse gas concentrations, and thus on mitigation activities and policies that are enacted in the coming decades. In a business-as-usual scenario (i.e. no substantial CO_2 reductions) the long term rate of warming could increase to values (far) in excess of 0.2°C per decade.

Aerosol concentrations are expected to be reduced globally, but at a different pace in different regions. Reductions will be achieved first in developed countries, whereas in developing countries the aerosol burden may first increase before it starts to decline at a later stage. A consequence of reductions in aerosol concentrations will be a more pronounced warming, as the underlying greenhouse warming trend will be revealed.

3.4 Climate-active emissions for selected sources

Fossil fuel and biomass burning

There are many linkages between the emissions of different climatically active compounds: Fossil fuel burning leads to emissions of both greenhouse gases (warming), black carbon (warming), sulfate and organic aerosol (cooling) and ozone (warming). The net effect of fossil fuel burning depends on the timescale of interest and on the way the emissions change in time. In the long term, the CO_2 induced warming inevitably dominates. In the short and medium term, however, it can go either way.

For coal-fired power plants, steadily increasing emissions without pollution controls do not cause warming (from CO_2), at least as long as the aerosol cooling (via emissions of SO_2 and NO_x) cancels the greenhouse warming. In the long run greenhouse warming will prevail, and large regional differences in climate forcing can still result from such coal fired power plants. On small spatial and temporal scales they may even lead to slight cooling dependent on the exact magnitude of aerosol indirect effects. This was the situation for coal-fired power plants up to ~1970: Their greenhouse warming and aerosol cooling effects nearly cancelled during that period. The aerosols that were emitted have long since disappeared, but a substantial part of the CO_2 is still around, exerting its warming effect.

Pollution control since then has led to a decrease in aerosol production from power plants. The masking by aerosol cooling gradually decreased, and greenhouse warming became evident since the 1970's (Shindell, 2009).

Black carbon is a primary aerosol emitted directly at the source from incomplete combustion processes such as fossil fuel and biomass burning. See Figure 3.12 for the different sources of three main aerosol components (black carbon, organic carbon and sulfate).

A similar pattern is valid for biomass burning, which can cause short term cooling (due to aerosols) but long term warming (due to greenhouse gases) (Jacobson, 2004). Aerosol from

biomass burning consists mainly of organic and black carbon. Their direct climate effects nearly cancel each other (direct radiative forcing estimated at $0.03 (\pm 0.12) \text{ W/m}^2$, IPCC, 2007). Note that this estimate does not include the indirect effects of the aerosol on cloud properties; they exert an additional negative forcing.

The climate effect of black carbon depends strongly on the albedo of the underlying surface. When above clouds, biomass burning aerosol can exert a significant positive radiative forcing (i.e. warming effect). This is due to radiation being absorbed which would otherwise be very effectively scattered by the reflective cloud. In clear skies the biomass burning aerosol exerts a negative forcing (cooling). Figure 3.13 gives the radiative forcing estimates of the major aerosol compounds, and how these estimates changed over time in the consecutive IPCC reports.

Cattle farming also contributes to aerosol cooling (via emissions of NH_3) in addition to greenhouse warming (via emissions of CH_4 , CO_2 and N_2O).

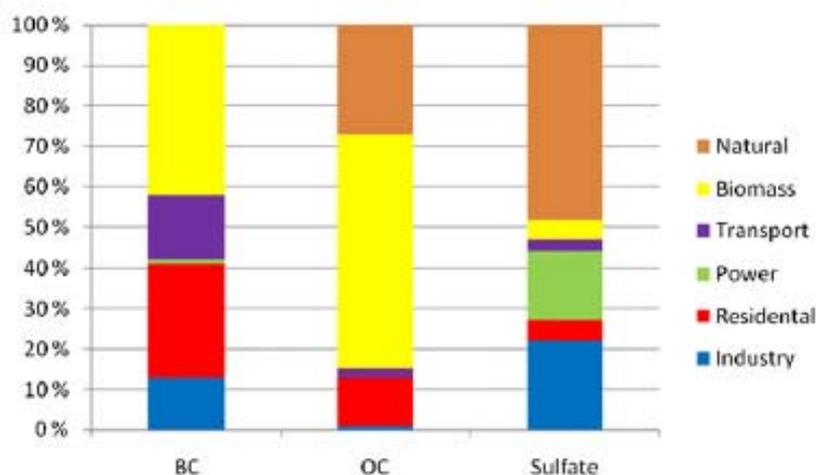


Figure 3.12 Sectoral contributions to aerosol forcings shown as percentages relative to total “top-of-atmosphere” forcing for each species. Results are taken from Koch et al. (2007). (From Isaksen et al, 2009)

	Global mean radiative forcing (W m^{-2}) ^a			Summary comments on changes since the TAR
	SAR (1750–1993)	TAR (1750–1998)	AR4 (1750–2005)	
Direct sulphate aerosol	-0.40 [2x]	-0.40 [2x]	-0.40 [± 0.20]	Better constrained
Direct fossil fuel aerosol (organic carbon)	Not evaluated	-0.10 [3x]	-0.05 [± 0.05]	Re-evaluated to be weaker
Direct fossil fuel aerosol (BC)	+0.10 [3x]	+0.20 [2x]	+0.20 [± 0.15]	Similar best estimate to the TAR. Response affected by semi-direct effects
Direct biomass burning aerosol	-0.20 [3x]	-0.20 [3x]	+0.03 [± 0.12]	Re-evaluated and sign changed. Response affected by semi-direct effects
Direct nitrate aerosol	Not evaluated	Not evaluated	-0.10 [± 0.10]	Newly evaluated
Direct mineral dust aerosol	Not evaluated	-0.60 to +0.40	-0.10 [± 0.20]	Re-evaluated to have a smaller anthropogenic fraction

Figure 3.13 Global mean direct radiative forcing of major aerosol species since 1750 and comparison with earlier IPCC assessments. Cloud albedo effect is only provided as an aggregate estimate of $-0.70 [-1.1, +0.4] \text{ W/m}^2$. Other indirect effects have not been quantified. (From IPCC, 2007)

Biomass use

The argument is often made that biomass is carbon neutral, because the carbon that is released from burning is taken up again at regrowth. However, this is only a small part of the picture of climate effects associated with biomass. Significant impacts on climate are associated with the production of biomass (e.g. through the use of fertilizer and associated nitrous oxide emissions) and with the change in land use that often accompanies the growing of more biomass. If 'new' land is used to grow (energy or food) crops, considerable changes in the carbon balance are often the result. The prime example is the burning of tropical rainforest to make space for the growing of crops or grass (De Santi et al, 2008), but it also plays a role in temperate regions².

Use of biomass as fuel is expected to increase in the Netherlands (as well as in other industrialized nations). This will lead to an increase in aerosol emissions. Industrial use of biomass generally leads to smaller particles than private wood burning (de Wilde et al., 2006). As a first approximation, climate effects are most strongly dependent on the number concentration, so industrial use of biomass leads to more short-term cooling than private stoves do (for the same amount of biomass burnt).

Alternative strategies

As explained above, the long term climate effects are most strongly dependent on CO₂ emissions (due to its long lifetime, and due to the expected increase in its emissions). However, since part of the CO₂ induced warming has been cancelled by simultaneous aerosol cooling, the observed warming until now could to a large extent be ascribed to the non-CO₂ greenhouse gases.

Based on that interpretation, Hansen and co-authors (Hansen, 2000) have suggested that the initial focus should perhaps be on those non-CO₂ greenhouse gases (methane, nitrous oxide, halocarbons, ozone) and black carbon. For the long term, the CO₂ forcing is however dominant, so CO₂ reduction should not take a backseat. The reduction of sulfur emissions has increased the net warming effect of fossil fuel burning (at least in industrialized countries).

In order to avoid abrupt climate change, fast-action regulatory strategies may be required, that can complement (the much slower acting) CO₂ emission reductions. Examples of such strategies are (Molina, 2009a; 2009b):

- Phasing down the use of halocarbons (HFC's and HCFC's). These chemicals replaced the banned CFC's, and while they do not substantially affect the ozone layer, they exert a large greenhouse effect.
- Reducing black carbon (which has important ancillary health benefits). Black carbon is thought to have disproportionately large effects on the melting of snow and ice in vulnerable regions such as the Arctic and Himalayan glaciers. Reducing its concentration has fast and substantial effects on reducing climatic warming.
- Reducing ozone precursors (such as CO, NO_x and VOC's). As is the case with black carbon, this has both health and climate benefits.

² Searchinger et al (2008) estimated the magnitude of this 'indirect land use effect' and concluded that it is a potentially very important factor in the total climate impact of producing and using biofuels, even though it is usually not included in life cycle analyses. It may take several hundreds of years for the carbon released by exploiting new land to be offset by the carbon saved from displacing fossil energy. These adverse consequences of changes in land use can be minimized by using biomass based on waste products, by growing biomass on degraded land, or by increasing the yield instead of using more land; the latter would however lead to increased nitrous oxide emissions. De Santi et al (2008) arrive at a very similar conclusion, though they stress the large uncertainty of the indirect land use effect, and how its magnitude depends on (control strategies in) the regions where the extra demand for crops will result in expansion of farmed area.

- Expanding and protecting carbon sinks by reforestation and the prevention of deforestation. Agricultural practices, such as the application of biochar³ could also aid in the expansion of carbon sinks.

More insight into the linkages between greenhouse gas and aerosol emissions also suggest ways in which air pollution strategies could be devised that lessen the warming expected from reducing cooling aerosols. An example is to address the transport and domestic sectors, emissions from which have a larger black carbon to sulfate ratio than those in the power and industrial sectors (Shindell et al., 2008). The efficacy of a certain measure depends critically on the timescale of interest though. Another example is the impact of the fuel sulfur content of oceangoing shipping. According to a recent modeling estimate, this causes significant (regional) cooling of around -0.5 W/m^2 globally averaged (Lauer et al., 2009). Future reductions in sulphur content (as foreseen in the IMO regulatory framework) have the potential to reduce this cooling (and thus reveal the masked warming), but it will not be entirely negated.

Many combustion processes result in the simultaneous emission of (warming) greenhouse gases and (cooling and warming) aerosols. Due to the different lifetimes of these compounds, the result can be short term cooling (e.g. biomass burning) or no short term impact (coal fired power plant), but long term warming (all sources). In devising policies to curb health effects, simultaneous climate impacts should be considered, and vice versa. It is becoming more and more clear that regional air quality and global climate issues are intricately linked with one another.

³ Biochar is based on low temperature pyrolysis (burning in the absence of oxygen) of biomass. By adding biochar to soil, carbon is stored for millennia. It also helps retaining nutrients, fertilizer and water, potentially reducing emissions of nitrous oxide (a potent greenhouse gas), increasing crop yield and increasing resistance to drought.

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