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Atmospheric aerosols in the earth system: a review of interactions and feedbacks

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Abstract

The natural environment is a major source of atmospheric aerosols, including dust, secondary organic material from terrestrial biogenic emissions, carbonaceous particles from wildfires, and sulphate from marine phytoplankton dimethyl sulphide emissions. These aerosols also have a significant effect on many components of the Earth system such as the atmospheric radiative balance and photosynthetically available radiation entering the biosphere, the supply of nutrients to the ocean, and the albedo of snow and ice. The physical and biological systems that produce these aerosols can be highly susceptible to modification due to climate change so there is the potential for important climate feedbacks. We review the impact of these natural systems on atmospheric aerosol based on observations and models, including the potential for long term changes in emissions and the feedbacks on climate. The number of drivers of change is very large and the various systems are strongly coupled. There have therefore been very few studies that integrate the various effects to estimate climate feedback factors. Nevertheless, available observations and model studies suggest that the regional radiative forcings are potentially several Watts per square metre due to changes in these natural aerosol emissions in a future climate. The level of scientific understanding of the climate drivers, interactions and impacts is very low.

1 Introduction

Aerosols are important components of most parts of the Earth system. In the atmosphere, they affect the radiative balance by scattering and absorbing radiation and affecting the properties of clouds (Haywood and Boucher, 2000). In the cryosphere, deposition of light absorbing carbonaceous and dust particles on ice and snow impacts the surface albedo and absorption of solar radiation (Warren and Wiscombe, 1980; Krinner et al., 2006), leading to modification of the snow/ice-albedo feedback mechanism (Flanner et al., 2007). The terrestrial biosphere is a major source of primary aerosol and aerosol precursor gases that form secondary organic aerosol (SOA), and the net primary productivity of plants can be influenced by the effect of aerosol on diffuse radiation (Gu et al., 2003). A large fraction of atmospheric dust is natural and is projected to increase in a future climate partly in response to vegetation changes (e.g., Mahowald et al., 2003, 2006). The oceans emit aerosol precursor gases such as dimethylsulphide (DMS) and primary sea spray particles, and marine biota are supplied with nutrients from aerosol deposition, particularly dust, which can impact the carbon cycle.

These natural systems make a very large and temporally variable contribution to global atmospheric aerosol. For example, observations from around the world suggest that in many places more than half of the aerosol mass is organic (Zhang et al., 2007a) and is dominated by compounds of biogenic origin (Hallquist et al., 2009). Likewise, model studies suggest that the oceanic emission of dimethylsulphide from plankton accounts for 42% of the global atmospheric sulphate aerosol mass (Chin and Jacob, 1996). There is therefore the potential for substantial long term changes in the atmospheric aerosol burden if the processes that drive the emissions are perturbed by climate change.

Natural aerosol sources vary substantially with climate on seasonal, interannual and decadal timescales. For example, wildfire aerosol emissions have an interannual variation of more than a factor of 2 driven partly by internal climate variability such as El Niño (Schultz et al., 2008), and decadal climate change seems to be affecting the occurrence of forest fires in some regions (Gillett et al., 2004; Westerling et al., 2006). In the boreal forest, particle growth rates correlate with seasonal variations in vegetation gross primary productivity, most likely due to varying emissions of terpenes that form SOA (Kulmala et al., 2004b), and interannual variations in temperature probably control large changes in biogenic isoprene (Palmer et al., 2006). There is evidence from ice cores that a tracer of marine DMS emissions varies on decadal and centennial timescales (Legrand, 1997), while seasonal variations in DMS from plankton in the Southern Ocean (Ayers and Gras, 1991; Ayers et al., 1997; Sciare et al., 2000a, b) appear to drive substantial changes in sulphate aerosol mass and cloud properties (Boers et al., 1994). Atmospheric dust is also changing partly due to climate effects. For example, dust concentrations at Barbados show a four-fold increase since the 1960s driven by meteorological changes in the African source region (Prospero and Lamb, 2003).

In addition to direct observational evidence for the effects of climate on natural aerosol emissions, process studies strongly suggest that large climatic effects can be expected. For example, the vegetation emissions of many terpene species that form SOA in the atmosphere are affected by temperature, radiation, soil moisture, foliar biomass, atmospheric composition, and vegetation type (Guenther et al., 1995, 2006), all of which are likely to change with climate. Likewise, the occurrence of forest fires is clearly related to temperature and hydrology (Westerling et al., 2006) and dust emissions to vegetation, soil wetness and wind speed, among other variables.

This review describes the response of these natural atmospheric aerosols to climate and environmental change on the century timescale. The climate drivers of changes in aerosol can be split broadly into physical changes (temperature, soil wetness, solar radiation, wind speed, sea ice extent, etc.) and biological changes (vegetation, plankton, etc.), although these changes are strongly coupled. The components of the Earth system respond to climate change and interact in ways that drive non-linear changes in aerosol emissions and sinks. A further important influence on the aerosol abundance and properties is the long term change in atmospheric composition, in particular the oxidative processes that drive production of secondary aerosol products from the emitted gases. For example, stratosphere-troposphere exchange of ozone may increase in a warmer climate (Butchart and Scaife, 2001; Collins et al., 2003), which could lead to increased tropospheric concentrations of ozone, an important oxidant. Atmospheric aerosol is itself a non-linear system and the impact on climate depends on the microphysical and chemical properties of the particles and not just their mass, and these can change non-linearly with emissions. For example, new particle formation, which is an important source of climate-relevant particles over forests, is suppressed by high particle loadings, so increased aerosol mass due to enhanced primary emissions could reduce net changes in number concentration (e.g. Spracklen et al., 2006; Mönkkönen et al., 2004).

These responses of the natural aerosol system to climate change may constitute a climate feedback. Feedbacks are processes that amplify or dampen the effect of a forcing. The climate system responds to radiative forcings – from greenhouse gases and aerosols in the first place – by adjusting the surface and atmospheric profile of temperature in order to maintain a radiative balance at the top of atmosphere. The role of non-radiative forcings is now also recognised (Betts et al., 2007; Boucher et al., 2009). However various parameters in the climate system respond to the temperature adjustment caused by the initial radiative imbalance. Water vapor, snow albedo and sea-ice albedo are examples of positive feedbacks that amplify the effects of a radiative forcing. It is now realised that biogeochemical cycles can play a role in climate feedbacks. While the importance of the carbon cycle in climate feedback is well accepted, the role of aerosols is much less well established.

We focus on four aerosol systems: terrestrial biogenic aerosol (including aerosols from biogenic gases and wildfires), marine aerosol, soil and desert dust, and stratospheric and volcanic aerosol. Although we discuss each aerosol system in a separate section, our review highlights that these aerosol systems are not self-contained and that there are interactions and feedback connections between them (Fig. 1). For example, wildfires alter the distribution of vegetation whose emissions of volatile organic compounds account for a large fraction of global aerosol mass, and changes in dust emissions directly impact aerosol radiative forcing but may also impact DMS emissions through changes in how dust fertilises the ocean with iron.

The status of our understanding of these aerosol systems, their interactions and feedbacks varies enormously. The DMS-aerosol-cloud-climate feedback (Charlson et al., 1987) is perhaps the most studied, with 20 years of research on marine biota, atmospheric chemistry and aerosol formation. Nevertheless, quantitative understanding of the direction and magnitude of the feedback in different parts of the ocean is still lacking, partly because of the challenge of scaling up and coupling the relevant processes in Earth system models. In other cases, such as wildfires, some potential impacts of climate change on regional fire distribution have been investigated but there has been no attempt to study feedbacks in an integrated way or on a global scale. Only in rare cases have these processes been incorporated in global models to calculate climate feedback factors.

The review is organized as follows. Sections 2 to 5 describe aerosol processes, interactions and feedbacks related to terrestrial biogenic aerosol, marine aerosol, stratospheric aerosol and dust. These sections describe the main sources of natural aerosol, the climatic factors controlling emissions as derived from observations and process models, and the status of their treatment as components of climate and Earth system models. In Sect. 6 we summarise the status of knowledge and identify where progress is needed in terms of observations and model development.

2 Terrestrial biogenic systems

The terrestrial biosphere emits primary biological aerosol particles (PBAP) and trace gas species that can partition into the particulate phase to form SOA. The terrestrial biosphere also makes sporadic and very large contributions to atmospheric aerosol during wildfires. In this section we evaluate potential feedbacks between the terrestrial biosphere, atmospheric aerosol and climate. Recent studies have started to quantify these feedbacks, but poorly quantified emissions and a limited understanding of aerosol formation mechanisms mean that the relevant climatic controls are not well defined. The direction and magnitude of future changes in aerosol and precursor gas emissions from the terrestrial biosphere are therefore very uncertain.

The interaction between the terrestrial biosphere and atmospheric aerosol operates in both directions. While the biosphere affects aerosol directly through emissions, the abundance and properties of aerosol can also affect the functioning of the biosphere through effects on photosynthetic radiation and nutrient supply. These effects are evaluated in Sect. 2.4.

2.1 Secondary organic aerosol from terrestrial sources

2.1.1 The impact of secondary organic aerosol on the atmosphere

Trace gas emissions from the terrestrial biosphere include biogenic volatile organic compounds (BVOCs), such as isoprene, terpenes, acetone and methanol, with an estimated total global source of carbon of 1150 Tg a^{-1} (Guenther et al., 1995). Once in the atmosphere, some BVOCs react to produce compounds that can partition into the particulate phase, forming SOA. The budget of SOA is very uncertain and current best estimates of 12–70 Tg a^{-1} (Kanakidou et al., 2005) may be an order of magnitude too small (Goldstein and Galbally, 2007). This uncertainty is due to our limited understanding of the principal SOA precursor gases, the magnitude of their emissions, and the dominant SOA formation mechanisms, which can include condensation and evaporation of semivolatile oxidation products, heterogeneous reactions, cloud processing and oligomerization/polymerization (Fuzzi et al., 2006).

SOA dominates sub-micron aerosol mass in a wide range of continental environments (Kanakidou et al., 2005; Zhang et al., 2007a) and therefore has a direct effect on atmospheric radiation. Additionally, SOA is important for the condensational growth of newly formed particles up to cloud condensation nuclei (CCN) sizes of 10's of nanometers (O'Dowd et al., 2002; Allan et al., 2006; Laaksonen et al., 2008). New particle formation (conversion of gas phase species to new particles) is observed in the air above terrestrial ecosystems all around the world (e.g. Kulmala et al., 2004a) and contributes between 5 and 50% to global mean CCN concentrations in the boundary layer (Kerminen et al., 2005; Spracklen et al., 2008a). In the boreal forest, particle growth rates correlate with seasonal variations in vegetation gross primary productivity and with monoterpene concentrations and radiation (Kulmala et al., 2004b). Whether biogenic emissions also control particle formation rates is unknown, although organics have been shown to enhance nucleation rates in laboratory experiments (Zhang et al., 2004; Verheggen et al., 2007) and a terpene-ozone mechanism has been proposed as an explanation for ambient particle formation rates (Bonn et al., 2008).

The contribution of terrestrial ecosystems to atmospheric aerosol is difficult to quantify accurately from observations because anthropogenic aerosol sources can mask natural cycles in many locations. Molecular marker methods and ^{14}C analysis suggest that biogenic aerosol sources dominate organic carbon aerosol even in urban locations (Hallquist et al., 2009, and references therein). Observations at several boreal forest sites that are relatively free of anthropogenic pollution (Andreae et al., 2007) suggest a very large natural modulation of aerosol by forest emissions. For example, aerosol mass is proportional to the time that air has spent over Scandinavian forest (Tunved et al., 2006a), with 12–50% of aerosol mass and ~50% of CCN coming from forest sources (Tunved et al., 2008). Kurten et al. (2003) used observations of new particle formation events made over the boreal forest in southern Finland to estimate that aerosol formation leads to a radiative perturbation over the forest of between −5 and $-14\,\mathrm{W\,m^{-2}}$ (global mean −0.03 to $-1.1\,\mathrm{W\,m^{-2}}$). Spracklen et al. (2008b) used a global aerosol model to predict that SOA from boreal forest terpene emissions doubles regional CCN concentrations compared to emissions from grassland and causes a local radiative forcing of between −1.6 and $-6.7\,\mathrm{W\,m^{-2}}$ due to changes in cloud albedo.

2.1.2 Feedback mechanisms involving biogenic SOA

Climate exerts a strong control over the emission of BVOCs, directly by modifying the emission rate through variables such as temperature, soil moisture and radiation, and indirectly by modifying vegetation distribution, species composition, primary productivity and foliar biomass.

Increases in temperature are likely to lead to increased BVOC emissions and aerosol concentrations, resulting in increased aerosol radiative cooling and a potential negative feedback mechanism linking forests, aerosols and climate (Kulmala et al., 2004b). There is observational support for such a hypothesis. Tunved et al. (2008) used correlations between observed aerosol at boreal forest sites and cumulative monoterpene emissions calculated over atmospheric back trajectories to estimate that a 1.4 K increase in temperature would increase CCN concentrations over Scandinavia by 8%

and a 5.8 K increase in temperature would increase CCN concentrations by 40%. Using the same approach as Spracklen et al. (2008b), this change in CCN would cause a local radiative forcing of between -0.6 and $-2.7\,\mathrm{W m^{-2}}$ due to changes in cloud albedo, assuming as an upper limit that all the CCN are active in cloud drop formation. The temperature-induced change in forest terpene emissions therefore represents a substantial negative climate feedback.

The atmospheric concentration of SOA is not determined solely by emissions of BVOCs, but also by temperature, precipitation and atmospheric chemistry, resulting in non-linear interactions between biogenic emissions and the SOA burden (Tsigaridis et al., 2005). Temperature affects the condensation of semi-volatile compounds to the aerosol phase, with increasing temperature resulting in decreased aerosol partitioning. The interactions between such processes further complicate how the distribution of SOA might change with climate. For example:

1. Increased temperature causes increased BVOC emissions (e.g., Guenther et al., 1995) but also reduces the partitioning of semi-volatile compounds to the particles. Increased temperature also modifies vegetation resulting in either further increased or decreased BVOC emissions.

2. Increased CO_2 concentrations may inhibit leaf-level isoprene emission (Arneth et al., 2007a, b) while simultaneously fertilizing plant growth, increasing emission rates.

2.1.3 Response of BVOC emissions to environmental change

BVOC emissions are controlled by a complex set of variables including leaf temperature, radiation, nutrient availability, soil moisture, foliar biomass, leaf age, atmospheric composition, vegetation type and species composition (Guenther et al., 1995, 2006), all of which are likely to change with climate. It has been predicted using a dynamic forest model that the concentration of reactive nitrogen (i.e. nitrogen, which plants can use) in soil will increase during the next 100 years (Hari et al., 2008). This effect alone will double the leaf area before 2100 and enhance BVOC emissions by factor of 2 (Bäck and Hari, 2008). Current BVOC emission algorithms treat only a subset of these variables. For example, they do not include the effect of changing O_3 concentrations (Loreto et al., 2004; Velikova et al., 2005), nutrient availability or physical stress and are only beginning to explore the effects of changing CO_2 concentrations.

The prediction of changes in BVOC emissions requires a BVOC emission model coupled either to meteorological observations or to meteorological output from a climate model. The interannual variability in BVOC emissions can yield useful information on how emissions are likely to change in a future climate. Using a land surface-vegetation model driven by observed climate Lathière et al. (2006) estimate that global emissions of biogenic organics varied by 8% from 1986 to 1995. Similarly, Levis et al. (2003) predict that global annual mean biogenic emissions vary by up to 5% and monthly mean emissions by up to 18%. Palmer et al. (2006) used a 6 year record of satellite-observed formaldehyde column to infer a 20–30% interannual variability in isoprene emission over the southeastern United States driven primarily by variations in surface air temperature.

2.1.3.1 Isoprene emissions

The vast majority of studies of future BVOC emissions have assessed possible changes in isoprene, taking into account changes in temperature alone, temperature and vegetation, land cover changes and the influence of CO_2 (Table 1). Although the importance of isoprene for SOA is still unclear (Clayes et al., 2004; Henze and Seinfeld, 2006), these studies indicate the typical responses that might be expected for other BVOCs. Studies of monoterpenes changes are discussed in Sect. 2.1.3.2).

Assuming fixed vegetation, changes in temperature are thought to be the dominant driver of isoprene emissions, with other variables such as radiation, cloud cover and soil moisture changing by less than 5% between 2000 and 2100 (Heald et al., 2008). Sanderson et al. (2003) used the IS92a greenhouse gas scenario to predict that a 4.7 K increase in global temperature will cause a 34% increase in isoprene emissions by the 2090s. Wu et al. (2008) used the A1B emissions scenario to predict temperature increases of 1.6 K and global isoprene emission increases of 25% by 2050 and Heald et al. (2008) used the same scenario to predict a 1.8 K increase in temperature and a 37% increase in isoprene emissions by 2100. Using a regional model and the A1B emissions scenario Jiang et al. (2008) predict a 20% increase in isoprene emissions within the Houston area of Texas by 2050.

Climate change results in complex changes to vegetation which can have very important but highly uncertain impacts on BVOC emissions. Projected climate driven changes to vegetation include longer growing seasons, increased leaf area index and changes in water stress. This may result in changes to vegetation distribution potentially including expansion of boreal and temperate forests (Lathière et al., 2005) and dieback of the Amazon forest (Cox et al., 2004, 2008). However, previous studies disagree as to whether the vegetation changes over the 21st century are likely to result in increased or decreased BVOC emissions. Turner et al. (1991) predict that under a $2\times CO_2$ environment vegetation change will cause a 25% increase in isoprene emissions due to an expansion of tropical humid forests. Heald et al. (2009) predict that vegetation changes amplify 21st century changes to isoprene emissions from an increase of 37% with fixed vegetation to a factor of 3.6 increase with dynamic vegetation. In contrast, Sanderson et al. (2003) predict that dieback of the Amazon tropical forest reduces the projected 21st century increase of isoprene emissions from 34% to 27%. Wiedinmyer et al. (2006) also predict that vegetation changes including the replacement of natural forest with grasslands offsets some of the climate driven changes to isoprene emissions. In all these studies the simulated vegetation distribution is in equilibrium with the climate, which will not be the case under transient climate change. Additionally, nutrient limitations not accounted for in these studies may limit the response of vegetation to elevated CO_2 concentrations.

The BVOC emissions predicted for future climates are further modified by potential anthropogenic changes to land cover. Lathière et al. (2006) predict that global tropical deforestation would reduce global isoprene emission by 29%. Guenther et al. (2006) use the IMAGE landcover database to calculate that anthropogenic land cover change (mainly tropical deforestation and expansion of croplands) reduces 2100 isoprene emissions by more than 30%. Heald et al. (2008) predicts that an A2 land-use scenario reduces global isoprene emissions by 15% by 2100. Wiedinmyer et al. (2006) predict that replacing 50% of both the Amazon and the southeastern United States with urban and pasture landuse reduces global isoprene emissions by 9%. Whereas replacement of forest with crops normally leads to reduced isoprene emissions, some crop species, such as oil palm, have large isoprene emission factors and the ongoing expansion of this crop in SE Asia may be leading to regionally increased isoprene emissions. Under a scenario where 25% of the Western United States was replaced by poplar plantations and 25% of the Amazon was replaced with oil palm and Eucalyptus plantations, Wiedinmyer et al. (2006) predict that global isoprene emissions would increase by 37%.

Model studies predict that BVOC emissions have increased since the last glacial maximum (LGM) due to higher temperatures and changes to vegetation (Adams et al., 2001; Lathière et al., 2005; Valdes et al., 2005; Kaplan et al., 2006). Valdes et al. (2005) and Kaplan et al. (2006) calculate BVOC emissions between the LGM and 1850, respectively predicting that isoprene emissions increased by a factor of 2.6 and 61%, while monoterpene emissions have increased by 80% and 51%. While both Adams et al. (2001) and Lathière et al. (2005) predict that isoprene emissions have approximately doubled since the LGM they disagree as to the dominant cause of the change. Adams et al. (2001) suggest that the direct effect of temperature on BVOC emissions dominates whereas Lathière et al. (2005) emphasise the role of climate-induced changes to vegetation distribution. Lathiere et al. (2005) predict that monoterpene emissions have increased by a factor of 3.5 over this period.

The impact of changing CO_2 concentrations on BVOC emissions is a major uncertainty that needs to be further understood. Increasing CO_2 concentrations may inhibit isoprene emissions (Sharkey et al., 1991; Rosenstiel et al., 2003; Possell et al., 2005; Arneth et al., 2007a) potentially offsetting some of the increased emissions due to higher temperature and changes in vegetation. Arneth et al. (2007b) predict that when CO_2-isoprene inhibition is accounted for, global isoprene emissions from the LGM to 2100 are maintained to within ±15% of present day values. Heald et al. (2009) predicts that CO_2-isoprene inhibition reduces isoprene emissions in 2100 by about 30% meaning that with fixed vegetation isoprene emissions are projected to decline slightly over the 21st century (–8%), whereas with dynamic vegetation they increase by more than a factor of 2.

2.1.3.2 Monoterpene emissions

In aerosol production monoterpenes and sesquiterpenes are much more effective than isoprene. Recently their concentrations, fluxes and emissions have been determined using PTR-MS with eddy covariance technique (e.g. Rinne et al., 2007; Ruuskanen et al., 2008). Raisanen et al. (2008) showed that a combination of increased CO_2 and temperature can substantially increase monoterpene emissions from Scots Pine trees, but the individual effects were small. Assuming no changes to vegetation, Heald et al. (2008) use the A1B emissions scenario to predict that changes to climate will increase monoterpene emissions by 19%, while Liao et al. (2006) use the A2 emissions scenario to predict a 58% increase. Lathière et al. (2005) predict that changes to climate and vegetation (no direct anthropogenic change) will increase monoterpene emission by 51%. However, more accurate predictions of their future behavior as well as more continuous measurements at different environments are needed.

2.1.4 Model studies of the response of SOA to environmental change

Recently, studies have predicted future SOA mass distributions due to changes in climate, biogenic and anthropogenic emissions and land use. These studies typically combine BVOC emission models (Sect. 2.1.3) with global atmospheric chemistry models driven by meteorological output from a general circulation model (GCM). While these studies are the most sophisticated attempts to simulate this complex system they account for a limited range of the variables known to affect SOA. For example, none of these studies accounts for the potential impact of changing CO_2 concentrations on BVOC emissions.

The global SOA mass burden is predicted to increase by between 26% and 150% by 2100 (Table 2), resulting in an increase of surface level continental particulate matter concentrations of about 0.5 $\mu g\,m^{-3}$. Increased BVOC emissions account for most of the change in SOA, with changes in aerosol processes and removal rates having a minor effect (–6% to +11%) (Liao et al., 2006; Tsigaridis and Kanakidou, 2007; Heald et al., 2008). The largest future increase in SOA burden is predicted by Tsigaridis and Kanakidou (2007), driven by the largest increase in BVOC emissions primarily because this study included dynamic vegetation. Although not directly comparable, the global changes in SOA mass in Table 2 are similar to the observationally derived estimate of a 40% increase in CCN over boreal forests due to a 5.8 K increase in temperature (Tunved et al., 2008, Sect. 2.1.2). While the global burden of SOA is projected to increase in a future climate, regional reductions in BVOC emissions and SOA concentrations are possible due to shifts in vegetation (Avise et al., 2009).

We use these studies to estimate that feedbacks due to changing SOA in a future climate (year 2100) will result in a global mean direct radiative forcing of between -0.14 to $-0.82\,W\,m^{-2}$ (assuming a present day SOA burden of 0.6 Tg with an aerosol optical depth (AOD) of 0.0091 (Seland et al., 2008), and assuming a radiative forcing efficiency of $100\,W\,m^{-2}$ per unit AOD (IPCC)). This direct forcing needs to be added to the indirect effect on cloud albedo for which there is no global assessment. However, the local cloud radiative forcing of -0.6 to $-2.7\,W\,m^{-2}$ estimated in Sect. 2.1.2 for temperature increases of 1.4 and 5.8 K suggests that the overall feedback effect of changes in biogenic SOA may be large by the end of the 21st century.

2.1.5 Impacts of change in oxidising capacity on BVOCs and SOA

Changes to climate and to anthropogenic and biogenic emissions can impact atmospheric chemistry and alter the conversion rates and total amount of BVOC converted into SOA. Projected future increases in primary anthropogenic aerosol emissions result in increased SOA formation from biogenic sources (Liao et al., 2006; Tsigaridis and Kanakidou, 2007; Heald et al., 2008). The concentrations of atmospheric oxidants are susceptible to change due to changing emissions and climate. Concentrations of OH may increase in a future climate, speeding up oxidation rates and modifying the distribution of SOA (Tsigaridis and Kanakidou, 2007; Heald et al., 2008). Global emissions of NO_x are predicted to increase 17% by 2100 (under the A1B SRES scenario) leading to modified distributions of SOA; high NO_x conditions reduce the formation of SOA from isoprene and α-pinene (Kroll et al., 2005; Ng et al., 2007) while enhancing the production from sesquiterpenes (Ng et al., 2007). The SOA yield increases in the presence of sulphate (Hallquist et al., 2009, and references therein) the concentrations of which are likely to change as described in Sect. 3.1.4.

BVOC emissions may themselves alter the oxidising capacity of the atmosphere. For example, increased isoprene emissions are predicted to cause a 10–30 ppb increase in surface ozone by 2100 (e.g., Sanderson et al., 2003) and increased methane lifetime (Valdes et al., 2005; Kaplan et al., 2006; Shindell et al., 2007) both leading to increased radiative forcing and additional climate change. It has been hypothesized that the reduced methane concentrations during the last glacial maximum could be explained by a reduction of biogenic VOC emissions during colder conditions (Valdes et al., 2005). However it is not clear how much of that is a direct temperature effect and how much is due to the large reduction in vegetation cover. Moreover, there are uncertainties in the isoprene oxidation mechanism that need to be resolved (Lelieveld et al., 2008).

2.2 Primary biological aerosol particles

Terrestrial ecosystems emit a wide range of particles directly into the atmosphere, including viruses (<0.3 µm), bacteria (~0.25 µm–10 µm), fungal spores (1–30 µm), pollen (10–100 µm), plant debris and algae. These particles, known collectively as Primary Biological Aerosol Particles (PBAP), have an estimated global emission of up to 1000 Tg a^{-1} (Jaenicke, 2005). New observations have demonstrated that in many continental locations PBAP contributes 10–45% of coarse particulate mass (Elbert et al., 2007) and is capable of acting both as CCN (Ariya et al., 2004) and ice nuclei (Christner et al., 2008). In this way PBAP can alter cloud properties and precipitation with potentially important but poorly quantified climate feedbacks. Global atmospheric models are only starting to consider PBAP and a quantitative assessment of their contributions to the global aerosol burden, optical depth and CCN population has not been made.

Very little is known about the emission mechanisms or atmospheric drivers of PBAP, but it is likely that the drivers vary with PBAP type and source. Jones and Harrison (2004) reported observed PBAP concentrations that varied with a range of meteorological and phenological variables. Emissions of dry-emitted fungal spores and bacteria are enhanced under warm, dry and windy conditions, requiring wind speeds greater than $\sim 1\,\mathrm{m\,s^{-1}}$ (Jones and Harrison, 2004; Elbert et al., 2007), whereas wet-emitted spores are enhanced by high relative humidity and precipitation (e.g., Zoppas et al., 2006).

The changing distribution of vegetation with climate change may also impact PBAP emissions but it is difficult to quantify this effect without a better understanding of the role of vegetation in emissions. Bacterial particle number fluxes are greater over vegetated regions ($\sim 500\,\mathrm{m^{-2}\,s^{-1}}$; Lindemann et al., 1982) compared to $100\,\mathrm{m^{-2}\,s^{-1}}$ over bare soil (Lindemann et al., 1982) and $\sim 0.5\,\mathrm{m^{-2}\,s^{-1}}$ over deserts (Lighthart et al., 1994). While fungal spore emissions might also increase with increasing vegetation coverage, emissions have also been observed over desert and ice-covered regions (Griffin et al., 2006; Polymenakou et al., 2008). Elbert et al. (2007) estimated a fungal spore terrestrial emission rate of $200\,\mathrm{m^{-2}\,s^{-1}}$, irrespective of vegetation type or meteorological conditions.

On the global scale, Heald and Spracklen (2009) used observed concentrations of the sugar mannitol, a biotracer of fungal spores, to constrain the fungal PBAP source to $28\,\mathrm{Tg\,a^{-1}}$, 25% of which was emitted at sizes below 2.5 µm. Their optimized emissions were based on leaf area index (LAI) and atmospheric water vapor concentrations. Mahowald et al. (2008) estimated a global coarse PBAP emission of 168 Tg a^{-1} based on above-ground biomass density.

It is difficult to quantify the impact of climate change on PBAP emissions without a better understanding of the emission mechanisms and drivers, although projected increases in above-ground biomass and LAI in a future climate are likely to drive increased emissions. The radiative feedback through changed PBAP concentrations is unlikely to be significant but there could be indirect radiative effects through their role as CCN or ice nuclei (Bauer et al., 2003; Ariya et al., 2004; Christner et al., 2008).

2.3 Wildfires

2.3.1 The impact of wildfires on aerosol and climate

Particles emitted from biomass wildfires are an important constituent of the atmospheric aerosol. They are particularly important for climate because of their light-absorbing properties, which can affect absorption of radiation in the atmosphere and at the surface when the particles are deposited on snow and ice. Penner et al. (2001) reported global emissions of organic matter of 45–80 Tg a^{-1} and of black carbon (BC) of 5–9 Tg a^{-1} for biomass burning (including biofuels). More recent inventories of large-scale (or open) burning rely on remote sensing estimates of fire counts (Generoso et al., 2003) or area burned (Hoelzemann et al., 2004; van der Werf et al., 2004). The range of estimates for annual emissions of particulate organic matter from wildfires is 20 to 35 Tg a^{-1} (see Fig. D1 in Dentener et al., 2006).

Biomass burning aerosols scatter and absorb solar radiation and there is no agreement on the sign of their radiative forcing at the global scale (Forster et al., 2007), with some models predicting a positive radiative forcing and others a slightly negative radiative forcing (range -0.05 to 0.22 W m^{-2}). It is conceivable that aerosols from wildfires contribute to either warm or cool the climate depending on which season or region or from which ecosystems they are emitted.

The climate impact of wildfires is not limited to the direct effect on aerosols and aerosol precursors. Aerosols from wildfires may affect the amount of direct and diffuse solar radiation at the surface as well as precipitation through their role as cloud condensation nuclei (Andreae et al., 2004), which may then feedback on the ecosystem productivity and vegetation dynamics. In addition, deposition of light-absorbing particles on snow and ice reduces the albedo and may accelerate warming of Arctic regions (Quinn et al., 2008). Flanner et al. (2007) have shown that the efficacy of BC/snow forcing is more than three times greater than for CO_2 because of the strong effect of the BC on snow melting rates, which amplifies the snow-albedo feedback. Given the current uncertainties in the atmospheric and cryospheric effects of wildfire aerosol it would be speculative at this stage to try to close the climate feedback loop involving wildfires and aerosols (see Fig. 3).

2.3.2 Feedback processes involving wildfires

Before discussing climate feedbacks involving aerosols from wildfires, it is important to note that the breakdown of wildfires and related emissions between natural and man-made origin is still being discussed. It is generally argued that i) emissions due to deforestation in the tropics have scaled with population over the last two centuries; ii) forest burning emissions at high latitudes of the Northern Hemisphere are lower now than in the past because of forest management and fire suppression policies (e.g. Marlon et al., 2008); and iii) some ecosystems, such as savannas, are essentially fire-regulated systems and would burn regularly irrespective of anthropogenic pressures. However, these are generalizations, and it is likely that in many regions wildfires occur because of a mix of natural and anthropogenic factors (van der Werf et al., 2008). This should be kept in mind when discussing possible climate feedbacks involving wildfires as these feedbacks could be different in the absence or presence of anthropogenic factors.

2.3.3 Response of wildfires to climate change

Emissions from wildfires show considerable interannual variability of more than a factor 2 partly in response to climate (Generoso et al., 2003; van der Werf et al., 2004; Schultz et al., 2008). For example, fire emissions are larger during El Niño years because drought conditions associated with El Niño trigger an increase in fire activity. Decadal climate change may be changing the occurrence of forest fires in some regions (Gillett et al., 2004; Westerling et al., 2006), although the global mean ~50% increase in wildfire carbon emissions from the 1960s to 1990s is most likely due to increased deforestation (Schultz et al., 2008). If wildfires respond to climate variability on interannual to decadal timescales then it is likely that they will also respond to climate change on centennial timescales. Marlon et al. (2008) have shown variations on even longer timescales in relation to both climate change and anthropogenic activities.

2.3.3.1 Changes in the occurrence of fires

There are several possible effects of climate change on wildfires. Wildfires require fuel availability, readiness of the fuel to burn depending on the atmospheric and soil conditions, and an ignition source (be it natural or anthropogenic). Moisture is one of the most relevant parameters for fires and drought indices can be used as a proxy to infer fire risk in the future climate. For instance Burke et al. (2006) applied the Palmer Drought Severity Index (PSDI) in an SRES A2 simulation of the HadCM3 model and estimated a net global drying trend, although there were regions of strong wetting and drying. Cox et al. (2004) predicted a dieback of the Amazon rainforest in coupled climate-carbon cycle simulations for the 21st century because of a significant shift in precipitation patterns over this region. One mechanism for the Amazon dieback would be through increased fire disturbances in the context of sustained drought (Laurance and Williamson, 2001; Nepstad et al., 2004; Aragão et al., 2008).

There is no global study of the change in wildfires in the future but regional studies have been performed. For the United States Bachelet et al. (2003) predicted an increase in the total biomass burnt using two different Dynamic Global Vegetation Models (DGVM) fed by two different climate change scenarios. Westerling et al. (2006) attribute the observed increase in wildfire activity in the Western United States to an increase in spring and summer temperature, which are responsible for an earlier melting of mountain snowpacks. The same process could be at work under climate change in this region where both a temperature increase and a precipitation decrease are predicted by climate models (Running, 2006). Increased wildfire due to climate change has also been projected for Canada (Flannigan and Van Wagner, 1991; Wotton and Flannigan, 1993; Stocks et al., 1998; Flannigan et al., 2005), North America (Flannigan et al., 2000), Russia (Stocks et al., 1998), Western United States (Brown et al., 2004; Westerling and Bryant, 2008) and the European Mediterranean area (Morriondo et al., 2006). Flannigan et al. (2005) projected area burned to increase by 74–118% in Canada by the end of this century in a 3×CO_2 scenario. These estimates do not explicitly take into account any changes in vegetation, ignitions, and human activity (fire management and land use activities) that may also influence area burned. In the Amazon, forest fires are closely related to both rainfall and the amount of human disturbance: future development of the Amazon is projected to increase fire activity there by between 22% and 123% (Cardoso et al., 2003).

Changes in ignition sources are also likely to be important. Price and Rind (1994) projected that increased convection under a 2×CO_2 scenario leads to increased lightning and a 78% increase in area burned in the United States.

Amiro et al. (2009) made the first projections of future wildfire emissions accounting for both changes to area burned and wildfire severity. They find that changes to wildfire biomass consumption in the Canadian boreal forest region are driven mainly by changes in area burned with limited contribution from increased fire severity. They project that biomass consumption increases by 33% in a 2×CO_2 scenario and 95% in a 3×CO_2 scenario. However, future climate change may not always drive increases in wildfire. Decreased area burned has been projected for parts of Eastern Canada due to projected increases in precipitation (Bergeron and Flannigan, 1995; Flannigan et al., 2001).

2.3.3.2 Changes in wildfire aerosol emissions

There are very few studies of the change in aerosol emissions and concentrations due to the change in wildfires. Interannual variability in wildfires has been shown to be the dominant driver of observed variability in summertime OC concentrations in the Western United States (Spracklen et al., 2007a; Jaffe et al., 2008). Spracklen et al. (2009) investigated the impact of future climate change on wildfire activity and carbonaceous aerosol concentrations in the same region. They regressed observed area burned onto observed meteorological fields and fire indices from the Canadian Fire Weather Index system and applied the same regression model to meteorological fields calculated by a GCM. They showed that increases in temperature cause annual mean area burned in the Western United States to increase by 54% resulting in a 90% increase in wildfire carbonaceous emissions by the 2050s relative to present-day conditions. Summertime organic carbon (OC) aerosol concentrations over the Western United States were projected to increase by 40% and elemental carbon concentrations by 20% by 2050. This study suggested that wildfire was the dominant driver of future OC aerosol in the Western United States causing 75% of the projected change with changes to climate and SOA being responsible for the remainder.

It is worth noting that predicting future changes in area burned is not the same as predicting future changes in emissions. There might be a negative feedback whereby more fires eventually imply less biomass to burn. Some authors have argued that this may not be the case in boreal forests as these have sustained more fires in the past in the absence of fire suppression policies. However there would be other ecosystems where wildfires caused by climate change would trigger large initial emissions of carbon and aerosols followed by a shift in ecosystems through vegetation dynamics. Clearly more elaborate modeling of the interaction between vegetation dynamics, wildfires and emissions is needed.

2.3.3.3 Coupled impacts of wildfires on greenhouse gases, albedo and aerosol emissions

Randerson et al. (2006) reported measurements and analysis of a boreal forest fire, integrating the effects of greenhouse gases, aerosols, black carbon deposition on snow and sea ice, and postfire changes in local surface albedo. They estimate that the radiative forcing of mature boreal forest compared with forest over an 80-year fire cycle was a warming of 2.3±2.2 W m^{-2} (due to contributions from storage of carbon of −1.6±0.8 W m^{-2} and the lower albedo of the forest compared to the burned area of +4.2±2.0 W m^{-2}, the radiative forcing due to smoke emissions is small when averaged across the fire life cycle). This result would mean that future increases in boreal fire would not accelerate climate warming. However, Randerson et al. did not include the radiative effects of aerosol generated by forest BVOC emissions. Spracklen et al. (2008b) used a global aerosol model to predict that boreal forest terpene emissions contribute 50% of regional (north of 60° N) CCN concentrations and cause a local indirect aerosol radiative forcing of between −1.6 and −6.7 W m^{-2} compared to grassland (Sect. 2.1.1), which may compensate the net positive forcing estimated by Randerson et al. (2006). Thus at present the long term climate impact of forest fires is not certain.

2.4 Aerosol impacts on terrestrial systems

2.4.1 Impact of aerosol radiative effects on vegetation

In addition to their effects at the top-of-atmosphere aerosols decrease the amount of solar radiation reaching the surface because of their scattering and absorbing properties (Fig. 1). In particular aerosols affect the amount of solar radiation in the 0.45 to 0.75 µm wavelength range (known as photosynthetically available radiation, or PAR), which may have an impact on plant productivity and therefore on the carbon cycle. Aerosols also decrease the amount of direct solar radiation but increase the amount of diffuse radiation reaching the surface. Increase in diffuse PAR is known to enhance plant productivity (e.g., Gu et al., 2002). It has been suggested by several authors that the increase in diffuse radiation due to stratospheric aerosols following large volcanic eruptions may be responsible for an additional uptake of carbon dioxide by terrestrial ecosystems (Gu et al., 2003). This effect was modeled by Mercado et al. (2009) who concluded that not only transient increases in stratospheric aerosol loadings but also the centennial trend in tropospheric aerosols due to anthropogenic emissions are responsible for an enhancement of the terrestrial carbon sink. If the burden of natural aerosols were to change over vegetated areas then the same process would apply. This process can provide a negative climate feedback if natural aerosols increase in response to climate change (e.g. larger temperature leads to enhanced BVOC emissions, increased SOA burden, increased diffuse radiation, increased carbon uptake leading to less severe temperature change (Kulmala et al., 2004b).

2.4.2 Aerosols as a source of nutrients for vegetation

The wet and dry deposition of particulates to the Earth's surface has an important impact on terrestrial ecosystems. This deposition may contribute a significant nutrient source to some ecosystems (Fig. 1), but in polluted regions excessive deposition of acidic particles can damage vegetation and lead to acidification of soils. Deposition of particles on leaf surfaces can also alter leaf-surface wetness, altering ecosystem water and CO_2 uptake and the risk of pathogen attack (Cape, 2008).

Much previous research has focused on Amazonian ecosystems which are thought to rely heavily on external inputs of nutrients because their soils are highly weathered and nutrient limited. It has been proposed that the 50 Tg of Saharan dust deposited annually to the Amazon basin (Kaufman et al., 2005) is a significant source of nutrients (Swap et al., 1992). Boy and Wilcke (2008) demonstrated that base-metal deposition at an Amazonian forest site was dominated by long-range transport of Saharan dust. The transport and deposition of Saharan dust to the Amazon is linked to the ENSO cycle and is therefore susceptible to changes in climate (Boy and Wilcke, 2008). Biomass burning is also a significant source of carbon, sulphur, nitrogen and phosphorous-containing compounds to downwind ecosystems. Biomass burning in the Amazon basin can result in sulphur deposition rates similar to those observed in polluted central Europe (Fabian et al., 2005). While the deposition of phosphorous on a global scale is dominated by dust (82%) and primary biological aerosol particles (13%) (Mahowald et al., 2008), biomass burning may be the dominant source of phosphorous to the Amazon basin (Mahowald et al., 2005). Increased biomass burning in the Amazon due to land-use change is likely leading to increased deposition of phosphorous to undisturbed Amazonian forests, where it may be contributing to observed increases in carbon sequestration. Deposition to the oceans may also fertilize phosphorous-limited waters (Mills et al., 2004).

Anthropogenic emissions can cause significant enhancement of nitrogen and sulfur deposition to surrounding ecosystems. Enhanced nitrogen deposition from anthropogenic sources may be driving net carbon sequestration in boreal and temperate forest ecosystems (Mencuccini et al., 2007). A large aerosol nitrate loading can contribute substantially to total nitrogen deposition (Cape, 2008). Acid deposition from anthropogenic emissions has also resulted in extensive forest dieback (e.g. Driscoll et al., 2001) but the impact on the global carbon cycle has not been quantified. Sulphate deposition also impacts natural wetlands and rice paddies resulting in suppressed methane emissions (Gauci et al., 2008). The future deposition of particulates to ecosystems will depend on both changes to anthropogenic emissions and climate (e.g., Tagaris et al., 2008).

2.5 Status and development of terrestrial biogenic aerosol in earth system models

GCMs have been developed that include BVOC emission models coupled to atmospheric chemistry models. At present they account only for a limited range of the variables known to affect BVOC emissions and SOA formation (Sect. 2.1.2). Ultimately, Earth system models will couple sub-models representing vegetation dynamics and the carbon cycle, nutrient availability, mechanistic treatments of wildfire occurrence and the factors that control BVOC emissions, together with sophisticated atmospheric models of SOA formation and properties.

Expanding the set of drivers used to calculate BVOC emissions is necessary. In particular, including the effects of soil nutrient limitations, atmospheric carbon dioxide and ozone may greatly alter our understanding of how BVOC emissions and SOA will change in the future. Current BVOC emission algorithms assume that the response observed over a period of days to weeks is applicable over annual to decadal timescales (Guenther et al., 2006). There is a need to develop algorithms that are less empirical and more process-based. The impact of climate on vegetation type and distribution is also likely to be important and is only now being included in simulations of aerosol (Tsigaridis and Kanakidou, 2007).

The poorly constrained SOA budget represents a major limitation in accurately quantifying changes due to climate. Current models may underpredict atmospheric SOA by up to an order of magnitude (Volkamer et al., 2006) with unknown consequences when these models are used for chemistry-climate predictions. Previous studies have focused on the SOA production from monoterpenes, isoprene and anthropogenic organics. The emissions of other SOA precursors such as sesquiterpenes may be very strongly temperature dependent (Duhl et al., 2008) and so their contribution to SOA may be greater in a warmer climate.

Previous studies have been limited to an assessment of the change to SOA mass budgets. While new global aerosol microphysics models can predict aerosol size distribution, these models have yet to be applied to changing BVOC emissions and SOA with a changing climate. This is an important step that is required before an accurate assessment of future impact on the aerosol indirect effect can be made.

Current Earth System models include interactive aerosols and an interactive carbon cycle with vegetation dynamics. However disturbances to the vegetation caused by pests and wildfires are not represented explicitly. There is ongoing work to estimate fire index, fire risk and fire emissions as diagnostics in climate models. Some fire models have been coupled to dynamic vegetation models (Bachelet et al., 2003; Arora and

Boer, 2005) but not yet interactively as part of a climate model where the potential for real or spurious feedbacks is greater. However the next generation of Earth System models is expected to explicitly couple fire models with dynamics vegetation models (e.g. Spessa et al., 2008). There is also a need for models to couple the biophysical effects of forests (albedo, evapotranspiration, roughness length, etc.), the biogeochemical effects (CO_2 sequestration, etc.) with the capacity of forests to produce aerosol (Sect. 2.1.1).

3 Marine aerosol

This section considers the response to climate change of aerosol produced directly or indirectly by marine regions. A wide range of responses have been studied based on observations, models or both, and several processes have been included in coupled ocean-atmosphere models to quantify the feedback on climate. The most extensively studied oceanic response to climate is that of dimethylsulphide (DMS or CH_3SCH_3) produced by phytoplankton, which is a major source of aerosol (Shaw, 1983; Charlson et al., 1987). Recent observations have led to an improved understanding of emissions of other primary and secondary aerosol components, which are also driven by biological activity, and suggest that a wider range of climate-emission feedbacks need to be considered.

3.1 Plankton, dimethylsulphide emissions and sulphate aerosol

3.1.1 The impact of DMS on atmospheric aerosol

DMS originates from the decomposition of dimethylsulphoniopropionate (DMSP) produced by marine organisms, particularly phytoplankton. It is found in varying concentration in seawater and is emitted into the atmosphere. Products of the gas phase oxidation of DMS contribute substantially to the mass of atmospheric aerosol. The sequence of reactions responsible for oxidation of DMS is given by Koga and Tanaka (1999) in their Table 1. The sequence begins with the reactions of DMS with OH and NO_3, also involves O_3, HO_2 and H_2O_2, and results in the eventual production of SO_2, methanesulphonic acid and gas-phase sulphuric acid (H_2SO_4) which can then condense onto aerosol particles (Pham et al., 1995) or nucleate to form new sulphuric acid particles (Kulmala et al., 1998). Chemical transport model studies suggest that ∼42% of global atmospheric sulphate aerosol is derived from DMS although it accounts for only 23% of sulphur emissions (the difference being due to the longer lifetime of DMS versus anthropogenic SO_2) (Chin and Jacob, 1996). Chin and Jacob (1996) also estimated that DMS accounts for 20–80% of sulphate in surface air over the Northern Hemisphere oceans and over 80% in most of the Southern Hemisphere and in the upper troposphere.

The observed correlation of DMS, aerosol sulphate, CCN and cloud properties on seasonal timescales (Ayers and Gras, 1991; Ayers et al., 1997; Boers et al., 1994) suggests that phytoplankton dynamics, DMS emissions and aerosol and cloud microphysics could be linked. Nevertheless, despite more than 20 years of research since the original proposal of a climate feedback involving DMS (Shaw, 1983; Charlson et al., 1987) and over 1500 related articles, the magnitude and even direction of the feedback remains uncertain (Ayers and Cainey, 2007).

3.1.2 Feedback processes involving DMS and aerosol

The climate feedback involving DMS emissions and aerosol has become known as the CLAW hypothesis after the authors of Charlson et al. (1987). At the most basic level, it is hypothesized that the emitters of DMS, phytoplankton, will respond to climate change and thereby affect the abundance of sulphate aerosol, and hence cloud properties and climate. Although CLAW is often cited as an example of a negative climate feedback, positive feedback scenarios have also been proposed. In addition, there are several proposed scenarios in which the direction of feedback is uncertain (see Fig. 3). It is important to recognize that different responses may operate together or in different regions or seasons (Boyd, 2002), so a single global mean feedback may not be a useful construct.

Several potential feedbacks are driven by changes in temperature and wind speed. A feedback that is likely to be negative is the reduction of the mixed layer depth (shoaling) due to atmospheric warming, causing phytoplankton to reside closer to the water surface, resulting in an increase in the received solar radiation dose (SRD) and increased DMS emission (Vallina and Simo, 2007a, b; Vallina et al., 2007a, b), although Larsen et al. (2008) has cautioned that changes in the different wavelength components of SRD could have different and perhaps compensating effects on DMS emissions. The mechanism is consistent with laboratory experiments showing that plankton DMSP production increases in response to solar radiation (Sunda et al., 2002). Although SRD and DMS are strongly correlated in the global ocean (Vallina et al., 2007a; Vallina and Simo, 2007b) the impact of climate change in a model seems to be small (Vallina et al., 2007b). Changes in ocean temperature and stratification can also result in shifts in ecosystem flora and structure (Boyd and Doney, 2002; Bopp et al., 2003). Increased wind speeds, and hence surface wind stress, result in deepening of the wind mixed-layer, which affects nutrient availability in an uncertain way. It also causes increased DMS air-sea exchange (Gabric et al., 1998), which alone would be a negative feedback, but also increases the sea spray flux (Sect. 3.2.1), which will impact atmospheric DMS oxidation in an as yet unquantified way (von Glasow, 2007).

Global satellite observations have been used to study the link between ocean productivity and climate, although the main indicator of productivity (chlorophyll) may not be a good indicator of changes in DMS itself. Gregg and Conkright (2002) showed that global mean ocean chlorophyll decreased by about 6% from the 1980s to 2000 with substantial regional variability most likely attributable to natural variability in wind stress, ocean warming and cooling. Behrenfeld et al. (2006) have shown using satellite observations of ocean color that ocean productivity has varied substantially over the past decade, most likely driven by climate induced changes in ocean stratification and nutrient supply. They suggest that future warming could increase ocean stratification and thereby reduce the supply of nutrients from deeper waters, limiting plankton growth, which would constitute a positive feedback. On a smaller scale, observations suggest a positive correlation between anomalies in sea water temperature and DMS in the Southern Indian Ocean, amounting to a 50% increase in DMS for a 1 K increase in temperature (Sciare et al., 2000b), although the cause was not established.

Several other drivers of changes in DMS production and emission have been proposed.

1. Retreat of Arctic summer sea ice exposes more water to solar radiation, which may lead to enhanced DMS production and emission (Gabric et al., 2005). In contrast, in the Southern Hemisphere Curran et al. (2003) showed that methanesulphonic acid (an oxidation product of DMS) has decreased by about 20% in Antarctic ice cores since the 1950s. This decrease is consistent with decreases in sea ice extent and the dominant source of DMS being from sea-ice algae. Increased aerosol optical depth during spring sea ice melt may provide corroborating evidence (Gabric et al., 2005) but effects of changes in temperature and atmospheric dynamics on DMS chemistry (Sect. 3.1.3) were not investigated. Thus, changes in sea ice may have opposite effects on DMS emissions in the Northern and Southern Hemispheres.

2. Increasing CO_2 raises ocean acidity (Caldeira and Wickett, 2003) resulting in physiological changes to plankton (Wingenter et al., 2007) and changes in DMS emission. However, the response of plankton communities to increased CO_2 and warming is complex and responses are not consistent across experiments (Vogt et al., 2008).

3. Changes in atmospheric dust due to changes in wind or vegetation influences iron fertilization and plankton growth (Jickells et al., 2005) – Sect. 5.2.2. Iron fertilization experiments have produced conflicting results (Boyd et al., 2007) and are difficult to extrapolate to regional and seasonal scales, although global analyses do suggest a link (Cropp et al., 2005). Significantly different responses of nat-

ural blooms to long term changes have also been observed (Blain et al., 2007). Changes in global dust deposition predicted by models are also highly variable, as are estimates of future changes in dust emissions, which may be positive or negative (Sect. 5.2).

4. Changes to atmospheric convection and lofting of DMS to the free troposphere could enhance particle nucleation and hence CCN (Shaw et al., 1998). In the GCM simulations of Kloster et al. (2007) global warming caused an overall decrease in the temperature lapse rate in the mid and low latitudes and a reduced lifetime of DMS, although the impact on CCN was not quantified.

5. Levels of surface ultraviolet radiation may affect phytoplankton productivity and the rate of seawater DMS destruction (Larsen, 2005). In a changing climate ultraviolet radiation at the surface may respond to changes in the aerosol loading, cloudiness and/or stratospheric ozone. Some models predict an increase in the stratospheric ozone layer (super-recovery) in the mid-latitudes because of a cooling (which reduces the destruction rate of ozone) and an increase in the Brewer-Dobson circulation (which increases the meridional transport of ozone from the tropics to the mid-latitudes) (Bodeker et al., 2007). Such an effect, while plausible, has not been quantified so far.

Two factors combine to limit accurate quantification of the direction and magnitude of DMS and sulphate aerosol in response to climate change: our understanding of the components of the system (e.g., factors affecting phytoplankton ecosystems, air-sea gas exchange, chemistry of DMS, aerosol microphysics, etc.) and the uncertainties in climate drivers (response to climate change of the mixed layer depth, dust deposition, wind speed, etc.). Below, we review the status of ocean DMS production models, atmospheric chemistry and aerosol models, and coupled ocean-atmosphere climate model studies of the CLAW mechanism.

3.1.3 Models of DMS-aerosol feedback

3.1.3.1 Plankton and DMS production models

The basis of CLAW as a climate regulator is the change in DMS production in the ocean. As with all components of the CLAW mechanism, it has become clear that DMS production is more complex than previously thought (Archer, 2007). In particular, grazers, viruses and bacteria all play a role in converting DMSP to DMS, but the budget of the various processes remains poorly quantified.

Prediction of DMS requires a model of the ecosystem regulation of DMS production (Stefels et al., 2007) and an ecosystem dynamics model to predict the response of the producers to climate change (e.g., Le Quéré et al., 2005). Models have grown considerably in complexity since the first DMS production models of Gabric et al. (1993) and Lawrence et al. (1993). They are able to reproduce observed chlorophyll and DMS dynamics where they are in phase at high latitudes (>40°) but have failed to capture behavior at low latitudes where high summer DMS concentrations are associated with low chlorophyll – the so-called "summer paradox" (Simo et al., 1999). Recent models incorporating solar radiation dose-dependent DMSP production do much better (Vallina et al., 2008).

There are two ways in which climate change can alter DMS production: through floristic shifts and changes in bulk global ocean productivity. Changes in phytoplankton composition are potentially very important for DMS because of the substantial variation in DMSP cell content. However, a definitive understanding of DMSP physiological function is still lacking, which limits the development of mechanistic models and makes it difficult to define an appropriate level of model complexity to capture the response to climate change (Archer, 2007). Furthermore, models have not been able to capture interannual changes in primary productivity (e.g., Behrenfeld et al., 2006).

3.1.3.2 Atmospheric chemistry and aerosol models

Prediction of the response of climate-relevant aerosol particles to perturbed DMS emissions requires a global model that couples atmospheric chemistry and aerosol microphysics. The chemistry of DMS is more complex than originally appreciated when the CLAW hypothesis was proposed, and the details appear to be important for quantifying the production of aerosol (von Glasow, 2007; Barnes et al., 2006; Lucas and Prinn, 2005). Models need to take account of several oxidants (OH, NO_3, BrO, O_3) as well as heterogeneous chemistry on sea spray particles (Boucher et al., 2003; von Glasow and Crutzen, 2004). There are large differences in predictions among gas phase chemistry schemes of different complexity, which may be as much as an order of magnitude for H_2SO_4 in some regions (Lucas and Prinn, 2005). Hence the ability of simplified reaction schemes in climate models to capture regional and seasonal variability in aerosol production from DMS and the response to climate change is at present questionable.

It is not so much the bulk quantity of DMS oxidized that matters for the CLAW hypothesis but the ability of oxidation products to increase the CCN population. However, the production of CCN from DMS-derived SO_2 and H_2SO_4 vapor is complex and there is no simple globally applicable relationship between them. The size of particles that can act as CCN depends on the in-cloud supersaturation, which can vary between $<0.1\%$ and $>1\%$. At 0.1% supersaturation particles of $\sim$90 nm dry diameter can serve as CCN, but much smaller particles can activate at 1% supersaturation. CCN concentrations are therefore influenced by DMS in three ways: by (i) condensation of DMS-derived H_2SO_4 onto small nucleation and Aitken mode particles, growing them to CCN sizes (these particles could come from sea spray or other sources); (ii) nucleation of new H_2SO_4-H_2O particles that eventually grow to CCN sizes; or nucleation of new particles via I_xO_y-H_2SO_4 mechanisms (Vuollekoski et al., 2009) and (iii) formation of aerosol sulphate through oxidation of SO_2 in cloud drops. Current global aerosol microphysics models (e.g., Spracklen et al., 2005; Adams and Seinfeld, 2002; Stier et al., 2005) include the microphysical processes needed to capture such processes explicitly, although simulations of remote marine aerosol show large (Spracklen et al., 2007b) and consistent (Trivitayanurak et al., 2008) regional disagreement with observations.

Based on model studies and observations a consistent picture of CCN production from DMS has emerged. An important source of DMS-derived CCN is from sulphuric acid-water particles nucleated in the cold free troposphere (FT) and subsequently entrained into the marine boundary layer (MBL) (Raes et al., 1995). This process has now been studied in detail using quite complex aerosol microphysical models in one (Caffrey et al., 2006) and three dimensions (Korhonen et al., 2008). Korhonen et al. (2008) were able to reproduce the observed CCN seasonal cycle at Cape Grim (Ayers et al., 1997) and estimated that, averaged over the Southern Ocean, the FT accounts for about 90% of the MBL CCN produced by DMS. However, their model results suggest that it is not possible to define a single value for the sensitivity of CCN to changes in atmospheric DMS (dCCN/dDMS) because of the non-linear processes linking H_2SO_4 production and particle nucleation. They estimated a peak (summer) zonal mean contribution of DMS to MBL CCN in the Southern Ocean of between 18 and 46% depending on latitude, which is much less than the 80% estimated from satellite observations (Vallina et al., 2006). Other work has shown how localised DMS emissions can impact CCN over large regions downwind (Woodhouse et al., 2008). This patchy response shows that dCCN/dDMS is a non-local, regionally variable factor and makes an experimental confirmation of modeled CCN response difficult.

3.1.3.3 Coupled model simulations of climate-DMS feedbacks

Coupled ocean-atmosphere GCMs have been used to study various combinations of the processes in Fig. 4 (Bopp et al., 2003b; Gabric et al., 2003, 2004, 2005; Vallina et al., 2007a; Kloster et al., 2007; Gunson et al., 2006), but no single study has yet combined them in a complete Earth System model. Model results are contradictory, although model intercomparison is difficult because of the different assumptions and experimental designs used. The uncertainty of the model predictions is likely to be large for the climate, ecosystem and chemistry (Lucas and Prinn, 2006) sub-models.

Bopp et al. (2003b) used an atmosphere–ocean GCM coupled to a marine biogeochemical scheme to study the response of DMS to a doubling of pre-industrial CO_2. The model predicted a 2% increase in the global DMS flux to the atmosphere but with large spatial heterogeneities (from −15% to +30% for the zonal mean). Bopp et al. (2004) prescribed these changes in seawater DMS in an atmospheric GCM and estimated a change in cloud radiative forcing of $-0.05\,\mathrm{W\,m^{-2}}$. However, large regional changes were also predicted, such as a perturbation of up to $-1.5\,\mathrm{W\,m^{-2}}$ in summer between 40° S and 50° S, which could impact the regional climate. A notable feature of their results is a strong response of DMS production to changes in the phytoplankton species composition, most apparent in the sub-Antarctic Pacific. The shift from diatoms to other species more efficient in producing DMS results in a net increase in DMS.

Gunson et al. (2006) used a coupled ocean-atmosphere GCM with an ocean ecosystem model to predict the climate response to changes in DMS. The ecosystem model was used to predict DMS based on an empirical relation with chlorophyll, shortwave radiation, and a nutrient limitation factor (Anderson et al., 2001). Halving DMS emissions caused a net global mean cloud radiative forcing of $+3\,\mathrm{W\,m^{-2}}$ and an increase in surface mean temperature of 1.6 K. Climate warming caused a feedback on the DMS flux, which increased by 2.1%. The negative feedback factor was calculated to be 0.06 which is a small negative feedback in support of the CLAW hypothesis.

Vallina et al. (2007a) used an ocean GCM forced by offline meteorology in a control and a $1.46\times CO_2$ atmosphere for a 56-year climate warming scenario. Two diagnostic equations defined the DMS response to either mixed layer depth and chlorophyll or solar radiation dose. Their model predicted a very small (1.2% global mean) increase in DMS flux in response to a net decrease in mixed layer depth. Regional differences were also small (6.3% at 95th percentile) with maximum changes of 10–15% in the equatorial Pacific.

Gabric et al. (2001) used a coupled ocean-atmosphere GCM and a mechanistic DMS model driven by sea-surface temperature, wind speed, cloud cover and mixed layer depth. They estimated a 5% increase in DMS flux from 1960 to 2080 (CO_2 tripling) for a region of the Southern Ocean. The main driver was a decrease in the mixed layer depth. They equated this change in DMS to a $-0.3\,\mathrm{W\,m^{-2}}$ radiative forcing (direct and cloud) and a negative feedback factor of 0.04, similar to Gunson et al. (2006).

Kloster et al. (2007) used a coupled ocean-atmosphere GCM coupled to a plankton dynamics model. Their model accounted for changes in ocean dynamics and mixing in response to climate change. Between 1861–1890 and 2061–2090 they calculated a global mean 10% decrease in DMS flux to the atmosphere, resulting in a 3% decrease in DMS concentration (owing to changes in atmospheric lifetime of DMS). The response of DMS was globally heterogeneous. In particular, decreases in summer seawater DMS of up to 40% occurred in the Southern Ocean caused by an increase in the summer mixed layer depth and mixing of phytoplankton to depths where photosynthesis is less favorable. This response, which is opposite to previous model results, was driven by changes in wind patterns caused by the poleward shift of the storm track. At low and mid-latitudes DMS sea surface concentrations decreased by ∼10–20% due to increased stratification, while at high latitudes DMS concentrations increased due to retreat of sea ice. They find changes in phytoplankton speciation to have only a small effect on DMS production, unlike Bopp et al. (2003b).

3.1.4 Impacts of changes in oxidising capacity on sulphate aerosols

The direct effect of climate change on natural sulphate aerosol is through modified DMS emissions. A further effect that cannot be ignored is the impact of changes in the oxidants responsible for conversion of DMS to sulphate. The dominant gas-phase oxidant of SO_2 is OH, while the dominant aqueous-phase oxidants are H_2O_2 and O_3 (Calvert and Stockwell, 1984; Seinfeld and Pandis, 1998; Finlayson-Pitts and Pitts, 2000; Warneck, 2000). These oxidants are formed from emitted precursors via sequences of gas-phase reactions (described by, e.g., Seinfeld and Pandis, 1998; Finlayson-Pitts and Pitts, 2000; Warneck, 2000).

Gas- and aqueous-phase reactions, and aqueous equilibria, are temperature-dependent. Aqueous-phase reactions depend additionally on cloud fraction and cloud liquid water content. Rates of reactions involving water vapor, or reactions catalysed by water vapor (such as the reaction of HO_2 with itself to form hydrogen peroxide) will increase with increased humidity. The rates of photolysis reactions may be inversely correlated with cloud cover (Koch et al., 2003). In addition, stratosphere-troposphere exchange of ozone may increase in a warmer climate (Butchart and Scaife, 2001; Collins et al., 2003), which could lead to increased tropospheric concentrations of ozone. Such climate-chemistry-aerosol interactions mean that concentrations of oxidants, and therefore sulphate aerosol burdens and radiative forcing, are likely to be affected by future climate changes as well as by changes in emissions (see, e.g., Johnson et al., 1999; Liao et al., 2003; Zeng and Pyle, 2003; Berglen et al., 2004; Pham et al., 2005; Unger et al., 2006; Rae et al., 2007).

Model studies of these effects have focused on anthropogenic sulphur sources and continental regions. Pham et al. (2005) gave the results of a study of the sensitivity of sulphate aerosol burdens to changes in oxidant concentrations. They ran experiments using the Laboratoire de Météorologie Dynamique GCM with 2100 SRES A2 sulphur emissions, and with oxidant concentrations appropriate for (i) the year 2000 and (ii) the year 2100 in SRES scenario A2. They found that the global burden of SO_2 was 4–5% less with (ii) than with (i), while that of sulphate decreased by less than 1%. They attributed this small decrease in sulphate to a more efficient oxidation, which results in sulphate being near the surface where it is scavenged, and therefore deposited, more efficiently.

Unger et al. (2006) ran simulations with the Goddard Institute for Space Sciences (GISS) ModelE GCM, with fully interactive chemistry and aerosols, for several model scenarios. In a 2030 simulation the rate of gas-phase SO_2 oxidation increased by 7% when 2030 oxidant precursors were used compared to 1995 oxidants. Rae et al. (2007) used the UK Met Office HadGEM1 climate model to study the effects on sulphate aerosol of changing the input oxidant concentrations from present-day values to values consistent with late-21st century predictions of emissions and climate. They compared this with the effects of changing the climate boundary conditions (sea surface temperature and sea ice fraction) and greenhouse gas concentrations in the model to predicted late-21st century values, while holding oxidant concentrations at present-day levels. They found that changing the oxidants alone caused the global total sulphate burden to decrease by about 3%, causing a global mean radiative forcing of $0.014\,\mathrm{W\,m^{-2}}$. Changing the climate alone led to an increase in global sulphate burden of about 9% (corresponding to a radiative forcing of $-0.047\,\mathrm{W\,m^{-2}}$). When both were changed simultaneously, there was an increase of about 7% in global total sulphate (corresponding to a radiative forcing of $-0.036\,\mathrm{W\,m^{-2}}$).

The studies described above show the importance of feedbacks in influencing sulphur-cycle oxidation reactions, and hence the influence they have on sulphate aerosol loading and consequently on radiative forcing. Traditionally climate models have used prescribed time-mean oxidant concentrations (e.g. Jones et al., 2001; Feichter et al., 2004; Pham et al., 2005). Now, however, oxidant concentrations are being calculated online in climate models (e.g. Liao et al., 2003; Unger et al., 2006; Rae, 2008). It is likely that, in the future, online oxidants will be used increasingly in Earth System models, allowing proper representation of climate-chemistry-aerosol feedbacks.

3.2 Marine primary aerosol

Marine primary aerosol includes both inorganic and organic components. Both emissions are susceptible to changes due to changes in wind speed, while the organic component may also respond to changes in marine biota, much like DMS.

3.2.1 Sea salt particles

It has been known for a long time that sea spray particles are numerous enough to constitute an important source of CCN at cloud base and that their production rate is wind speed dependent (O'Dowd and Smith, 1993; see Lewis and Schwartz, 2004, for a review). More recent studies have shown that sea spray emissions extend down to a few nanometres (Martensson et al., 2003; Geever et al., 2005; Clarke et al., 2006) and that emission rates depend on both wind speed and sea surface temperature. Microphysical model studies incorporating these latest emission schemes (Caffrey et al., 2006; Pierce and Adams, 2006; Korhonen et al., 2008) suggest that sea spray makes a substantial but highly variable (typically 10% to 60%) contribution to monthly mean regional MBL CCN concentrations depending on season and supersaturation, a far higher contribution than assumed originally by Shaw (1983).

Given the substantial contribution of sea spray to CCN, the dependence of production rate on temperature and wind speed, and the long term changes in wind speed observed at southern high latitudes (Le Quéré et al., 2007) and predicted by models, the potential exists for regional or global changes in CCN in response to climate change. Penner et al. (2001) found that sea salt emissions may increase in a warmer climate because of the increase in wind speed. However this result was model-dependent, with some climate models predicting an increase in wind speed in 2100 and other models predicting a decrease. Moreover sea-salt emissions were not calculated interactively in these models, and Penner et al. (2001) had to rely on time-averaged wind speeds for their calculations. This may alter the frequency of high wind speeds which are critical to predict the sea-salt emission flux. Jones et al. (2007) examined the change in sea-salt aerosols in response to 2×CO_2 forcing in the HadGEM1 climate model coupled to a mixed-layer ocean and sea ice model. In their simulation the wind speed decreases over most of the tropical and mid-latitude oceans but increases at high latitudes. This increase appears to be related to the reduction in sea ice and the decreased roughness length over the open ocean as compared to sea ice. This causes a sizable increase in sea-salt concentration in these areas (75% and 51% increases in sea-salt burdens poleward 60° N and 60° S, respectively) in response to a doubling of CO_2. Given that sea salt particles comprise a significant fraction of CCN concentrations in these regions (e.g., Spracklen et al., 2005), such large changes are likely to cause a large forcing through changes in cloud drop number.

3.2.2 Marine organic primary and secondary aerosol

It has been known for some time that the ocean has a layer of surface-active organic material that can be emitted into the atmosphere (Blanchard, 1964) and substantial amounts of particulate organic carbon (OC) have been observed at several marine sites (Novakov et al., 1997; Putaud et al., 2000; Cavalli et al., 2004; O'Dowd et al., 2004; Leck and Bigg, 2005a, b; Pio et al., 2007; Bigg and Leck, 2007; Spracklen et al., 2008c).

The correlation of marine aerosol OC with chlorophyll (O'Dowd et al., 2004, 2008; Spracklen et al., 2008c) and the strong seasonal cycle in OC abundance suggests a biologically driven production mechanism of potential importance to climate. It has been proposed that marine OC has the potential to modify or augment the CLAW mechanism via DMS oxidation (Leck and Bigg, 2007). The marine OC source is highly uncertain with emission estimates ranging from 2 to more than 75 $\mathrm{Tg\,a^{-1}}$ (Roelofs, 2008; Spracklen et al., 2008c; Langmann et al., 2008). Spracklen et al. (2008c) used a combination of new OC observations in remote regions and inverse modeling simulations to suggest a global marine OC source of $\sim 8\,\mathrm{Tg\,a^{-1}}$, comparable to the global anthropogenic OC source of 5–30 $\mathrm{Tg\,a^{-1}}$ and the 22.6 $\mathrm{Tg\,a^{-1}}$ of sulphur emitted as DMS. Based on these emissions and the correlation of aerosol OC and chlorophyll it seems likely that marine OC will impact climate, but the effect depends on the relative contribution the OC particles make to CCN alongside sea salt and DMS-derived aerosol.

Model simulations to quantify the contribution to CCN have not been feasible yet because of our limited understanding of the sources and physical and chemical properties of the marine OC, which is likely to have various and complex biological sources. For example, some studies have detected degradation products of bacteria and viruses in the aerosol (Leck and Bigg, 2005a, b). Other studies suggest that much of the organic material is water-insoluble when emitted from the ocean (Ceburnis et al., 2008), but that SOA of marine origin can also contribute to particle growth (Vaattavorra et al., 2006). Meskhidze and Nenes (2006) suggested that marine isoprene emissions could explain observed correlations between chlorophyll and cloud drop sizes observed by satellite, but a follow-up study (Miller and Yuter, 2008) casts doubt on the correlations. Furthermore, the abundance of atmospheric marine OC is much higher than can be explained in terms of isoprene emissions alone (Spracklen et al., 2008c; Palmer and Shaw, 2005; Arnold et al., 2009), suggesting that primary emissions may dominate (Leck and Bigg, 2007). Observations in the north east Atlantic suggest that approximately 70% of water-insoluble organic carbon exists at submicron sizes (Cavalli et al., 2004). Such enrichment in small particles, unlike sea salt, suggests a high potential efficiency of the OC to act as CCN.

Another open question, as with DMS, is how biological processes control the organic aerosol emissions and the extent to which wind speed or other physical processes play a role (Nilsson et al., 2007; O'Dowd et al., 2008). However, with present knowledge on the magnitude of emission and the correlation with chlorophyll, marine organic aerosol needs to be considered as a potentially important component of the climate-ocean-aerosol feedback alongside DMS.

3.3 Other marine emissions

There are many other biologically driven species emitted from the ocean that may also affect atmospheric chemistry or climate, such as organohalogens (Carpenter et al., 2003), ammonia (Schlesinger and Hartley, 1992), isoprene (Bonsang et al., 1992; Palmer and Shaw, 2005; Arnold et al., 2009), monoterpenes (Yassaa et al., 2008) and other non-methane hydrocarbons. The influence of these emissions on climate has not been quantified, but their potential to influence oxidants and aerosol processes in remote marine regions is plausible and needs to be evaluated. For example, oceanic organohalogens are strong sources of reactive halogen species in the atmosphere, such as the halogen oxide radicals IO and BrO, which influence tropospheric oxidation processes, DMS chemistry and particle formation in some environments (O'Dowd et al., 2002). These species may strongly control photochemistry of remote ocean regions

(Read et al., 2008) and impact DMS chemistry and aerosol production.

3.4 Status and development of marine aerosol in earth system models

Coupled ocean-atmosphere GCMs have been used to study the impact of climate change on marine DMS and aerosol emissions. These studies point to a weak positive or negative global response of sulphate aerosol to climate change due to increased CO_2. Most studies suggest a negative feedback (more aerosol in a warmer climate) driven by multiple factors outlined in Sect. 3.1.2. A positive feedback was predicted by Kloster et al. (2007) due to physical changes to the ocean. Some studies predict large regional changes in aerosol and forcing of as much as $-1.5\,\mathrm{W\,m^{-2}}$, which would be important for regional climate change.

The next stage in model development will involve the coupling of more advanced ocean ecosystem models (e.g., Le Quéré et al., 2005), ocean-atmosphere physical models, and the available global aerosol microphysics models. However, there is considerable scope for evaluation of the sub-models against observations. In addition, large differences in GCM predictions of the impact of climate change on DMS emissions (Sect. 3.1.3.3) appear to be due in large part to differences in the physical climate of the atmosphere and ocean, which drive DMS production and emission. Accurate predictions of changes in sea ice and the effects of changing temperature and wind speed on the mixed layer depth and stratification are essential.

There are several major observational and modeling challenges to understand the environmental factors that govern phytoplankton processes (Boyd, 2002) and DMS emission. A major challenge for the global models is to reproduce the interannual variability of surface chlorophyll (Le Quéré et al., 2005; Behrenfeld et al., 2006), an indicator of ocean productivity. Present global biogeochemical models do not achieve such agreement. Sophisticated plankton dynamics models have been developed (e.g., Le Queré et al., 2005). The predictive skill of these models should be demonstrated in a GCM against observed regional and seasonal variations in plankton and DMS, including the “summer DMS paradox”. Such evaluation would give increased confidence in predictions of DMS changes this century.

The impact of iron deposited in dust on DMS production is poorly understood. While an attempt can be made to simulate it in Earth system models, fundamental work is needed on iron speciation in dust, transformation in the atmosphere, and availability to biota (Sect. 5.2.2).

In the atmosphere, good progress has been made in understanding the key uncertainties in DMS chemistry (Lucas and Prinn, 2006; Barnes et al., 2006). However, despite an appreciation of the substantial differences between complex and simple models the more complex models remain too computationally demanding for Earth System models. The explanatory power of complex chemical schemes, particularly for the halogen components and mixed-phase processes, needs to be assessed if simulations are to be feasible on centennial timescales. Many global models now include microphysical schemes sufficient to simulate the response of CCN to changes in DMS (e.g., Korhonen et al., 2008; Kloster et al., 2008), and these models are suitable for inclusion in Earth system models. A major challenge is to evaluate these aerosol models against observations of DMS and CCN on seasonal and interannual timescales in different regions. However, a major limitation is the lack of observed DMS emissions (which need to cover very wide regions around the aerosol observations (Woodhouse et al., 2008)), as well as the lack of long-term reliable datasets of CCN. Satellite observations of CCN, cloud drop size and chlorophyll (Meskhidze and Nenes, 2006; Vallina et al., 2007b) are potentially valuable in aerosol model assessment, but need to be carefully evaluated against in situ observations if they are to be used quantitatively.

Our understanding of marine aerosol and the impact on climate has some large and fundamental gaps. The substantial organic component of sub-micron sea spray aerosol has been a significant discovery in recent years (Sect. 3.2.2) but an understanding of the production mechanisms, spatial and temporal variability and CCN properties prohibits inclusion in global models at present. Air-sea gas and aerosol exchange is very uncertain (>factor 2) and only takes account of wind speed, neglecting currently poorly understood factors such as surface surfactants, fetch and temperature.

Given the very large uncertainties in air-sea exchange rates, the impact of long term changes in these factors should not be ignored. Also, given the current resolution of climate models, the sub-grid scale variability in wind speed may be important to simulate air-sea exchange and parametrisations of gustiness need to be introduced in the models (Morcrette et al., 2008).

4 Stratospheric and volcanic aerosols

There is a thin layer of aerosols in the stratosphere, between the tropopause and 30 km, known as the Junge layer. For a long time stratospheric aerosols were thought of being mostly composed of sulphuric acid of submicron size. However, it is realized now that organic aerosols may play a role as well, at least in the lowest 2 km of the stratosphere (Murphy et al., 1998). The Junge layer is partly sustained by natural emissions of carbonyl sulphide (OCS) which is produced through biogenic processes. OCS is relatively stable in the troposphere and can reach the stratosphere where it is photochemically broken down, eventually resulting in the formation of sulphuric acid droplets. Sulphur dioxide is also transported through the tropopause but efficient oxidation and scavenging in the troposphere reduces its concentration. Models show that OCS is the main contributor to stratospheric aerosol above 25 km and SO_2 plays a larger role below (Weisenstein et al., 2006). Despite its short lifetime, there could be a small contribution from DMS to the Junge layer as DMS is not scavenged; however the contribution is likely only a few percent. Finally some VOC are long-lived and can be oxidized in the stratosphere into semivolatile species that can condense into SOA.

In background conditions (i.e. in the absence of large volcanic eruptions), the Junge layer is so thin (aerosol optical depth smaller than 0.005 at 550 nm) that it is hard to imagine that background stratospheric aerosols can play a significant role in climate feedbacks.

Explosive volcanic eruptions can inject SO_2 directly into the stratosphere where it is oxidised into sulphuric acid. However most volcanic plumes do not penetrate into the stratosphere and only a small number of eruptions have a significant impact on the Junge layer. The most recent examples are the 1982 El Chichón and the 1991 Mt Pinatubo eruptions. The stratospheric aerosol layer was perturbed for several years following these eruptions, with significant impacts on climate. In particular surface air temperatures cooled by up to 2 K over the Northern Hemisphere continents in the summer following the Mt Pinatubo eruption (Robock, 2002; Soden et al., 2002) and there is evidence that vegetation and the global carbon cycle have also responded to both the global cooling and the increase in diffuse solar radiation at the surface (Gu et al., 2003). By altering levels of surface ultraviolet radiation, significant perturbations of stratospheric aerosols can also impact aquatic and terrestrial ecosystems (Larsen, 2005; Caldwell et al., 2007; Häder et al., 2007).

Flood lava eruptions can also have a dramatic effect on the environment and affect the troposphere and stratosphere (Thordarson and Self, 2003). The 1783–1784 Laki flood lava eruption in Iceland emitted ∼122 Tg SO_2 into the atmosphere. The environmental impacts were widespread, including extreme pollution and crop damage, possible high latitude cooling of around −1.3 K that lasted for 2–3 years, and wider impacts on the African and Indian Monsoons (Oman et al., 2006). Eruptions of Laki magnitude have occurred in the recent past in Iceland and will occur again (Thordarson and Larsen, 2007). The impact on the biosphere through acid deposition and changes in direct and diffuse radiation (Sect. 2.4) are likely to be substantial but are poorly understood.

Although climate change is not thought to affect the intensity or the frequency of large volcanic eruption, it is conceivable that climate change will affect the fate of stratospheric aerosols after large volcanic eruptions, thus modulating the climate impact of stratospheric volcanic aerosols. The most likely way climate change can affect stratospheric aerosols is through changes in stratospheric transport and mixing. It has been shown that climate change will enhance the mass exchange between the troposphere and the stratosphere (Butchart and Scaife, 2001). In an intercomparison study Butchart et al. (2006) showed that all the models consistently predict an increase in the mass exchange rate in response to growing greenhouse gas concentrations, irrespective of whether or not the model includes interactive ozone chemistry. The mean trend is 11 kt s^{-1} a^{-1}, or about 2% per decade, but varies considerably between models. In all but one of the models the increase in mass exchange occurs throughout the year though, generally, the trend is larger during the boreal winter. Garcia and Randel (2008) showed that in their model the circulation strengthens as a result of increased wave driving in the subtropical lower stratosphere, which in turn occurs because of enhanced propagation and dissipation of waves in this region. Enhanced wave propagation is due to changes in tropospheric and lower-stratospheric zonal-mean winds, which become more westerly.

The increase in the Brewer-Dobson circulation means a shorter lifetime for stratospheric aerosols following large volcanic eruptions in the 21st century. This would shorten the cooling period and reduce the associated terrestrial carbon uptake experienced after such eruptions.

5 Dust aerosol

5.1 The impact of dust aerosol on climate

Dust particles interact with solar as well as terrestrial radiation. Over the cloud-free ocean the net direct radiative forcing at the top of the atmosphere (ToA) due to dust is negative due to increased shortwave albedo, which dominates over the longwave effects. In contrast, dust reduces the planetary albedo over bright surfaces (e.g., snow and ice, some deserts) and has different effects above and below bright clouds. The magnitude of all these effects depends strongly on particle size, refractive index and the altitude of dust layers (Liao and Seinfeld, 1998; Arimoto et al., 2001). The composition of dust is especially important in determining the longwave effect (Sokolik et al., 1998; Highwood et al., 2003) since refractive indices in the infrared vary considerably between dusts from different source regions. The net direct radiative impact of dust is a combination of the shortwave and longwave effects and can be positive or negative.

Estimates of global mean forcings are very uncertain, and typically vary between -0.7 and $+0.5\,\mathrm{W\,m^{-2}}$ at ToA and between -0.82 and $-1.92\,\mathrm{W\,m^{-2}}$ at the surface (Balkanski et al., 2007; Myhre and Stordal, 2001; Tegen et al., 1996; Woodward, 2001). Dust outbreaks result in a cooler surface ocean, thus affecting circulation patterns with potential regional weather and climate feedbacks (Miller and Tegen, 1998).

The accumulation of soluble material during transport as a result of heterogeneous chemistry enables dust particles to act as giant CCN and activate to cloud droplets with the potential to affect the climate through aerosol indirect effects (Levin et al., 1996). Dust particles may play a role in suppressing rainfall via the aerosol second indirect effect (Rosenfeld et al., 2001) but have also been suggested to enhance precipitation by acting as giant CCN (Feingold et al., 1999; Posselt and Lohmann, 2008) and ice nuclei. Impacts of Saharan dust on the development of tropical cyclones have also been proposed through the effects of CCN and giant CCN (Zhang et al., 2007b).

Dust can also have other indirect effects on climate. For instance, Lee et al. (2008) show that in dusty regions, CCN concentrations can be reduced by 10–20% due to particles providing a condensation sink for sulphuric acid vapor and a coagulation sink for ultrafine particles. Also, increased dust deposition on snow can reduce surface albedo leading to enhanced snow-melt and feedbacks on regional climate (Krinner et al., 2006).

Mineral dust aerosol also plays a key role in the Earth's climate system by providing iron and other nutrients to marine phytoplankton after atmosphere to ocean deposition. Jickells et al. (2005) describe how dust deposition can alleviate nutrient limitation to marine phytoplankton, linking dust emissions, ocean biogeochemistry and climate.

5.2 Climate controls on dust and feedback processes

Wind speed, soil moisture and vegetation cover are climate-driven variables that strongly affect dust emission fluxes, size distribution and mineralogical composition and hence, indirectly, control dust transport, deposition and radiative effects. Transport is also controlled by the atmospheric circulation and wet deposition by precipitation. There is, consequently, clear potential for feedbacks involving dust associated with a changing climate. However, as in the case of emissions of biomass burning aerosols from wildfires, there are both natural and anthropogenic factors governing dust emissions (Moulin and Chiapello, 2006). The attribution of changing dust emissions to natural or human causes is very uncertain for this reason and also because, unlike for purely anthropogenic species, the choice of a base time to define natural emissions is essentially arbitrary.

Some early studies (e.g., Tegen and Fung, 1995) found human influences to have increased dust emissions directly via changes in land use by as much as 20–50% (Tegen et al., 1996; Sokolik and Toon, 1996; Moulin and Ciapello, 2006). However, some studies find the contribution of dust from currently cultivated land to total dust load to be less than 10% (e.g., Tegen et al., 2004; Prospero et al., 2002). Elevated CO_2 concentrations may result in changes in the extent of desert regions in future, though the direction of such change is currently uncertain (Cox et al., 2000; Harrison et al., 2001). The combined role of climate and human intervention in controlling dust emissions makes predictions of future dust emissions unclear. Studies of the effects of anthropogenic climate change on dust loadings (neglecting land use changes) give a wide range of results from large increases (e.g., Woodward et al. (2005) find a factor of 3 increase in 2100) to large decreases (e.g., Mahowald and Luo (2003) and Mahowald et al. (2006) find a 60% decrease under double CO_2 concentration), to moderate ($\sim$10––20%) increases/decreases (e.g. Tegen et al., 2004). The large range reflects different responses and couplings of the climate and vegetation models used.

5.2.1 Interaction with the hydrological cycle

There is evidence that major dust sources tend to be located in topographic lows containing alluvial or aeolian sediments or ephemeral lakes (Prospero et al., 2002; Tegen et al., 2002; Washington et al., 2003). Some of these sources are supply limited and therefore dependent on events such as flooding to provide a source of erodible material for enhanced dust emissions (e.g., Bullard et al., 2008; Niemeyer et al., 1999; Bryant et al., 2007). Future changes in frequency of high rainfall events are likely to affect activation of such sources.

Atmospheric dust concentration measurements made routinely at Barbados since 1965 show a four-fold increase since the 1960s (Prospero and Lamb, 2003). This long-term record has been shown to have a significant correlation with drought occurrence in the Soudano-Sahel region and with dust-event frequency from Mali visibility records (Mbourou et al., 1997). However, global models have struggled to replicate the observed dust record, and the relative contributions of changes in source strength and transport efficiency are unclear (Mahowald et al., 2002).

5.2.2 Biogeochemical effects

Jickells et al. (2005) describe how dust deposition can provide nutrients to oceanic phytoplankton, linking dust emissions, ocean biogeochemistry and climate. Martin et al. (1990) hypothesized that high dust loadings during glacial periods would provide more nutrients to the phytoplankton population thereby enhancing primary productivity and oceanic CO_2 uptake, which could explain part of the glacial-interglacial difference in atmospheric CO_2 levels. A combination of field experiments and modeling (Watson et al., 2000; Bopp et al., 2003a) and analysis of sedimentary records (Kohfeld et al., 2005) suggests that when a glaciation starts, increases in the dust supply of iron to the ocean could drive up to half of the decrease in atmospheric CO_2. While there is a reasonable amount of evidences for long term and substantial changes in dust on CO_2 uptake or DMS emission by the oceans in past climates, there is still little or no observational or modeling evidence on how such feedbacks could operate in the modern atmosphere or in a warmer climate. However, studies have found strong correlations between simulated iron deposition and satellite chlorophyll (Erickson et al., 2003) and observations of net community production (Cassar et al., 2007) in the Southern Ocean suggesting aeolian dust deposition has an important role for present-day ocean biogeochemistry. Cropp et al. (2005) also found strong correlation between satellite-derived chlorophyll and aerosol optical depth, consistent with dust deposition enhancing the productivity of marine biota. It is also possible that changes in dust deposition can modulate the DMS-sulphate-cloud albedo feedback discussed in Sect. 3.1.

5.2.3 Dust-Chemistry interactions

Dust particles can react with acids during transport and become coated with soluble material (Usher et al., 2002; Underwood et al., 2001). Such dust chemistry has been shown to reduce the particle lifetime (e.g., Fan et al., 2004) and hence affect dust burden, radiative forcing and deposition to the oceans. As well as enabling dust particles to act as CCN, reactive uptake of acids also increases the solubility of iron, making it more available to the phytoplankton in the ocean (Fan et al., 2006). These observations have led to the suggestion that changes in anthropogenic SO_2 emissions over East Asia (for example) may affect carbon fixation in High Nutrient Low Chlorophyll regions of the ocean via atmospheric dust deposition (Meskidhze et al., 2003) – see Sect. 5.2.2. A further effect may be to increase the CCN activity of dust particles, thereby impacting rainfall (Levin et al., 1996).

The reactive uptake of gases on dust depends strongly on dust mineralogy. Particles with significant carbonate content (e.g. containing calcite, dolomite) are alkaline and hence strongly reactive in the presence of atmospheric acids (e.g. Claquin et al., 1999). Bauer and Koch (2005) found that interaction between sulphate and dust would reduce the sulphate direct forcing from -0.25 to $-0.18\,\mathrm{W\,m^{-2}}$ when heterogeneous oxidation of SO_2 is included, as a result of a reduced concentration of externally mixed sulphate. Dentener et al. (1996) found that 50–70% of global sulphate formation is associated with dust, however this is likely to be an overestimate because of the large mass accommodation coefficient used in this study.

Uptake of other gases to dust particles and the impact on photolysis rates can also affect oxidant concentrations and atmospheric chemistry (e.g. Bian and Zender, 2003; Liao et al., 2003).

5.3 Status and development of dust in earth system models

Studies of the feedbacks associated with dust are at an early stage. Development is needed in several sub-components of Earth system models, including the land surface, vegetation and ocean biogeochemical as well as atmospheric dust schemes.

Dust modeling has progressed considerably over the last two decades, but there are still many major uncertainties associated with all aspects of the dust lifecycle. Basic emission processes are not yet fully understood and a range of emission schemes are used which each seem to capture some but not all of the details of the deflation process (e.g. Marticorena and Bergametti, 1995; Alfaro and Gomes, 2001). The widely used saltation models do not apply to areas where other processes such as abrasion are dominant, and indeed may not apply to the largest single dust source found in the Bodele depression (Todd et al., 2007). The effects of crusting, soluble salts in the soil, supply limitation and resuspension are all generally ignored. Further research on these processes is required, to allow them to be parametrized in emission models, together with the behavior of different geomorphic types and better characterization of sub-grid-scale wind variability and soil properties. New models representing the response of vegetation to climate change are also needed, as this is a major controlling factor for dust emissions, especially in arid and semi-arid areas. Global datasets of surface properties including geomorphology, soil size distribution and mineralogical composition would aid the development of emission schemes significantly. Improved understanding and representation of the relation between horizontal and vertical dust fluxes are fundamental requirements. The use of satellite data to constrain emissions (e.g. through the use of preferential source areas) has allowed significant improvement in the identification of source regions (e.g., Prospero et al., 2002; Schepanski et al., 2007) and the agreement of models with observed optical depths (Zender et al., 2003). However in order to simulate the various feedbacks on dust production that may be influenced by climate change, more of the basic production processes will need to be represented explicitly in models.

In the atmosphere, changes in dust chemistry during transport may be important for iron availability in the oceans, although more fundamental research is needed (e.g., Fan et al., 2006). Improved understanding and parametrizations of wet and dry deposition processes are also required (Jung and Shao, 2006; Petroff et al., 2008). The representation of radiative properties is another area where development would be useful. Balkanski et al. (2007) have shown that the haematite content is critical for a correct simulation of atmospheric absorption by African dust. On a global scale, haematite in potentially arid soil types can vary at least from 0 to 7% by mass (Claquin et al., 1999), so this factor will have to be included if local dust forcings are to be calculated correctly (Krinner et al., 2006). A further complication is that haematite often forms a coating on the surface of the dust particle so mass fraction may be a poor guide to its radiative impact. In the longwave, dust refractive indices are even more variable between different source areas than in the shortwave, and the effect of this will also have to be addressed.

Better characterization of soil particle size is essential because it can substantially alter the long range transport to remote oceans (Grini and Zender, 2002) as well as affecting the radiative impact of the dust and the bio-availability of its iron content to plankton. This requires improvements to the modeling of emission, transport and deposition, all of which are size-dependent, as well as enhanced surface soil datasets.

Further observational data for model validation is required. A particular challenge is the provision of long-term data on a global scale, which are needed due to the strong spatial and temporal variability of dust concentration, size distribution and hence radiative effects. Direct measurements of dust concentrations are needed as well as AODs, to allow both modeled concentrations and parametrized radiative properties to be assessed.

In the ocean, present global biogeochemical models do not reproduce the interannual variability of surface chlorophyll as observed by the SeaWiF Satellite (Le Quéré et al., 2005), or the interannual variability of CO_2 exchange with the atmosphere as produced by atmospheric inversions (Le Quéré et al., 2007). Accurate modeling of the response of marine ecosystems and plankton speciation to changes in iron input will require a representation of plankton functional types.

6 Summary

6.1 Direction and magnitude of aerosol feedbacks

Table 3 summarises the interaction of aerosols with the Earth system, including the expected impacts, the status of existing models, and the developments in models and observations required to improve our understanding.

A review of available studies of aerosol interactions with the Earth system suggests that there is the potential for substantial feedbacks on the climate. Future climate change is likely to have significant effects on the natural environment and hence the physical and biological systems that generate aerosol. Natural aerosol is a large component of the total atmospheric aerosol load and natural climate variability already causes emissions to vary on seasonal, inter-annual and decadal timescales.

Most of the feedbacks operate through the effect of climate change on the rate of emission of natural aerosols, leading to future changes in aerosol burden and properties, with associated effects on direct and indirect radiative forcing. There are also less direct feedbacks involving impacts of climate on aerosol emissions, which subsequently impact other components of the Earth system, such as the carbon cycle or the cryosphere. For example, dust is in this latter category since it is an important source of iron for phytoplankton, which has a critical effect on the ocean carbon cycle.

The drivers of changes in aerosol are very numerous and present a substantial challenge for the development and evaluation of Earth system models. The direct climate drivers are: temperature, wind speed, precipitation (and associated convection and lightning), radiation, sea ice and snow coverage, CO_2 concentrations, nutrients (reactive nitrogen, iron, etc.), and changes in oxidants and other trace species. Anthropogenically driven changes in land use, fire suppression, etc. are also important.

The indirect drivers, which respond to the direct climate drivers, include: wind stress, ocean mixed layer depth and stratification, soil wetness, vegetation distribution, terrestrial vegetation and marine phytoplankton species composition, foliar biomass, primary productivity, and fire severity and frequency. All of these need to be simulated in Earth system models.

With our present state of knowledge and with very few integrating global model studies the natural aerosol-related feedbacks are hard to quantify in terms of a meaningful feedback factor as has been attempted for physical feedbacks. Nevertheless, existing studies suggest that responses are potentially large, at least regionally.

The terrestrial biosphere is a very large source of SOA mass in the present atmosphere and most observational and modeling studies suggest this source will increase substantially, primarily due to increases in temperature and changes in vegetation (Sect. 2.1). There is the potential for $>1\,\mathrm{W\,m^{-2}}$ global mean negative forcing due to the direct and indirect radiative effects of extra biogenic SOA in the atmosphere by mid to late century. Forcings of several Watts per square metre have been estimated or can be inferred close to aerosol sources, such as over forests.

The net effect of changes in wildfires is not known. Several factors are important but have not been studied together in Earth system models or observations. Fires emit absorbing aerosol that has either a positive or negative direct radiative effect (Sect. 2.3.1); deposition of absorbing material on snow accelerates melting; the local radiative effect is a combination of changes in biophysical effects (albedo, hydrology), carbon sequestration, and changes in biogenic secondary aerosol formation (Spracklen et al., 2008b). Despite the complex couplings and competing effects, regional studies suggest increases in wildfire emissions of >100% by 2050 and associated increases in aerosol organic carbon of >40%.

In the marine system, the effect of climate change on DMS emissions and atmospheric sulphate aerosol remains ambiguous. Global models suggest changes in DMS emissions and/or aerosol sulphate of a few percent, but regionally the forcing due to changes in cloud properties may be $>1\,\mathrm{W\,m^{-2}}$, which would impact regional climate.

However, large gaps remain in the ability of models to capture interannual changes in ocean productivity (Sect. 3.1.2), so confidence in models of DMS is probably low. The effects of changes in primary marine aerosol (sea salt and organic material) are potentially large and could conceivably exceed the impact of DMS, although more fundamental research is needed to estimate the effects.

6.2 Research requirements

6.2.1 Key uncertainties

The main open questions and gaps in our knowledge have been addressed in Sects. 2.5, 3.4 and 5.3 for each aerosol system and are summarised in Table 3. An important finding of this review is that the uncertainties in climate drivers are a very large component of the overall uncertainty in aerosol changes. For example, in marine regions the DMS emissions depend on changes in the mixed layer depth, wind speed, ocean surface temperature and sea ice extent, none of which can be confidently predicted on regional scales over the next century. For dust, it is changes in the hydrological cycle, vegetation and wind speed that will largely determine future dust levels. Some components of the Earth system have a substantial impact on the emissions of several important aerosols. For example, better quantification of biogenic secondary organic aerosols, wildfires and dust will require an improved understanding of the response of vegetation to climate change. Thus, better quantification of aerosol feedbacks requires improvements in several physical and biological components of Earth system models in parallel with improvements in aerosol processes themselves.

Some processes reviewed here have only recently been studied and much more needs to be done before we can even begin to estimate likely couplings in the Earth system. A good example is primary organic aerosol emissions from the ocean and the terrestrial biosphere. These have the potential to exert a large feedback effect on climate.

6.2.2 Measurements

Both field and laboratory measurements can and have been applied to study feedbacks and processes behind them. Process level understanding can be achieved using laboratory experiments, continuous comprehensive field observations and process models. To utilise the process understanding in global models, satellite retrievals, airborne experiments and long term continuous ground-based experimental monitoring networks are needed.

The coupling of natural aerosol emissions and processes with the functioning of physical and biological systems makes continuous comprehensive measurements particularly useful. A good example of comprehensive measurements is the SMEAR II station in the Finnish boreal forest (Hari and Kulmala, 2005). The dataset enables direct comparison of different feedbacks; for example how biogenic aerosol production is related to CO_2 concentration and fluxes as well as changes in temperature, cloudiness and rain. A network of such stations provides a rich dataset for evaluation of aerosol-Earth system interactions on regional scales (Kulmala et al., 2008a, b). A good example of the use of long data sets from several stations is the study of boreal aerosol production and change (Tunved et al., 2006a, b, 2008). In marine regions, station observations have established links between DMS, aerosol, temperature and cloudiness (e.g., Ayers and Gras, 1991; Ayers et al., 1997; Boers et al., 1994) although the lack of supporting measurements of ocean biological and physical state highlights the difficulty of comprehensive measurements in all environments. Although the number of long term dust records is small, we have learned a great deal about climate impacts by combining observations with climate data and global models (e.g., Prospero and Lamb, 2002; Mahowald et al., 2002).

Several feedback mechanism and particularly processes behind them can be tested in laboratory experiments. For example, Joutsensaari et al. (2005) and Mentel et al. (2009) studied photochemical production of aerosols using living plants in environmental chambers. In the ocean, iron solubility of real aerosol in natural sea water has been studied in a flow reactor (Wu et al., 2007), and the physical and chemical properties of aerosol generated from bubble bursting in natural sea water has been measured (Keene et al., 2007). Mesocosm experiments that probe the response of natural marine (Vogt et al., 2008) and terrestrial (Pegoraro et al., 2005) systems are valuable controlled experiments. In a similar way the effect of changing climate conditions can and has been investigated for other aerosol systems.

At larger scales, satellite observations enable long term global changes to be observed. Satellite observations have been used, for example, to measure ocean chlorophyll (as a proxy for primary productivity) and its relation to CCN, and to detect and quantify changes in wildfires. However, quantities such as chlorophyll and aerosol optical depth, while useful for some aspects of model evaluation, cannot provide the detailed information required to evaluate some key model processes.

6.2.3 Development of models

Quantification of feedbacks associated with aerosol requires an Earth system model. Such models differ from climate models in the number and complexity of the physical, chemical and biological processes that are represented. They typically aim to couple the Earth's various components: physics and chemistry of the atmosphere, oceans, cryosphere, and the terrestrial and oceanic biosphere, and to quantify the interactions between these components and their impacts on each other.

A common thread in all of the aerosol systems we have reviewed is the need for development of increasingly complex chains of models even for single feedbacks. The coupling of systems further complicates matters. Model evaluation and uncertainty analysis is therefore substantially more challenging than for physical climate systems alone. For example, modeling the effect of BVOC emissions on climate requires models of photosynthesis, autotrophic respiration and VOC synthesis at cell and leaf scales. Atmospheric models of VOC chemical degradation, particle nucleation together with condensation/evaporation and coagulation are required to understand SOA aerosol dynamics. The drivers of change in this system are related to the carbon cycle, soil nutrients and hydrology and the fate of the aerosol is related to changes in boundary layer dynamics, temperature, changes in oxidants and deposition processes on a global scale. In order to be able to simulate global climate and air quality, the most recent progress on this chain of processes must be compiled, integrated and implemented in numerical Earth system models via novel parameterizations.

What are the requirements for atmospheric models of the aerosol and chemical processes specifically? Aerosol and number and mass concentrations are not necessarily linearly dependent on each other. Therefore both should be known to predict feedbacks. For example, dust, sea salt and biomass burning will enhance mass and surface area, which will decrease new particle formation and number concentrations. As a consequence of this coupling, changes in CCN may not scale linearly with emissions (Merikanto et al., 2009). The present generation of global aerosol microphysics models are a necessary and computationally feasible component of Earth system models. However, a major obstacle to the development of Earth system models is the requirement to simplify complex and numerically demanding gas phase chemical schemes, such as for DMS, halogens, BVOC degradation and SOA formation (Sects. 2.1.1 and 3.1.3.2). Studies should seek to find minimum schemes with maximum explanatory power if they are to be most useful in Earth system studies.

Glossary

AOD: aerosol optical depth

BC: black carbon

BVOC: biogenic volatile organic compounds

CCN: cloud condensation nucleus

CLAW: Charlson-Lovelock-Andreae-Warren hypothesis

DMS: dimethylsulphide
DMSP: dimethylsulphoniopropionate
ESM: Earth System Model
FT: free troposphere
GCM: general circulation model
MBL: marine boundary layer
MSA: methanesulphonic acid
OC: organic carbon
PAR: photosynthetically available radiation
PBAP: primary biological aerosol particles
RF: radiative forcing
SOA: secondary organic aerosol
SRD: solar radiation dose
VOC: volatile organic compounds

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Table 1. Effects of various environmental changes on global isoprene emissions in modeling studies.

	Scenario	Δisoprene emission	Time period
		Climate effect only	
Wu et al. (2008)	A1B; CO_2 522 ppmv, +1.6 K	+25%	2000–2050
Sanderson et al. (2003)	IS92a, +4.7 K	+34%	1990–2090
Heald et al. (2008)	A1B; +1.8 K	+22%	2000–2100
Liao et al. (2006)	A2; +4.8 K	+55%	2000–2100
		Climate effect and dynamic vegetation	
Heald et al. (2009)	A1B; 717 ppmv	Factor 3.6	2000–2100
Turner et al. (1991)	$2\times CO_2$	+25%	N/A
Sanderson et al. (2003)	IS92a, +4.7 K	+27%	1990–2090
Lathiere et al. (2005)	560 ppmv CO_2	+27%	1990–2100
Wiedinmyer et al. (2006)	$2\times CO_2$; +1.7 K	+70%	1990–2100
Arneth et al. (2007b)	Various	+77 to +90% (A2 SRES); 38% to 48% (B1 SRES)	1980–2100
		(Anthropogenic) land cover changes only	
Lathiere et al. (2006)	Global tropical deforestation	−29%	–
Guenther et al. (2006)	IMAGE landcover database	−30%	2000–2100
		Climate and CO_2 inhibition	
Heald et al. (2009)	A1B	−8%	2000–2100
		Climate and CO_2 inhibition and dynamic vegetation (no anthropogenic vegetation change)	
Heald et al. (2009)	A1B	Factor 2.4	2000–2100
Arneth et al. (2007b)	Various	−9% to −16% (A1 SRES); 0% to −7% (B1 SRES)	1980–2100

Table 2. Projected change to BVOC emissions (isoprene (I), monoterpenes (M)) and SOA burden by 2100 due to changing climate and emissions.

	Emissions scenario	Global surface ΔT	Vegetation	Change in BVOC emissions	Change in SOA burden
Liao et al. (2006)	A2 SRES	+4.8 K	Fixed	58% (M), 55% (I)	+54% (M only)
Heald et al. (2008)	A1B SRES	+1.8 K	Fixed	+19% (M), +22% (I)	+26%
Tsigaridis and Kanakidou (2007)	IS92a	Uncoupled (+1 K BL, +2 K FT)	Dynamic	+120% (M), +37% (I)	+150%

Table 3. Status of earth system models, developments needed, key observations, possible feedbacks and the level of scientific understanding.

Aerosol type	Model status	Model challenges and developments	Key observations needed	Direction and magnitude of feedback by 2100	Level of Scientific Understanding
Biogenic Secondary Organic Aerosol (SOA)	Some studies of the effect of T on BVOC emissions (mostly isoprene and monoterpenes) based on semi-empirical models. Limited studies of the effect of CO_2 and vegetation changes on BVOC emissions. Limited studies of the impact of changing climate and BVOC emissions on SOA.	Response of vegetation dynamics to climate change (resolving BVOC-emitting species) Expanded range of biogenic emission drivers (soil nutrients, nitrogen deposition, CO_2, ozone, etc.). Expanded range of emitted compounds (e.g., sesquiterpenes) and controlling factors Size segregated aerosol dynamics	Biogenic SOA budget Interannual variations in emissions and drivers. Role of isoprene and other terpenes in SOA formation Physico-chemistry of the gas-aerosol equilibrium of semi-volatile organic species. Connections and interaction between carbon and nitrogen cycles with aerosols and their precursors Continuous and comprehensive measurements to observe soil, ecosystem and atmosphere relations	Likely increase in biogenic SOA (25 to 150%). Likely negative radiative forcing. Locally a few $\mathrm{W\,m^{-2}}$. Globally $>1\,\mathrm{W\,m^{-2}}$.	Poor
Primary Biological Aerosol Particles (PBAP)	Very limited studies of global PBAP emissions. No studies of potential change due to climate.	Inclusion of PBAP	High temporal resolution PBAP observations in a range of environments to elucidate emission mechanisms and drivers.	Unknown	Very poor
Aerosol from wildfires	Very limited uncoupled regional studies of changes in burned area with climate change.	Coupling of fire models and vegetation models. Impact of fire suppression policies on fire occurrences. Global analysis of response to climate change. Role of secondary organic aerosol.	Optical properties of biomass burning aerosols. Records of deposition on snow and ice, including optical properties. Size distribution and CCN properties of particles versus age. Long term global records of fire occurrence.	Likely increase in wildfire occurrence (∼100% possible) and carbonaceous aerosol concentrations (up to 40%). Global mean climate impact positive or negative. Positive in Arctic	Very poor
Marine sulphate aerosol	Some coupled studies of the phytoplankton-DMS-aerosol-cloud feedback. New generation sub-models of DMS chemistry, aerosol dynamics and marine biota not yet fully coupled	Ocean response to climate change (mixing, stratification, nutrients, etc.) Ocean biota models to capture observed ocean productivity and DMS observations DMS production in region of sea-ice and sea-ice changes Budget of DMS in the seawater including destruction by UV. Response of plankton dynamics to climate change (resolving DMS-emitting and non-DMS-emitting species). Fast global models of sulphur-halogen-oxidant chemistry, including multiphase processes	DMS production in region of sea-ice. DMS in coastal regions Global marine CCN and variability	Positive or negative. Global mean few % increase in sulphate. Locally $>1\,\mathrm{W\,m^{-2}}$	Poor

Table 3. Continued.

Aerosol type	Model status	Model challenges and developments	Key observations needed	Direction and magnitude of feedback by 2100	Level of Scientific Understanding
Marine organic aerosol	Lack of model	Organic aerosol source function Role of organic aerosols in the aerosol indirect effect	Organic aerosol source function linked to biological and physical drivers Speciation and solubility of the marine organic aerosol	Unknown	Very poor
Sea salt	Some studies of the response of sea salt emissions to climate change	Indirect effect of sea salt on clouds. Improved sea-salt emission scheme Evaluation of GCM wind speed response to climate change	Long term change in surface wind speed in response to climate change, including in regions of changing sea ice. Additional factors controlling sea spray emission flux (surfactants, fetch, etc.) Mixing state of sea-salt with primary marine organics.	Likely negative	Poor to medium
Dust	A few studies of the dust response in climate change simulations	Response of ocean biota models to iron. Evolution of dust sources with changing climate (e.g. activation of supply limited regions after high rainfall events). Allow soil mineralogy to affect dust properties (iron content and speciation, refractive index) Better characterization of soil wetness and vegetation response to climate change. Improved emission schemes	Interannual variability of ocean, chlorophyll and CO_2 exchange Iron speciation, haematite content and geomorphology of soils Factors controlling iron bioavailability Ice nucleating ability of dust versus mineralogy Refractive indices of dusts from different source regions Long-term observations of atmospheric dust concentrations with wide geographic coverage Global observational climatology of soil moisture	Possible large positive or negative change in dust burden (up to factor 3). Positive or negative forcing.	Very Poor
Stratospheric and volcanic aerosols	Some studies of the change in stratospheric tropospheric exchange with climate change. Few studies of volcanic aerosol impacts on climate	Role of organics in the stratospheric aerosol system Aerosol effects on diffuse radiation	Budget of stratospheric aerosols Diffuse radiation and biospheric response	Small feedback effect of climate on processes Potential large impact of eruptions on biosphere	Low

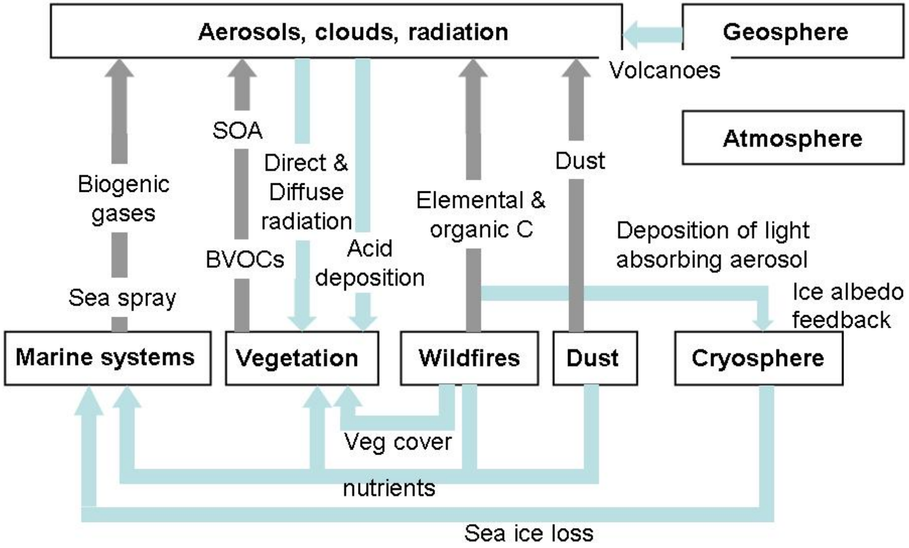

Fig. 1. Interactions between different components of the Earth system connected by aerosol.

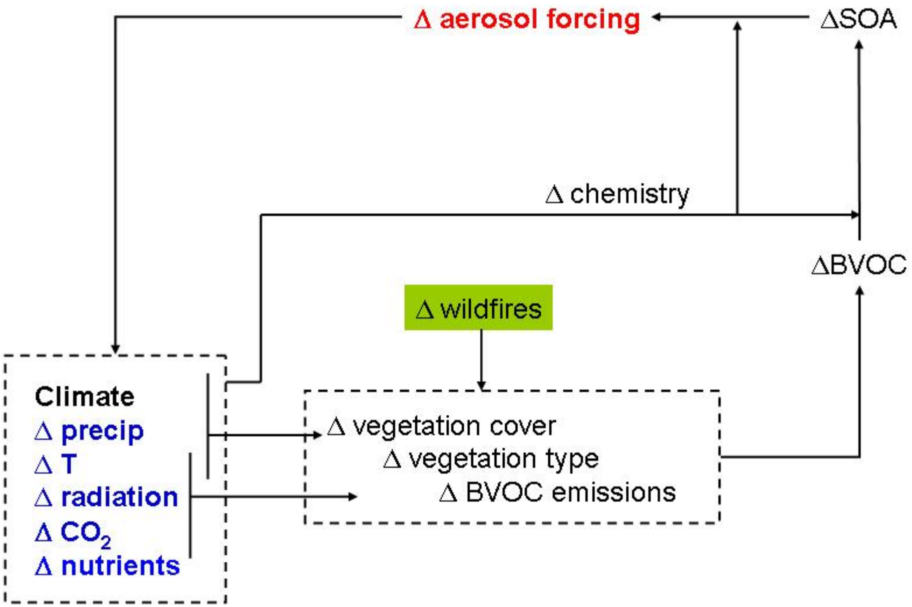

Fig. 2. The response of terrestrial biogenic secondary organic aerosol to climate change. Climate change drivers are indicated in blue, processes in black, and atmospheric impacts in red. The green box contains another set of drivers/feedbacks described in Sect. 2.3.

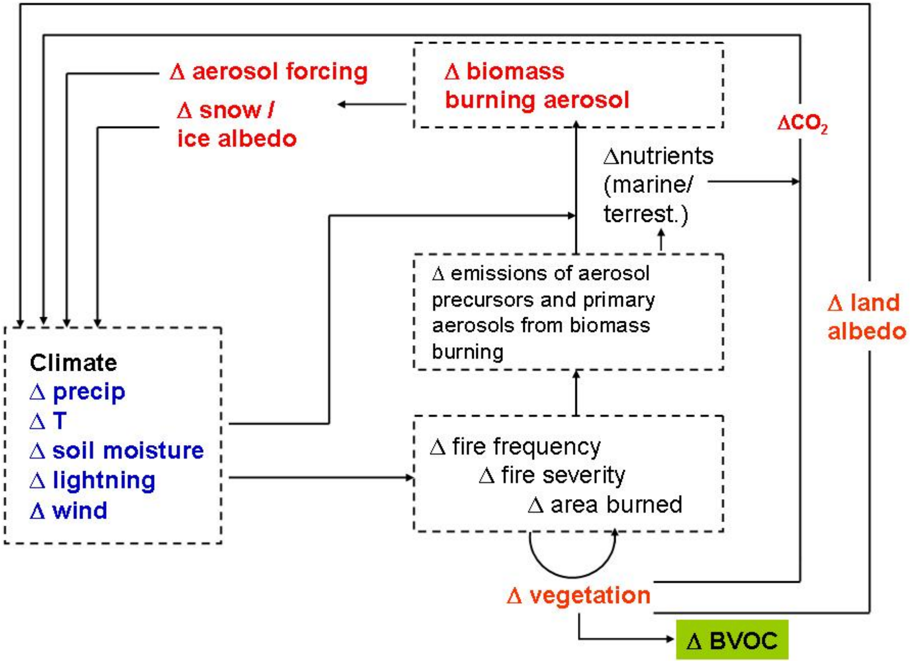

Fig. 3. Possible feedbacks associated with wildfires. Climate change drivers are indicated in blue, processes in black, atmospheric impacts in red. The green box contains another set of drivers/feedbacks described in Sect. 2.1.2. Primary biological aerosol particles are not included.

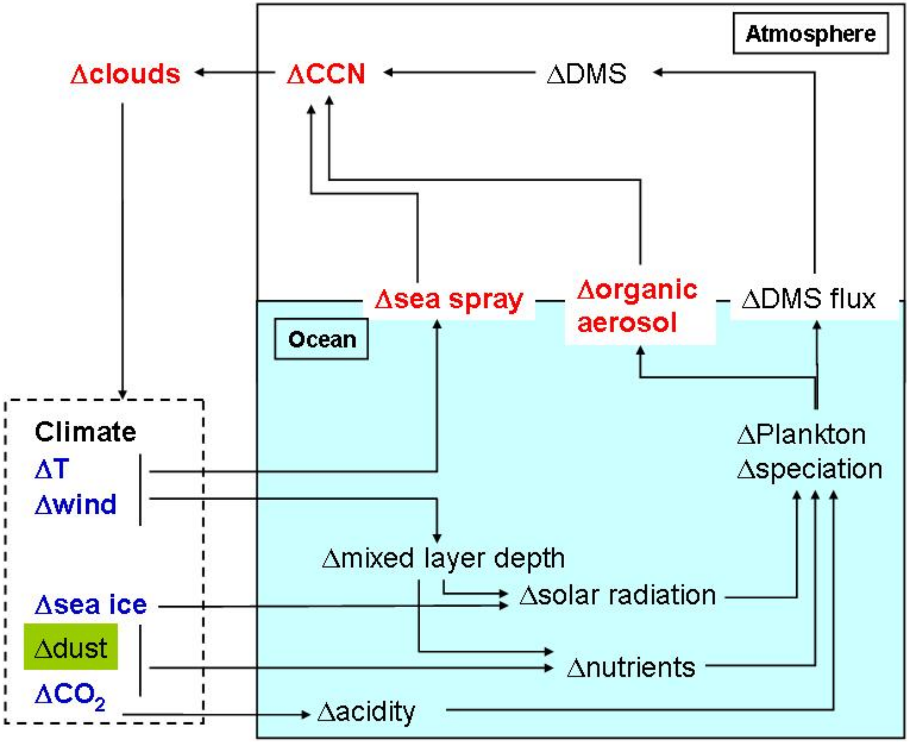

Fig. 4. The response of marine primary and secondary aaerosol to climate change. Climate change drivers are indicated in blue, processes in black, and impacts in red. The green box includes a separate set of feedbacks discussed in Sect. 5.

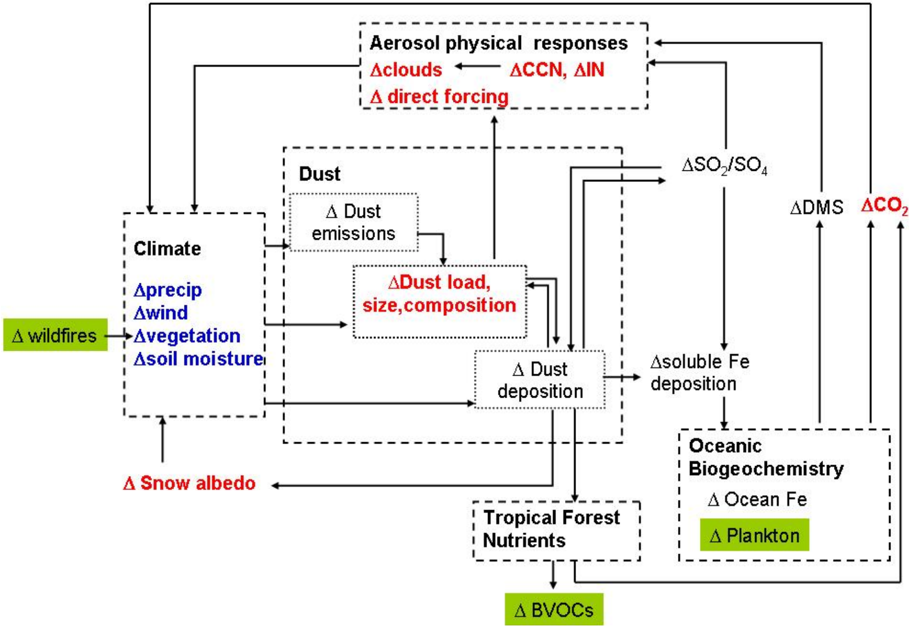

Fig. 5. Possible feedbacks involving dust. Climate change and anthropogenic drivers are indicated in blue, processes in black, and impacts in red.