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Simulating aerosol microphysics with the ECHAM4/MADE GCM – Part II: Results from a first multiannual simulation of the submicrometer aerosol

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Abstract. First results of a multiannual integration with the new global aerosol model system ECHAM4/MADE are presented. This model system enables simulations of the particle number concentration and size-distribution, which is a fundamental innovation compared to previous global model studies considering aerosol mass cycles only. The data calculated by the model provide detailed insights into the properties of the global submicrometer aerosol regarding global burden, chemical composition, atmospheric residence time, particle number concentration and size-distribution. The aerosol components considered by the model are sulfate (SO_4), nitrate (NO_3), ammonium (NH_4), black carbon (BC), organic matter (OM), mineral dust, sea salt and aerosol water. The simulated climatological annual mean global atmospheric burdens (residence times) of the dominant submicrometer aerosol components are 2.25 Tg (4.5 d) for SO_4, 0.46 Tg (4.5 d) for NH_4, 0.26 Tg (6.6 d) for BC, and 1.77 Tg (6.5 d) for OM. The contributions of individual processes such as emission, nucleation, condensation or dry and wet deposition to the global sources and sinks of specific aerosol components and particle number concentration are quantified. Based on this analysis, the significance of aerosol microphysical processes (nucleation, condensation, coagulation) is evaluated by comparison to the importance of other processes relevant for the submicrometer aerosol on the global scale. The results reveal that aerosol microphysics are essential for the simulation of the particle number concentration and important but not vital for the simulation of particle mass concentration. Hence aerosol microphysics should be taken into account in simulations of atmospheric processes showing a significant dependence on aerosol particle number concentration. The analysis of the vertical variation of the microphysical net production and net depletion rates performed for particle number concentration, sulfate mass and black carbon mass concentration unveils the dominant source and sink regions. Prominent features can be attributed to dominant microphysical processes such as nucleation in the upper troposphere or wet deposition in the lower troposphere. Regions of efficient coagulation can be identified.

1 Introduction

Aerosols are known to play an important role not only in many atmospheric processes, but also in environmental protection policy and when concerning adverse effects on public health. Absorption and scattering of solar radiation by aerosols (direct effect) as well as aerosol impacts on cloud formation and cloud microphysical properties (indirect effect) are important aspects concerning the Earth's climate, in particular when predicting future climate change. In addition, aerosols are involved in atmospheric chemistry, e.g. by providing surface area for heterogeneous reactions. Recently, aerosols received increasing attention in environmental policy as the directive 1999/30/EC of the European Council obliges mandatory limit values for particulate matter (PM10) in ambient air since 2005.

Because of the high spatial and temporal variability of aerosols, measurements alone are not sufficient to assess climatological average properties of the aerosol over the whole globe, which are needed to address global climate impact and health effects of particulate matter. Thus, the assistance of evaluated global models including detailed representations of atmospheric aerosols is needed to make further progress in understanding the global aerosol cycles and the global impact of particulate matter.

Most current global climate models include aerosols in the form of prescribed climatologies or treat aerosol mass

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only (e.g. Roeckner et al., 1996, 2003; Feichter et al., 1996; Lohmann et al., 1999a; Adams et al., 1999). Important aspects such as particle size and particle number concentration have to be obtained diagnostically in such models. The high computing power of current super computers allows for a more detailed representation of aerosols not only in global transport models (e.g. Spracklen et al., 2005a,b) but also in general circulation models (e.g. Gong et al., 2003; Easter et al., 2004; Stier et al., 2005) and enabled the successful coupling of the general circulation model ECHAM4 (Roeckner et al., 1996) and the modal aerosol microphysics module MADE (Lauer et al., 2005). Due to the high relevance to climate research, the representation of aerosols is currently also subject to improvement in some other general circulation models. Different approaches and techniques are applied regarding the representation of the aerosol size-distribution (modal with fixed standard deviation or bin scheme), the number of modes or bins, the aerosol components considered and the number of aerosol microphysical processes included. The progress achieved so far is well documented under the framework of the AeroCom Aerosol Model Intercomparison Initiative (Textor et al., 2006). Model results and further information can also be found on the AeroCom web page http://nansen.ipsl.jussieu.fr/AEROCOM/.

The new model system ECHAM4/MADE is able to calculate particle number concentration and the aerosol size-distribution in addition to the mass concentrations of the aerosol components sulfate (SO_4), ammonium (NH_4), nitrate (NO_3), black carbon (BC), organic matter (OM), sea salt, mineral dust and aerosol liquid water (H_2O). In the present study, a so-called time slice experiment has been performed to obtain new insights into the properties of the global submicrometer aerosol. Averaging all 10 model years calculated, climatological properties of the global aerosol, representative for the beginning 1990s, are obtained. Besides the characterization of the global submicrometer aerosol, the simulations are used to assess the roles of individual processes such as emission or deposition. In particular aerosol microphysical processes (nucleation, condensation, coagulation) acting on the aerosol size-distribution through changing the aerosol mass or particle number concentration are investigated. This analysis allows a quantitative assessment of the specific role of aerosol microphysics newly included in the model system for the simulation of submicrometer particles on the global scale. Previous investigations of the contribution of individual sources and sinks to the global aerosol budget (e.g. Adams and Seinfeld, 2002; Stier et al., 2005) are upgraded by taking into account particle mass concentration, particle number concentration, and, in particular, different size-ranges (modes).

Section 2 gives a brief overview of the model system and model configuration used in this study. The characteristics of the global aerosol regarding particle mass concentrations, particle composition, atmospheric residence time and global burden, particle number concentration, and size-distribution are presented in Sect. 3. The role of aerosol microphysics for the global simulation of particle mass concentration and particle number concentration is discussed in Sect. 4. The main conclusions of this study are summarized in Sect. 5.

2 Model description

The coupled model system ECHAM4/MADE consists of two main components: the general circulation model (GCM) ECHAM4 and the modal aerosol microphysics model MADE. Each part will be described briefly in the following. Details on the model and techniques used can be found in Lauer et al. (2005) and the references therein. For a detailed evaluation of ECHAM4/MADE and comparison of modeled mass concentrations of multiple aerosol components, particle number concentrations, and size-distributions with observations, we also refer to the first part of this study (Lauer et al., 2005).

2.1 The ECHAM4 GCM

The ECHAM4 GCM (Roeckner et al., 1996) is a spectral model. The horizontal resolution applied in this study is T30, which corresponds to a Gaussian grid of about $3.75^\circ \times 3.75^\circ$ (longitude by latitude). The atmosphere is divided into 19 vertical layers ranging from the surface up to 10 hPa ($\approx$30 km). The ECHAM4 version applied in this study includes two major upgrades of the standard version: upgraded cloud microphysics (Lohmann et al., 1999a) and an aerosol module describing the mass concentrations of several aerosol species (Feichter et al., 1996; Lohmann et al., 1999a). In order to be consistent with Lauer et al. (2005), this aerosol module is used to drive the cloud and the radiation schemes of the GCM. MADE has been run in passive mode, neglecting feedbacks from the aerosol calculated by MADE to the model dynamics. This allows us to study the impact of aerosol microphysics on aerosol properties alone. Changes introduced by altered model dynamics and cloud properties would introduce additional uncertainties in investigating the relevance of aerosol microphysics to the simulation of global aerosol. For a more detailed description of this model set up, we refer to the first part of this study (Lauer et al., 2005).

2.2 Aerosol microphysics model MADE

The aerosol microphysics model MADE (Ackermann et al., 1998) (modal aerosol dynamics model) describes the aerosol size-distribution in a simplified manner by the sum of three log-normally distributed modes: the Aitken (diameter typically smaller than 0.07 µm), accumulation (diameter between about 0.07 µm to 1 µm) and coarse mode (diameter larger than 1 µm). As this study focuses on the submicrometer aerosol, coarse particles are not considered to reduce the computational burden of the model. This is justified by the typically rather little interaction between fine and coarse particle size-ranges (Whitby, 1978). Coarse particles are mostly produced by mechanical processes, whereas fine particles are usually produced by combustion, condensation, or nucleation processes. Hence, the sources of fine and coarse particles are generally well separated. This counteracts the interaction between these size modes. Furthermore, the time scale of transferring mass from fine to coarse mode by coagulation is typically in the order of weeks whereas the usual residence time of fine particles is only in the order of days. A calculation of Binkowski and Roselle (2003) for a typical average continental aerosol size-distribution showed, that the Aitken mode loses only about 0.1%/hr of particle number concentration and about 0.02%/hr of particle mass to the coarse mode due to intermodal coagulation. The calculated loss rates of the accumulation mode to the coarse mode are even lower (0.002%/hr and 0.0008%/hr for number and mass concentration, respectively). Thus, about 4 weeks would be needed to reduce Aitken mode particle number concentration to 50% due to intermodal coagulation with coarse particles, which is substantially longer than the typical residence times of Aitken mode particles. Hence, the coarse mode can be omitted without losing much accuracy when focusing on average properties of submicrometer particles under typical continental conditions. Nevertheless, the concentrations of sea salt and dust in the accumulation mode could be overestimated by the model due to neglect of intermodal coagulation with coarse particles which can show comparatively large concentrations close to their sources. However, this overestimation is limited by the short residence time of the coarse particles due to efficient sedimentation. Nevertheless, the coarse mode can be easily switched on to allow, for instance, the calculation of aerosol optical properties.

All particles are assumed to be internally mixed (see Lauer et al. (2005) for a discussion of this assumption). Aerosol components considered are SO_4, NH_4, NO_3, BC, OM, sea salt, mineral dust and aerosol liquid water. In addition to the mass concentration of these individual aerosol components of each mode, particle number concentration and the median particle diameter of each mode are calculated taking into account emission of primary particles, nucleation and condensation of sulfuric acid vapor, coagulation, size-dependent wet and dry deposition, aerosol chemistry, gas/aerosol partitioning and cloud processing. To reduce numerical expenses, the standard deviations of the modes are kept constant in this MADE version (σ=1.7 for the Aitken mode and σ=2.0 for the accumulation mode).

3 Characteristics of the global aerosol

Results obtained from a 10-year integration with the new model system ECHAM4/MADE provide a detailed picture of the global distribution and physical and chemical characteristics of atmospheric submicrometer particles. In addition to chemical composition, aerosol number concentration and size-distribution are calculated. Furthermore, mass concentration, global burden and average residence time of each aerosol component are computed for the two size-ranges Aitken and accumulation mode.

For the present study, the GCM has been run in quasi-equilibrium mode with sea surface temperatures (SST), greenhouse gas concentrations and other boundary conditions prescribed according to measurements taken around the year 1990. (Such a simulation is usually called "1990 time slice-experiment"). The 10 years analyzed here were preceded by a 4-year spin-up period, which has been discarded. The majority of the emission data used for characteristic global aerosol precursor and primary particle emissions are representative for the mid 1980s to early 1990s. Details and references on the emission data and the size-distributions used for the emissions of primary particles are given in Lauer et al. (2005).

3.1 Particle mass concentration

Global distributions of particle mass concentrations have been simulated in numerous previous GCM studies (e.g. Feichter et al., 1996; Adams et al., 1999; Chin et al., 2000; Takemura et al., 2000; Chung and Seinfeld, 2002). As particle mass concentrations are closely related to particle number concentration and size-distribution, we will also present particle mass concentrations of selected species simulated with ECHAM4/MADE to provide a complete picture of the characteristics of the global submicrometer aerosol. Figures 1–4 show the climatological annual means of the near surface mass concentrations of selected species as well as corresponding zonal mean vertical cross sections obtained from the 10-year integration. As a new feature results obtained for both individual submicrometer modes are presented, rather than total concentrations. In order to facilitate the analysis of the altitudinal variation, all aerosol mass concentrations presented in this section are transformed to standard temperature and pressure (STP) conditions, i.e. 273 K, 1013 hPa.

3.1.1 Sulfate and ammonium

Figure 1 shows the global distributions of the annual mean sulfate mass concentrations of the Aitken and accumulation mode in the lowest model layer and corresponding vertical distributions. In general, the total sulfate mass concentration is dominated by the accumulation mode, whereas the Aitken mode has only a small contribution.

The geographical distribution shows sulfate maxima in Central Europe, the eastern United States, and Southeast Asia. These regions correspond with areas of high SO_2 emissions from fossil fuel combustion. In contrast to, for instance, BC, sulfate is a secondary aerosol, i. e. almost no sulfate is emitted by the sources directly, but results from gaseous precursors which are transformed to SO_4 by chemical reactions.

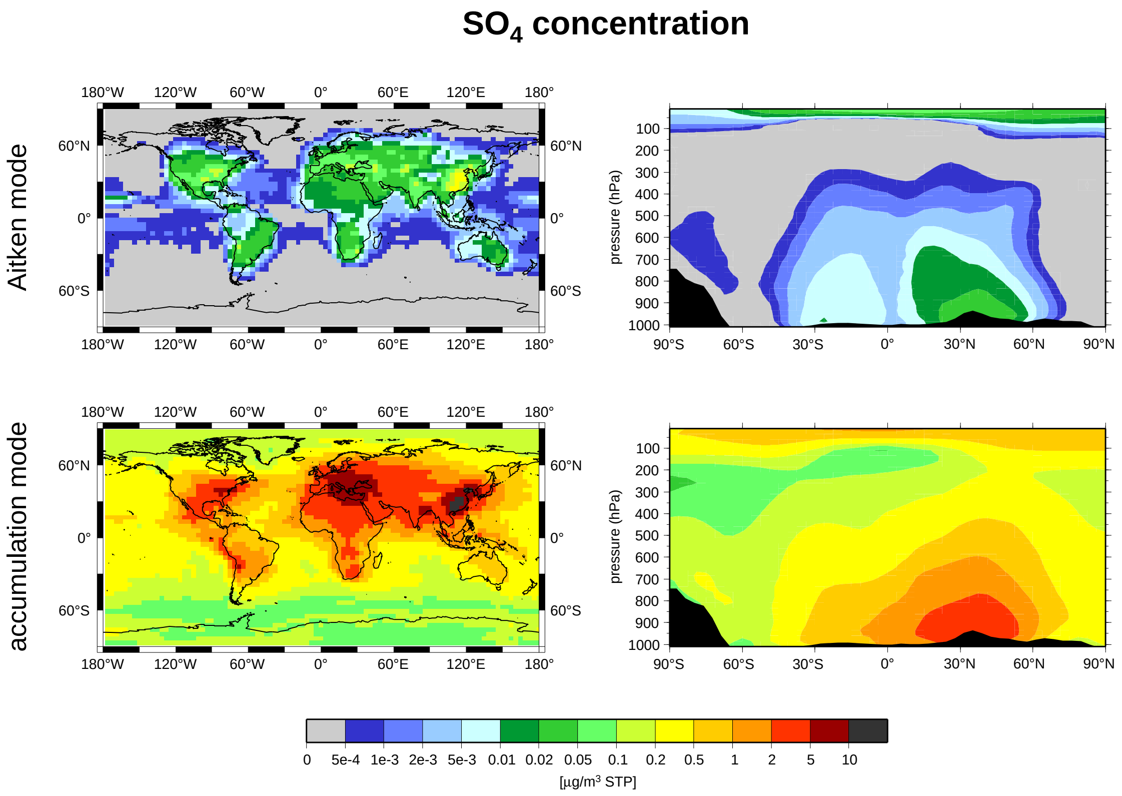


Fig. 1. Climatological annual means of simulated SO_4 mass concentration for Aitken (top) and accumulation mode (bottom) obtained from a 10-year integration. For each mode, the geographical distribution of the lowermost model layer (left), which is about 60 m thick, and the zonally averaged vertical cross section (right) are shown. The concentrations are given at STP conditions (see text for details).

Thus, the maxima in the geographical distribution have a larger extent than in the case of primary aerosols (e.g. Fig. 2). Overall, the basic characteristics of the patterns of the geographical SO_4 distribution follow the distribution of the SO_2 emissions. These basic characteristics of the geographical distribution of total SO_4 mass (sum of Aitken and accumulation mode) obtained in our simulations are similar to the results of other model simulations (e.g. Feichter et al., 1996; Adams et al., 1999; Takemura et al., 2000). Compared to measurements, ECHAM4/MADE reproduces the SO_4 concentrations reasonably well. However, sulfate mass is overestimated compared to measurements for European summer conditions (see Lauer et al. (2005) for details).

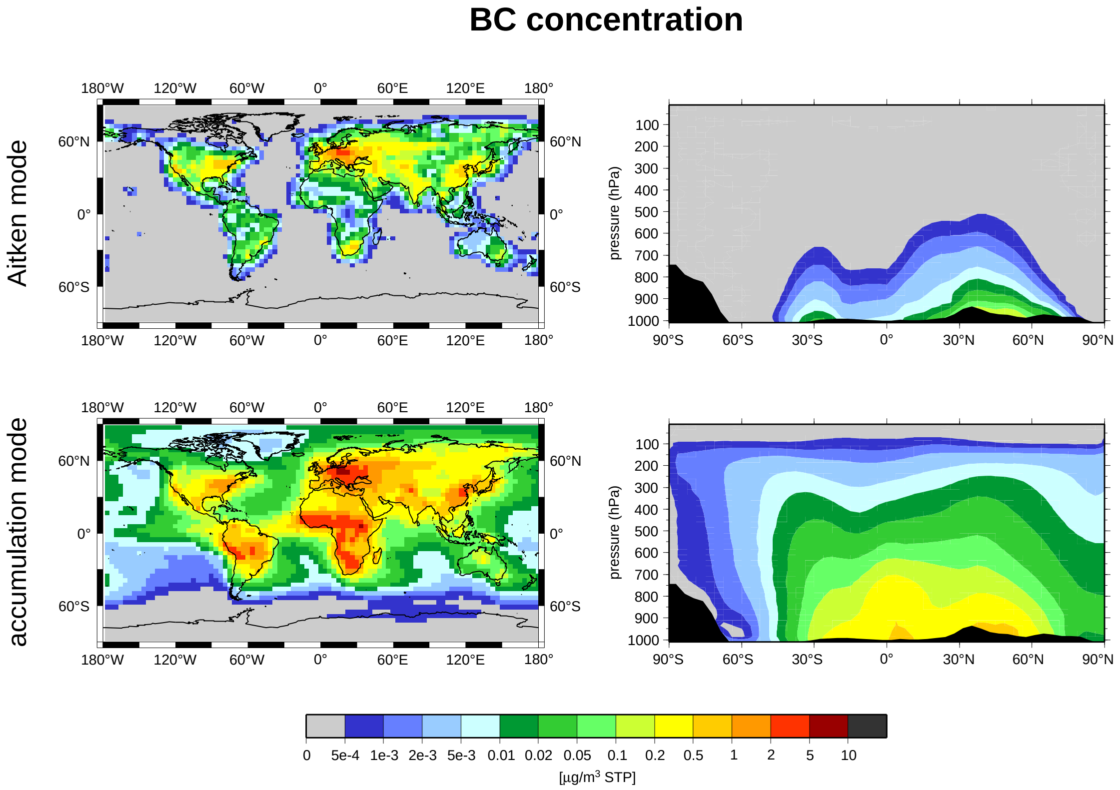
As a direct result of the smaller average residence time of Aitken particles compared to accumulation mode particles, the pattern of the Aitken mode is spread out less around the areas of maximum emissions than the one of the accumulation mode. (For a detailed analysis of the efficiency of the major sinks of sulfate in the different modes, we refer to Sect. 4.2.)

The zonal mean cross sections of both modes show highest sulfate concentrations in the boundary layer of the northern hemisphere between 20°N and 50°N (Central Europe, USA, and Asia). With increasing height, the SO_4 mass concentrations decrease. This results from sink processes, in particular wet deposition, which compensate the source processes for sulfate (gas and liquid phase production of SO_4) at higher altitudes. The tropopause region shows no enhanced SO_4 mass concentrations, although strong nucleation of the binary system H_2SO_4-H_2O takes place in these altitudes (Sect. 3.4). The newly formed particles tend to remain small on average due to the limited availability of SO_4 precursors.

In the model, the geographical distribution of ammonium (not shown) closely follows the one of sulfate, since most NH_4 is associated with SO_4.

3.1.2 Black carbon and organic matter

In contrast to the geographical distribution of sulfate, the annual mean black carbon mass concentration does not only show maxima in Central Europe, the eastern United States and Southeast Asia, but also in central parts of South America and Africa (Fig. 2). The maxima in the highly polluted areas of the northern hemisphere are mainly due to fossil fuel combustion, whereas enhanced BC concentrations in South America and Africa are caused by biomass burning activities. As for sulfate, the geographical distribution of BC in



BC concentration

Fig. 2. As Fig. 1, but for black carbon (BC).

the Aitken mode reveals a less distinctive long-range transport than the accumulation mode. As in the case of sulfate, this is due to the shorter average residence time of Aitken mode particles compared to particles in the size-range of the accumulation mode. (For a detailed analysis of the efficiency of the major sinks of BC in the different modes, we refer to Sect. 4.3.) The geographical distribution as well as absolute values of the total BC mass concentration simulated by ECHAM4/MADE are within the range spanned by the results of other model studies (e.g. Chung and Seinfeld, 2002; Cook et al., 2002; Jacobson, 2002). Nevertheless, the absolute values simulated by ECHAM4/MADE tend to be at the upper end of this range. Comparisons of measured BC mass concentrations with the ECHAM4/MADE results reveal that the model tends to overestimate BC by a factor of about 2-3 in regions of high emissions. This is mainly caused by the relatively high emission rates (mid 1980s) considered in the model. For further details of the comparison we refer to Lauer et al. (2005).

The zonal mean cross sections also shown in Fig. 2 reveal that the highest accumulation mode BC concentrations are simulated for the lowermost boundary layer at the northern mid-latitudes (resulting in particular from European emissions) and near the equator (mainly caused by emissions in Central Africa). Due to tropical convection, accumulation mode BC particles emitted near the equator can reach higher altitudes than particles emitted at mid-latitudes. The BC mass concentration decreases with increasing height and with increasing distance to the source regions. Aitken mode BC shows maximum concentrations near the surface at northern mid-latitudes, but not in the tropical latitudes. This is a consequence of assuming different size-distributions of BC particles emitted by fossil fuel combustion in the mid-latitudes and biomass burning in the tropics. Fossil fuel combustion results in a much larger fraction of primary BC particles emitted in the size-range of the Aitken mode than biomass burning (for details, see Lauer et al. (2005)). Due to the relatively short average residence time of Aitken mode particles, the Aitken mode mass concentration decreases more rapidly with increasing distance to the source regions at the surface than the accumulation mode mass concentration.

The most basic features of the geographical distribution of organic matter (not shown) follow the distribution of black carbon with the exception of South America. Here organic matter is also produced from Terpene emissions, which do not alter the BC distribution.

3.1.3 Mineral dust

Currently, the submicrometer fraction of mineral dust in ECHAM4/MADE is represented by the accumulation mode

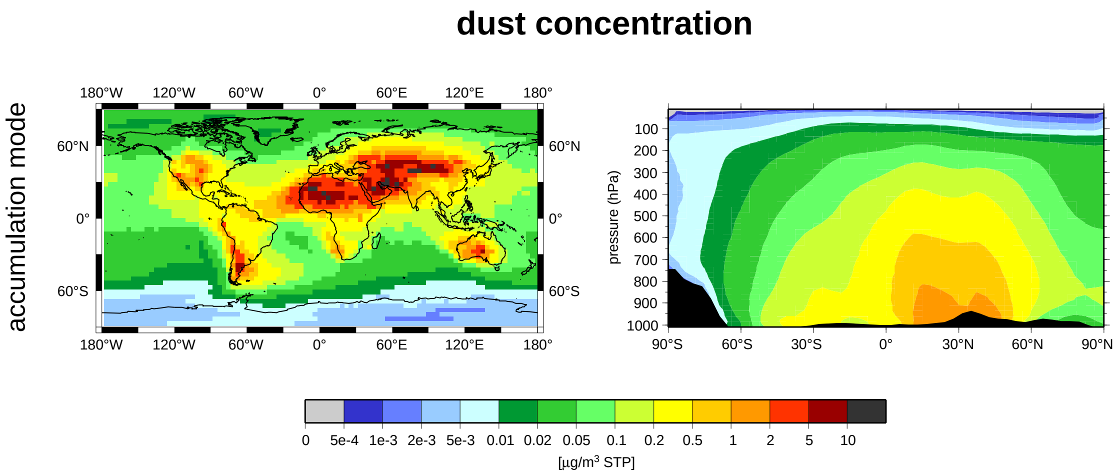


Fig. 3. As Fig. 1, but for mineral dust (accumulation mode only).

only. As discussed in Sect. 2.2, the coarse fraction has not been taken into account to reduce the overall computational burden of the model. Since most of the total mass of mineral dust is associated with particles in the size-range of the coarse mode, the omission of these particles complicates a reasonable comparison with results of other model studies simulating mineral dust. (For a comparison of the total atmospheric burden of submicrometer dust simulated here and in another study, see Sect. 3.3).

The main source regions for mineral dust are the Sahara (North Africa), Arabian deserts (Arabian Peninsula and Asia), Gobi (Asia), the Great Victoria Desert (Australia) and the Atacama (South America). In addition, mineral dust is also emitted at the Great Plains in the western United States (Fig. 3).

The vertical cross section of simulated accumulation mode mineral dust concentrations shows maxima in the boundary layer around 30° N, where the major source regions are located. As mineral dust is emitted at the surface only, mass concentrations decrease with increasing height. As for all other aerosol compounds simulated, the mineral dust concentration south of 75° S is very low. The calculated dust concentration does not exceed 0.01 $\mu g/m^3$ at all heights at these latitudes. This is a result of the, on climatological average, weak airmass exchange of the southern polar zone with the circumventing atmosphere, preventing mineral dust particles from being transported into these latitudes.

3.1.4 Sea salt

As discussed for mineral dust and in Sect. 2.2, the coarse particle mode is not considered in the present study. Therefore only sea salt particles in the size-range of the accumulation mode are taken into account. Maximum accumulation mode sea salt concentrations are simulated for the marine boundary layer of the mid-latitudes (Fig. 4), a well known feature of the geographical distribution of sea salt particles.

Inspecting the modeled vertical distribution of sea salt, the highest mass concentrations are found in the boundary layer of the main source regions, in particular between 30° S and 60° S. The sea salt mass concentration decreases rapidly with height. This is a direct consequence of the hygroscopicity and the large diameters of the primary sea salt particles emitted, facilitating a very effective removal by wet deposition.

3.2 Particle composition

Figure 5 shows the climatological global average (dry) particle composition of submicrometer aerosols obtained from the 10-year model simulation with ECHAM4/MADE on four different model levels (≈1000 hPa, 850 hPa, 500 hPa and 250 hPa). In the lowermost boundary layer (1000 hPa), particle mass in the size-range of the accumulation mode is dominated by organic matter (48%), followed by sulfate (23%), mineral dust (12%), ammonium (8%), black carbon (6%), sea salt (3%), and nitrate (1%). This shows that the surface emissions play a dominant role determining the particles main components near the surface, as the composition of the emissions of particulate matter is similar to the composition of the accumulation mode particles at this altitude. The average mass concentration of accumulation mode particles decreases from about 4.6 $\mu g/m^3$ in the lowermost model level to 0.25 $\mu g/m^3$ (STP conditions) at 250 hPa. The sulfate fraction increases with height to about 55% at 250 hPa. This is a consequence of sulfate gain due to gas phase oxidation of SO_2 followed by condensation of the sulfuric acid vapor formed and in-cloud production of SO_4 throughout the atmosphere. As long as free excess ammonia is present in the atmosphere, the increase in sulfate causes an increase in ammonium (7.7% in the accumulation mode at 1000 hPa to 9.1% at 850 hPa) as additional ammonium sulfate is formed.

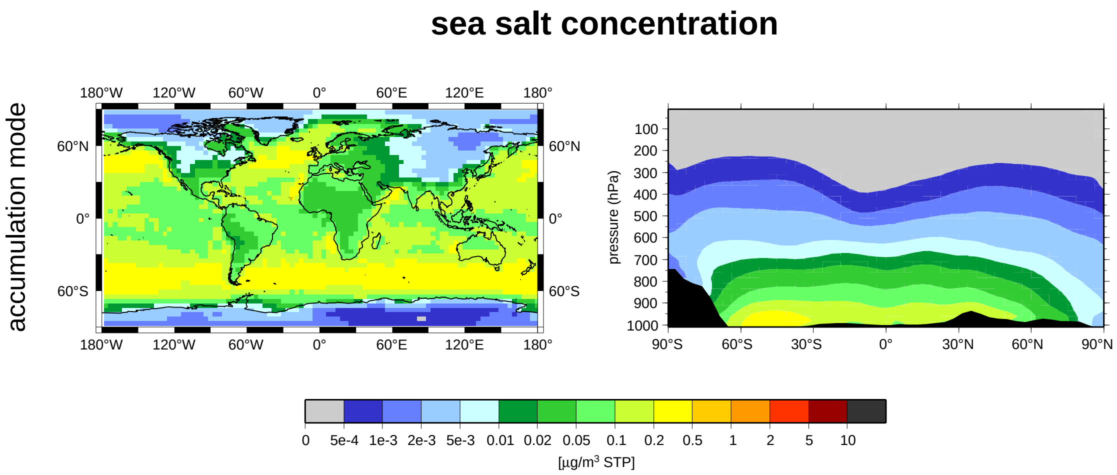


Fig. 4. As Fig. 1, but for sea salt (accumulation mode only).

If no excess ammonia is left, the relative contribution of ammonium decreases again (6.7% in the accumulation mode at 500 hPa and 4.9% at 250 hPa). The increasing sulfate fraction corresponds with increasing acidity of the aerosol. Aerosol nitrate plays only a minor role on global annual average. The contribution in the near surface layer is rather low (about 1% in the accumulation mode) and further decreases with height.

Also the average composition of Aitken mode particles in the lowermost boundary layer is controlled by the emissions at the surface. Here, the composition is dominated by black carbon (70.5%), followed by sulfate (15.9%), organic matter (6.7%), ammonium (5.5%), and nitrate (1.4%). The very high fraction of BC is caused by fossil fuel combustion. It is assumed that particles emitted by fossil fuel combustion mostly have sizes in the range of the Aitken mode (for details and references see Lauer et al. (2005)). As for accumulation mode particles, the sulfate fraction increases with height as a consequence of sulfate production due to oxidation of SO_2 and other precursors.

This increase in relative contribution of SO_4 to the total particle mass in the Aitken mode with height is much stronger than in the accumulation mode. Condensation is proportional to the surface area of the pre-existing particles. Since Aitken particles are smaller than accumulation mode particles, they show a larger surface area to particle volume ratio, which facilitates a more efficient increase in relative contribution of SO_4 due to condensation than in the case of larger particles. The average total mass concentration of Aitken mode particles decreases from 57 $\mathrm{ng/m^3}$ at 1000 hPa to 1 $\mathrm{ng/m^3}$ (STP conditions) at 250 hPa. The contribution of the Aitken mode to the total mass of submicrometer particles decreases from 1.2% in the boundary layer to about 0.1% in the tropopause region.

3.3 Atmospheric residence time and global burden

The average atmospheric residence time of each aerosol component can easily be estimated from Eq. (1) assuming steady state of sources and sinks:

$$\text{residence time [days]} = \frac{\text{burden [Tg]}}{\sum \text{sources [Tg/day]}} \qquad (1)$$

The main component of submicrometer aerosols is sulfate with an average burden of 2.25 Tg, followed by organic matter (1.77 Tg), mineral dust (0.83 Tg), ammonium (0.46 Tg), and black carbon (0.26 Tg) (Table 1). Sea salt (0.08 Tg) and nitrate (0.02 Tg) are only of minor importance for the average global burden of submicrometer particles. The simulated burdens of the aerosol compounds dominated by submicrometer particles are within the range spanned by the results of other model studies (Table 2). For the aerosol components mineral dust and sea salt, most of the mass is associated with coarse particles, which are not included in this model study. Thus, it is not reasonable to compare the atmospheric burden of the submicron fraction of these two components with the total burdens simulated in other model studies. However, for mineral dust Tegen and Lacis (1996) give size-dependent global average mass loadings, which amount to 0.2 $\mathrm{mg/m^2}$ for the radius range 0.1–0.18 µm, 1.5 $\mathrm{mg/m^2}$ for the radius range 0.18–0.3 µm, and 4.2 $\mathrm{mg/m^2}$ for the radius range 0.3–0.6 µm. These mass loadings correspond to global burdens of 0.10 Tg, 0.77 Tg, and 2.14 Tg respectively. According to the size of the mineral dust particles emitted in ECHAM4/MADE (d=0.14 µm, σ=1.95), only the two smallest size-ranges reported by Tegen and Lacis (1996) seem appropriate for comparison. Summing up these two size intervals of particles smaller than 0.6 µm in diameter results in a burden of 0.87 Tg. This value compares well to the ECHAM4/MADE burden for accumulation mode mineral dust (0.83 Tg).

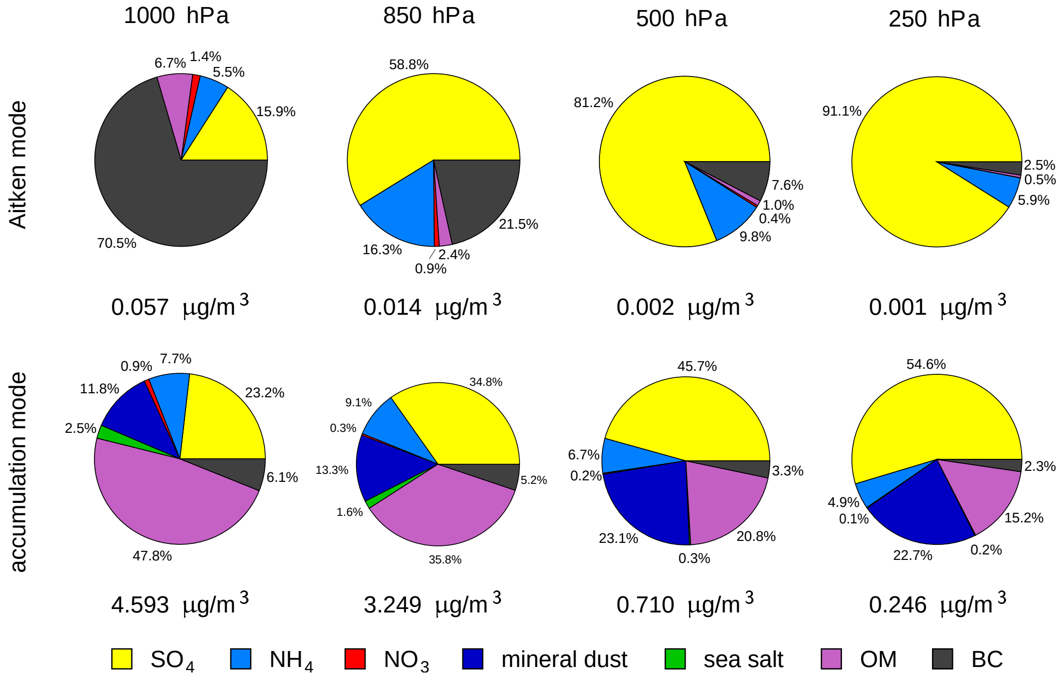


Fig. 5. Average composition and total mass concentration (STP conditions) of the global (dry) aerosol of the Aitken and accumulation mode for the pressure levels 1000 hPa, 850 hPa, 500 hPa and 250 hPa.

Table 1. Average global atmospheric burden and residence time (Eq. 1) of the submicrometer aerosol components calculated by ECHAM4/MADE for the Aitken mode, the accumulation mode and the total submicrometer aerosol.

component	atmospheric burden [Tg]			average residence time [days]		
	Aitken mode	accumulation mode	total	Aitken mode	accumulation mode	total
SO_4	0.02	2.23	2.25	1.7	4.6	4.5
NH_4	0.003	0.46	0.46	0.7	4.7	4.5
NO_3	<0.001	0.02	0.02	0.4	5.2	5.0
BC	0.01	0.25	0.26	0.4	8.9	6.6
OM	0.001	1.77	1.77	0.5	6.6	6.5
sea salt	–	0.08	0.08	–	1.6	1.6
mineral dust	–	0.83	0.83	–	18.0	18.0

Mineral dust has the longest residence time of all aerosol components in the accumulation mode (18 d), followed by accumulation mode black carbon (8.9 d), organic matter (6.6 d), sulfate, ammonium, and nitrate (4.6–5.2 d) and sea salt (1.6 d) (Table 1). The longer residence time of mineral dust in the accumulation mode, BC, and OM is caused by the reduced scavenging efficiency of these particles by cloud droplets due to partly hydrophobic properties. The short residence time of sea salt is caused by an efficient wet scavenging due to large particle sizes and high solubility (for details, see Lauer et al. (2005)). On average, the atmospheric residence time of Aitken mode particles is shorter than of accumulation mode particles. It amounts to around 0.5 d for Aitken mode NH_4, NO_3, BC and OM and about 1.7 d for Aitken mode SO_4. This reduced residence time is caused by additional sinks such as intermodal coagulation or growth of Aitken particles into the size-range of the accumulation mode as well as a very effective dry deposition of Aitken mode particles. The average residence time of the total particle mass (sum of Aitken and accumulation mode) simulated by ECHAM4/MADE is also in the range spanned by the results of other model studies. This is summarized in Table 2.

3.4 Particle number concentration

In contrast to many current climate models, ECHAM4/MADE calculates particle number concentration and particle size-distribution. This allows a more detailed view on the global aerosol characteristics than

Table 2. Atmospheric burden and residence time for the aerosol compounds SO_4, NH_4, NO_3, BC and OM from various global aerosol modeling studies.

component	atmospheric burden [Tg]	residence time [days]	reference
SO_4	2.09		Adams et al. (1999)
	2.31	6.5	Adams and Seinfeld (2002)
	1.89	5.8	Chin et al. (2000)
	1.71	4.3	Feichter et al. (1996)
	3.09		Lohmann et al. (1999b)
	2.4	3.9	Stier et al. (2005)
	2.11	4.2	Textor et al. (2006), AeroCom Exp. A (8 model mean)
	2.0	4.1	Textor et al. (2006), AeroCom Exp. B (8 model mean)
	2.25	4.5	this study
NH_4	0.39	4.2	Adams et al. (1999)
	0.46	4.5	this study
NO_3	0.13		Adams et al. (1999)
	0.02	5.0	this study
BC	0.22	6.4	Chung and Seinfeld (2002)
	0.28	6–10	Cooke and Wilson (1996)
	0.13	4–4.5	Liousse et al. (1996)
	0.26	6.8	Lohmann et al. (1999a)
	0.11	5.4	Stier et al. (2005)
	0.25	7.8	Textor et al. (2006), AeroCom Exp. A (8 model mean)
	0.16	7.3	Textor et al. (2006), AeroCom Exp. B (8 model mean)
	0.26	6.6	this study
OM	1.39	5.3	Chung and Seinfeld (2002)
	1.87	5.1	Lohmann et al. (1999a)
	0.99	5.4	Stier et al. (2005)
	1.71	7.0	Textor et al. (2006), AeroCom Exp. A (8 model mean)
	1.21	6.7	Textor et al. (2006), AeroCom Exp. B (8 model mean)
	1.77	6.5	this study

most previous studies, in which particle number concentration had to be calculated diagnostically using prescribed size-distributions.

Figure 6 shows the climatological annual mean particle number concentration simulated for the two size-ranges Aitken and accumulation mode. The geographical distribution in the near-surface layer follows the emission-patterns of primary particles and particle precursors (SO_2), resulting in regions of high particle number concentrations due to fossil fuel combustion (Central Europe, Southeast Asia, eastern USA) and biomass burning (Central Africa, South America). There are no significant sources of primary particles at the surface south of 30°S other than sea salt.

The largest emission rates of primary particles are usually found over the continents. These include emissions of BC and OM by industry, traffic, burning of domestic fuel and biomass burning as well as emission of mineral dust over the continental deserts. In contrast, sea salt particles are the only important primary particles emitted over the oceans (not taking into account emissions from international shipping). Nevertheless, due to its large particle sizes, sea salt shows comparatively small particle number concentrations.

The highest particle number concentrations in the Aitken mode are simulated for Central Europe, Southeast Asia and the eastern United States. Here, typical annual mean number concentrations of Aitken mode particles range between 10,000 and 50,000 particles per cm^3. Above the central regions of North and South America, typical annual mean particle number concentrations between 2000 and 5000 cm^{-3} are simulated. The lowest particle numbers in the Aitken mode are obtained above the oceans, far away from any strong source of primary particles. Here, particle composition is dominated by sea salt and typical annual mean number concentrations do not exceed 100 particles per cm^3. The vertical structure of the Aitken mode particle number concentration simulated for latitudes north of 30° S shows decreasing concentrations from the surface up to about 700 to 600 hPa, where the minimum particle number concentrations are reached. This reduction of particle number is mainly caused by coagulation (particularly intermodal coagulation), which is much more effective than wet deposition of Aitken mode particles. The formation of new particles by nucleation is very weak in this altitude range, resulting in a net depletion of particle number. There are no significant sources of

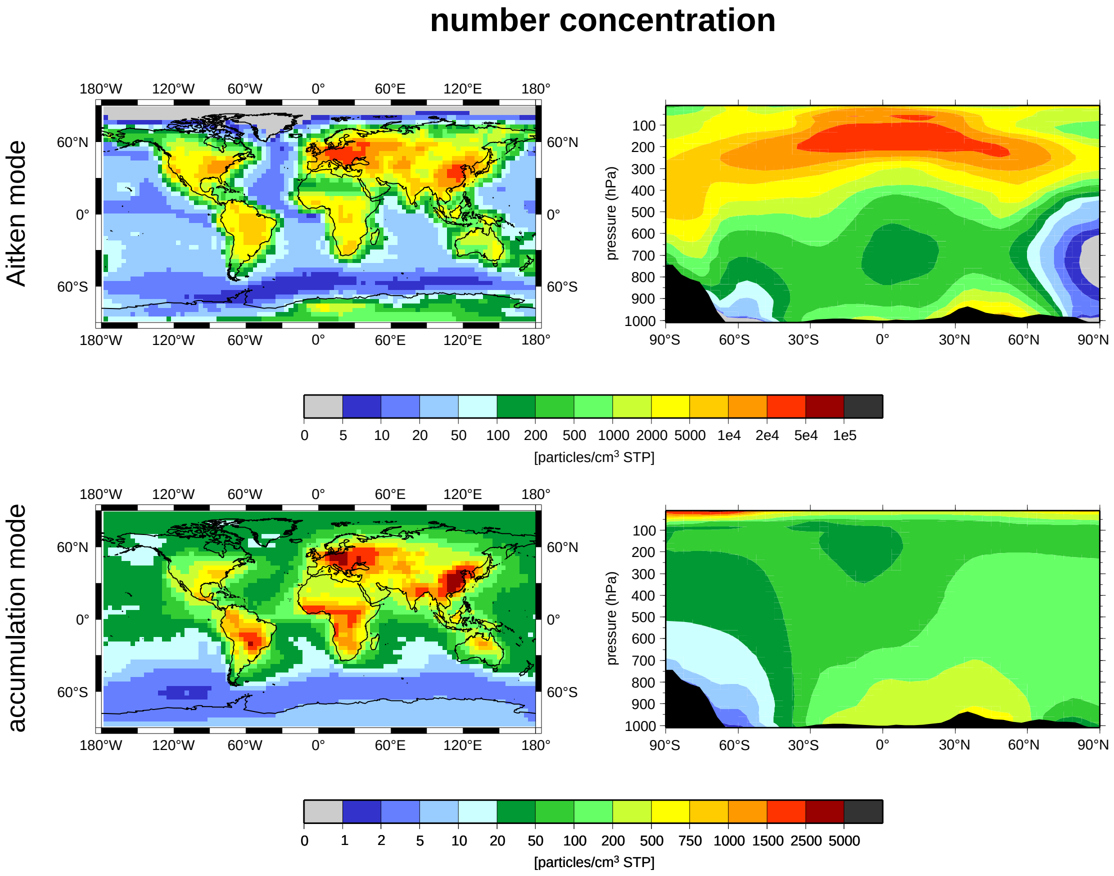


Fig. 6. As Fig. 1, but for total particle number concentration.

primary particles at the surface south of 30° S. This results in very low number concentrations of Aitken mode particles in this region in the lower troposphere. Above 600 hPa, particle number concentrations increase rapidly with altitude at all latitudes. Maximum particle numbers are found in the tropopause region, where zonally averaged annual mean concentrations range between 20 000 and 50 000 particles per cm^3 (STP). This strong increase in particle number concentration is caused by nucleation of sulfuric acid vapor. The low ambient temperatures and the low aerosol surface area available for condensation (which competes with nucleation for the condensable gas) favors efficient nucleation. This basic vertical structure of the mean number concentration of small particles, showing highest number concentrations in the tropopause region and only small number concentrations near the surface, is also supported by other recent model studies (e.g. Stier et al., 2005) and measurements (e.g. Clarke and Kapustin, 2002).

The most important source for accumulation mode particle number concentration is the emission of primary particles at the surface (see Sect. 4.1). Thus, the highest annual mean number concentrations of particles in the accumulation mode are simulated for the areas around the continental source regions of primary particles in Central Europe, Southeast Asia, the eastern United States and central regions of South America and Africa. Typical values of 1000 to 2500 cm^{-3} are simulated for these regions. In Central Europe and Southeast Asia the concentrations even exceed 2500 cm^{-3}. The simulation reveals that emissions of BC and OM by fossil fuel combustion are the dominant sources in the highly industrialized countries of the northern hemisphere. Emissions of BC and OM by biomass burning are dominant in South America and Africa. Because of their large sizes, mineral dust and sea salt particles are of minor importance for particle number concentration, even though they might contribute significantly to the total particle mass. For the continental regions not discussed above, typical annual mean near-surface particle number concentrations around 100 to 500 cm^{-3} are simulated. Above the oceans in the northern hemisphere, the simulation shows typical particle numbers of about 20 cm^{-3}. These northern hemispheric oceanic concentrations are slightly larger than typical accumulation mode concentrations simulated above the oceans in the southern hemisphere ($<$20 cm^{-3}) and Antarctica ($<$10 cm^{-3}). The zonally averaged vertical profile shows maximum annual mean accumulation mode particle number concentrations in

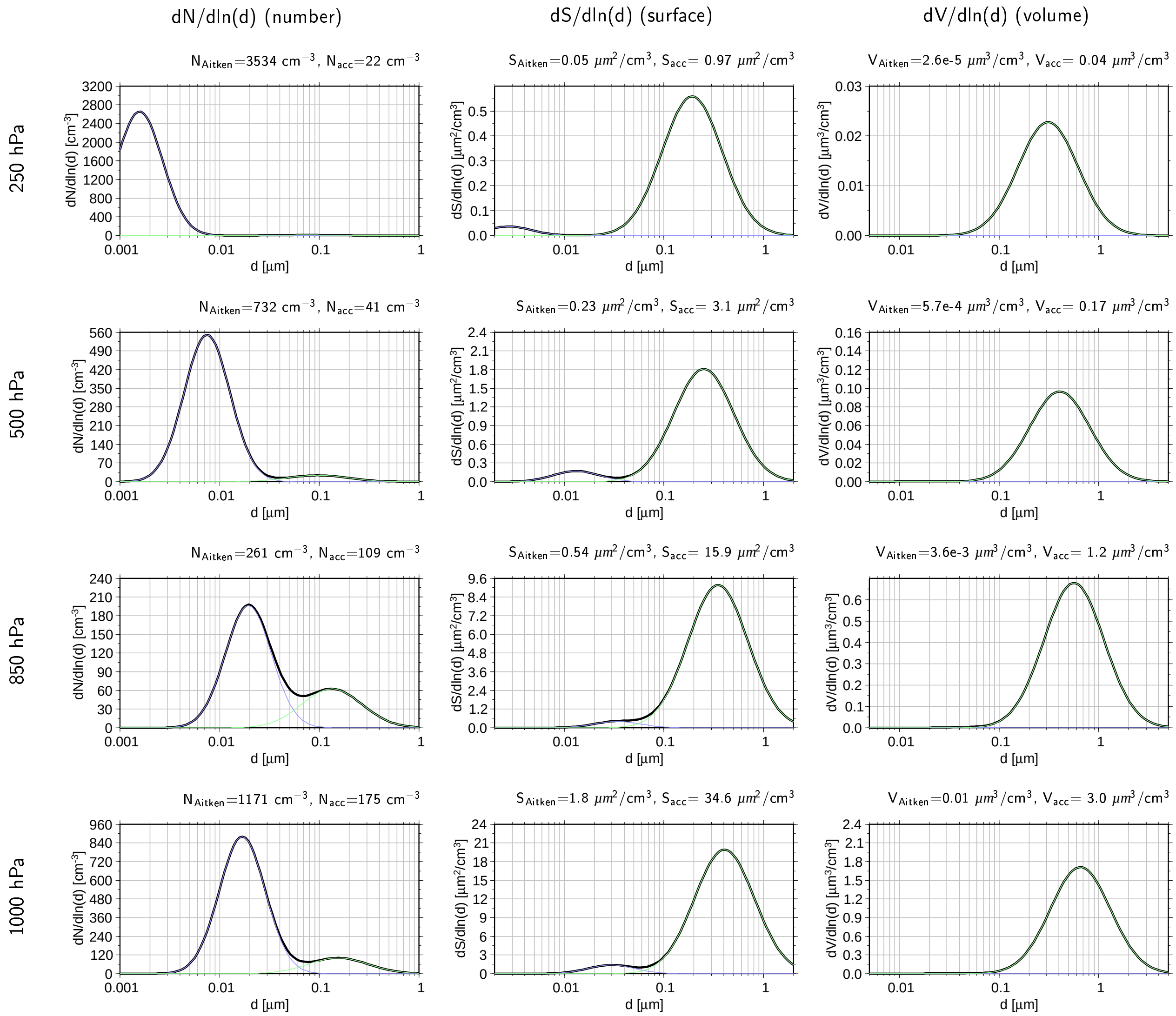


Fig. 7. Climatological annual means of the global average size distributions (lognormal) of particle number, dry surface area and dry volume for the pressure levels 1000 hPa, 850 hPa, 500 hPa and 250 hPa. The Aitken mode is drawn in blue, the accumulation mode in green and the total size-distribution in black. Total number concentration (N), surface area (S), and volume (V) of the Aitken and accumulation mode (acc) are listed above the individual plots. The concentrations shown refer to ambient (not STP) conditions.

the boundary layer above the continents north of 30° S. Due to weak sources at the surface, accumulation mode particle number concentrations are very low (below 10 cm^{-3}) in the boundary layer south of about 45° S. With increasing height, accumulation mode particle number concentration decreases at all latitudes north of 30° S. With the exception of a minor source resulting from Aitken mode particles growing into the accumulation mode size-range, no further sources of accumulation mode particle number exist but sources at the surface. Overall, this results in a net loss of particle number due to wet deposition and coagulation with increasing distance (height) to the surface sources.

3.5 Size-distribution

Figure 7 shows the climatological annual mean of the globally averaged particle number, dry particle surface and dry particle volume size-distribution simulated for different altitudes. The figure presents results obtained for the lowest model layer (≈1000 hPa), for about 1.5 km altitude (≈850 hPa), as well as the middle (≈500 hPa) and upper troposphere (≈250 hPa).

Near the surface, the simulated size-distribution is strongly influenced by the prescribed size-distributions of primary particles emitted. The median diameters of the Aitken and the accumulation mode are about 17 nm and 155 nm respectively. The ratio of total particle number of Aitken and accumulation mode is about 7:1. The ratio amounts to 1:19 in the case of surface area and 1:285 in the case of total volume.

Compared to the near surface conditions, the median diameter of the Aitken mode simulated for 850 hPa is slightly larger (19 nm). This is caused by the growth of Aitken particles due to intramodal coagulation and condensation of sulfuric acid vapor occurring during vertical transport from the surface to upper levels. The increase of the Aitken mode median diameter is also caused by intermodal coagulation and dry deposition which both are significant sink processes for Aitken mode particles. The efficiencies of both processes increase with decreasing particle size which results in a reduced residence time of smaller Aitken particles. Thus, the probability of larger Aitken particles to be transported upwards is higher than for smaller particles. In contrast to the Aitken mode, the average median diameter of the accumulation mode (about 134 nm) at 850 hPa is smaller compared to the 1000 hPa level. This is caused in particular by wet deposition, which removes large particles more efficiently than smaller ones. Another reason can be the transformation of growing Aitken particles into the accumulation mode, which also benefits the decrease in accumulation mode median diameter. The efficiencies of processes such as condensational growth or coagulation of accumulation mode particles causing an increase of particle size, are exceeded in particular by wet deposition efficiency, resulting in an overall decrease of the modal median diameter. Due to the sink processes described, particle number concentrations in both modes are smaller at 850 hPa than in the boundary layer (1171 cm^{-3} vs. 261 cm^{-3} in the Aitken mode and 175 cm^{-3} vs. 109 cm^{-3} in the accumulation mode). The ratio of particle numbers of Aitken and accumulation mode is 2.4:1 at 850 hPa. The corresponding ratios of total surface area and volume concentration amount to 1:29 and 1:332.

In the middle troposphere (500 hPa), the average median diameter of the simulated accumulation mode amounts to only 96 nm, which is considerably smaller than the values simulated for lower altitudes. As discussed above, an important reason for this reduction is that the efficiency of wet deposition increases with particle size. The resulting reduction in residence time of larger particles counteracts an efficient upward transport. Compared to the lower troposphere, the median diameter of the modeled Aitken mode is smaller, too. It amounts to only 7.5 nm at 500 hPa. Since the nucleation activity increases with altitude and the newly formed nucleation particles are very small (wet diameter 3.5 nm (Lauer et al., 2005)), the total size-distribution of the Aitken mode is shifted towards smaller median diameters and the Aitken particle number concentration increases. The number ratio of Aitken and accumulation mode is about 18:1. The corresponding surface and volume ratios amount to 1:14 and to 1:295, respectively.

The average median diameters of both modes further decrease with altitude. In the upper troposphere (250 hPa), they amount to only 1.6 nm and 74 nm in the case of the Aitken and accumulation mode, respectively. Particle number concentration in the Aitken mode is larger than simulated for lower altitudes. The ratio of particle number in Aitken and accumulation mode in the upper troposphere amounts to 162:1, which is about one order of magnitude higher than in the middle troposphere. This is mainly caused by strong nucleation, which reaches its maximum activity in the tropopause region. The ratio of the modal surface area concentrations of Aitken and accumulation mode amounts to 1:20 at 250 hPa. The corresponding ratio of the modal volumes is 1:1540. In case of strong nucleation, the formation of new very small particles might lead to an underestimation of the median diameter of the Aitken mode compared to observations. This has been identified as a principle limitation of the bimodal representation of the submicrometer aerosol as discussed in Lauer et al. (2005). Thus, the modal median diameter of the Aitken mode should be regarded as a lower limit rather than a typical value for this altitude range. Future modifications of the model towards the representation of a trimodal size distribution including separated Aitken and nucleation modes will enable a more sufficient representation of the upper tropospheric aerosol.

4 The role of aerosol microphysics on the global scale

To assess the relevance of aerosol microphysics (i.e., nucleation and condensation of gaseous precursors, coagulation of particles) for the simulation of submicrometer particles on the global scale, the roles of individual sources and sinks in the mean budgets of different aerosol compounds and particle number are analyzed for both aerosol modes considered. The specific role of the individual processes can be easily judged from the intercomparison of the strength of the different sources and sinks. To realize this, we saved the actual concentration changes induced by each individual process considered in the model. These changes can be integrated over time to calculate the total change and, hence, the corresponding contributions of the individual processes to the global budgets. This technique offers the possibility to gain new insights in the role of individual microphysical processes in the global aerosol cycles. Such detailed insights cannot be gained by observations alone, as measurements spanning the whole globe, covering at least one full seasonal cycle, and resolving each individual process would be required to obtain balanced data on all relevant sources and sinks. Such measurements are currently not available. Hence, various model studies have addressed the contribution of different sources and sinks to the global aerosol burden in the past (e.g. Feichter et al., 1996; Chin et al., 2000; Stier et al., 2005). The budgets provided by these studies considered aerosol mass cycles, but do not resolve particle number, size-dependencies, and aerosol microphysical processes. The first steps have been done investigating sources and sinks of particle number concentration including aerosol

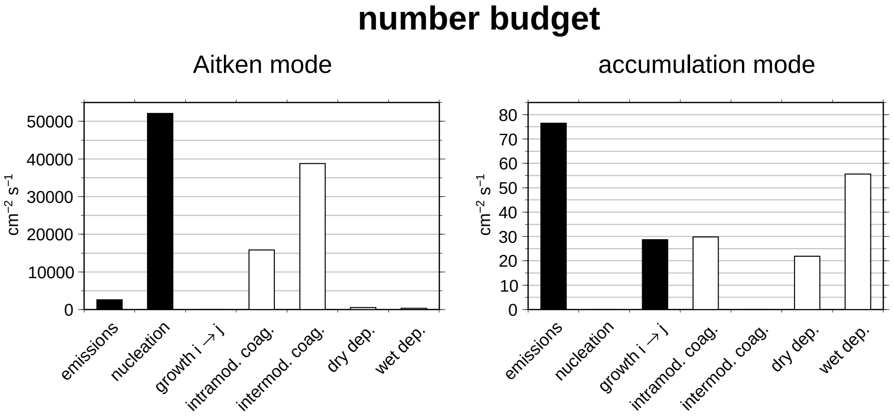


Fig. 8. Contributions of different processes to the global aerosol particle number budget. Mean column changes (particles cm^{-2} s^{-1}) due to sources (black) and sinks (white) calculated from a 10-year model simulation with ECHAM4/MADE are presented for Aitken mode (left) and accumulation mode (right) particles.

microphysics by Adams and Seinfeld (2002). However, their study is limited to pure sulfate aerosols only. The present study addresses both aerosol mass concentrations and particle number concentrations. Different particle size-ranges are analyzed separately taking into account aerosol microphysical processes. Budgets of particle number concentrations as well as of the masses of sulfate (secondary aerosol) and black carbon (exemplary for primary aerosol) are investigated.

4.1 Particle number

The emission of primary particles is a source for both modes in the model, whereas new particle formation by nucleation of sulfuric acid vapor is a source for the Aitken mode only. The growth of Aitken mode particles into the size-range of the accumulation mode acts as sink of the Aitken mode particle number and as source of accumulation mode particles. The following sinks reduce particle number concentration in the model: intramodal coagulation (coagulation of particles within the same mode), intermodal coagulation (coagulation of particles from different modes; reduces Aitken mode particle number concentration here), as well as dry and wet deposition. Figure 8 depicts the global climatological annual mean, vertically integrated changes in particle number induced by the individual sources and sinks as calculated from the 10-year model simulation with ECHAM4/MADE.

On global annual average, the most important source of Aitken mode particles in the simulation is the formation of new particles by nucleation of sulfuric acid vapor in the tropopause region. Since a high production rate of sulfuric acid vapor in the lower troposphere usually coincides with a high particle surface concentration, condensation prevents efficient nucleation in the model. However, due to the coarse spatial and temporal resolution of the model, potentially occurring local nucleation bursts cannot be resolved resulting in a potential underestimation of nucleation at these altitudes. It should also be noted, that comparisons with observations show, that the model tends to underestimate particle sizes in the tropopause region, where the particle formation rate due to nucleation reaches its maximum (Lauer et al., 2005). This might indicate that the newly formed particles grow too slowly or that nucleation might be generally overestimated. Nevertheless, we do expect nucleation being the most important source of new particles in the size-range of the Aitken mode, since the emission of primary particles at the surface does not contribute very much to the total sources although emissions dominate near the surface. Since the Aitken mode generally dominates particle number concentration, nucleation in the tropopause region should be the most important source of particle number concentration in terms of the global mean. This finding agrees with Adams and Seinfeld (2002), who also found that nucleation is the dominant source of particle number on global annual average. In contrast, emission of primary particles is the dominating source for accumulation mode particles. In addition, also the growth of Aitken mode particles into the size-range of the accumulation mode ("growth i→j") provides an important contribution to the global sources of accumulation mode particles. Due to the comparatively high particle number concentration of the Aitken mode, the corresponding contribution to the sinks of Aitken mode particle number is negligibly small.

While wet deposition is the most important sink for accumulation mode particle number, coagulation is the dominant sink for Aitken mode particles. Here, intermodal coagulation is more effective than intramodal coagulation. Dry and wet deposition have only a small impact on particle number concentration in the Aitken mode. Coagulation (intramodal) is an important sink for accumulation mode particle number, too. Besides this, also the removal of particles by dry deposition is relevant for the accumulation mode. Again, this is in accordance with the study of Adams and Seinfeld (2002), who also found that coagulation is the dominant sink of particle number concentration on global annual average.

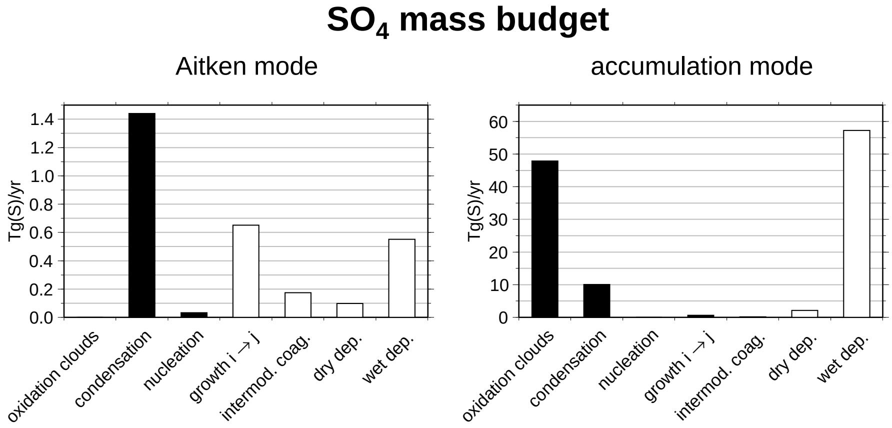


Fig. 9. As Fig. 8, but for the aerosol sulfate mass budget.

4.2 Sulfate mass

For the investigation of the budgets of particle mass concentration further processes such as condensation or sulfate production in cloud droplets, need to be analyzed, which change the mass concentration, whereas the number concentration remains unchanged. To assess differences between primary and secondary aerosol components, the budgets are not presented for total mass, but for sulfate and black carbon, which are representative for both classes of aerosol components in the model. Figure 9 shows the budgets of Aitken and accumulation mode sulfate for all individual sources and sinks.

Source processes of the secondary aerosol component sulfate are condensation of sulfuric acid vapor (both modes), formation of new particles by nucleation (Aitken mode only) and sulfate production in cloud droplets by oxidation of SO_2 (accumulation mode only). Most of the Aitken mode sulfate is produced by condensation of H_2SO_4. Nucleation contributes only about 2% to the sources of Aitken mode sulfate. In the accumulation mode, sulfate production by oxidation of SO_2 in cloud droplets (82%) dominates all other sources. In-cloud production contributes about 81% to the total sulfate mass (sum of both modes). This is within the range spanned by the results of other model studies assessing the global atmospheric sulfate budget. These studies suggest a contribution of in-cloud oxidation of SO_2 to the total SO_4 sources of, for instance, 77% (Berglen et al., 2004), 64%–83% (Chin et al., 2000), 55% (Koch et al., 2006), and 90% (Takemura et al., 2000). The aerosol model comparison initiative AeroCom calculated an average contribution of chemical sulfate production in the aqueous phase to the total SO_4 sources from 11 global models of 74% (Textor et al., 2006). In the present study, the contribution of condensation to the sources of accumulation mode mass amounts to about 17%. The corresponding contribution of the growth of Aitken particles into the accumulation mode size-range is negligibly small.

In both modes, sulfate is removed by wet and dry deposition. Intermodal coagulation and the growth of Aitken mode particles into the accumulation mode act as sinks for sulfate in the Aitken mode size-range. Wet deposition is the most important sink of accumulation mode sulfate, whereas dry deposition is only of minor importance. In the Aitken mode, besides wet deposition, also the loss of sulfate mass due to particle growth into the accumulation mode size-range is of major importance. But also coagulation and dry deposition must not be neglected when considering the sinks of Aitken mode sulfate. Considering total sulfate mass (sum of Aitken and accumulation mode), wet (dry) deposition contributes about 96% (4%) to the total sinks of sulfate. This is at the high (low) end of the range given by other model studies for the contribution of wet (dry) deposition to the sinks of sulfate: 85% (15%) (Berglen et al., 2004), 80%–93% (20%–7%) (Chin et al., 2000), 89% (11%) (Koch et al., 2006), 88% (12%) (Takemura et al., 2000), and 95% (5%) Stier et al. (2005). The AeroCom multi model mean suggests a contribution of wet (dry) deposition of 89% (11%) to the total sinks of SO_4 (Textor et al., 2006).

Thus, aerosol microphysics are only of minor importance for the simulation of accumulation mode sulfate burden on the global scale, but are essential for simulating sulfate mass concentration in the size-range of the Aitken mode.

4.3 Black carbon mass

Figure 10 shows the contributions of the individual sources and sinks to the modeled climatological global mean black carbon mass budget.

Emissions are the only source of BC in the Aitken mode size-range. The simulations suggest that emissions of primary particles constitute also the main source (74%) of accumulation mode BC, whereas the growth of Aitken particles into the accumulation mode and intermodal coagulation contribute only 17% and 9%, respectively. For both modes, wet

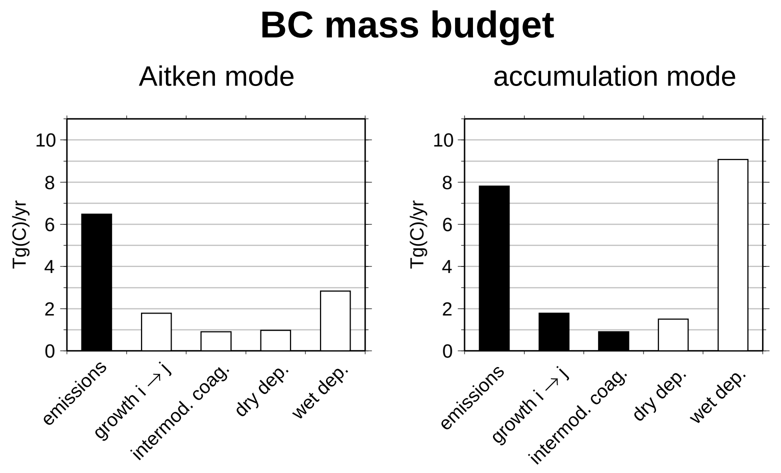


Fig. 10. As Fig. 8, but for the black carbon mass budget.

deposition is the most important sink for BC (Aitken mode: 44% of total sinks, accumulation mode: 86% of total sinks). The reduced efficiency of wet deposition compared to SO_4 results from the hydrophobic properties of the primary particles at emission. Only aged (hydrophilic) BC particles are subject to activation scavenging. The growth of Aitken particles into the accumulation mode (27%), intermodal coagulation (14%), and dry deposition (15%) have important contributions to the sinks of the Aitken mode BC. For the accumulation mode, dry deposition cannot be neglected, too (contribution about 14%). Considering the total BC mass budget (sum of Aitken and accumulation mode), the contribution of wet (dry) deposition to the total sinks calculated by ECHAM4/MADE amounts to 81% (19%). Stier et al. (2005) calculated a corresponding contribution of 92% (8%). Koch (2001) simulated contributions ranging from 58% to 63% (42% to 37%). Takemura et al. (2000) obtained 82% (18%), the AeroCom multi model mean (Textor et al., 2006) amounts to 78% (22%).

Overall, aerosol microphysics play an important role for the simulation of BC particles on the global scale, particularly for the Aitken mode. Although the budget of accumulation mode BC is dominated by emission of primary particles and wet and dry deposition, the contribution of aerosol microphysics should not be neglected.

4.4 Net production/depletion

In addition to the total budgets discussed in the previous section, the vertical distributions of the microphysical change rates (sum of sources and sinks discussed above) of particle number and mass are analyzed and the dominant processes are identified for different altitudes. For particle mass, we selected black carbon and sulfate as representative primary and secondary aerosol components, respectively. Figures 11, 12 and 13 depict the vertical distributions of the climatological zonal mean net change rates of particle number, sulfate and black carbon mass concentrations. Sources are considered as positive change rates, sinks as negative change rates. The results allow the identification of regions with net production and net depletion of particle number and sulfate or black carbon mass.

4.4.1 Particle number concentration

Net production of Aitken mode particles near the surface above the continents between about 45° S and 75° N is caused particularly by emission of primary particles from sources at the surface. The analysis of the contribution of the individual sources (not shown) reveals, that formation of new particles by nucleation is of minor importance in the near surface layer. Above this layer, intra- and intermodal coagulation effectively remove particles due to the high number concentrations resulting in a strong net depletion of particle number concentration in the Aitken mode. With increasing height this net depletion decreases as particle number concentration in the Aitken mode decreases up to altitudes around 600 hPa. Above 600 hPa, in particular in the tropopause region, very effective formation of new Aitken mode particles due to nucleation occurs. Just below this region of strong net production newly formed Aitken particles are efficiently removed by coagulation leading to a net depletion in the altitude range of about 300–400 hPa where coagulation is not compensated by nucleation.

In terms of annual means, efficient net production of accumulation mode particles takes place in the boundary layer only. In contrast to Aitken mode particles, strong emissions of accumulation mode particles lead to a net production even south of 45° S above the oceans. This is mainly caused by emissions of sea salt particles. The maximum net depletion of accumulation mode particle number takes place in the altitude range between 700 and 900 hPa. Here, the reduction

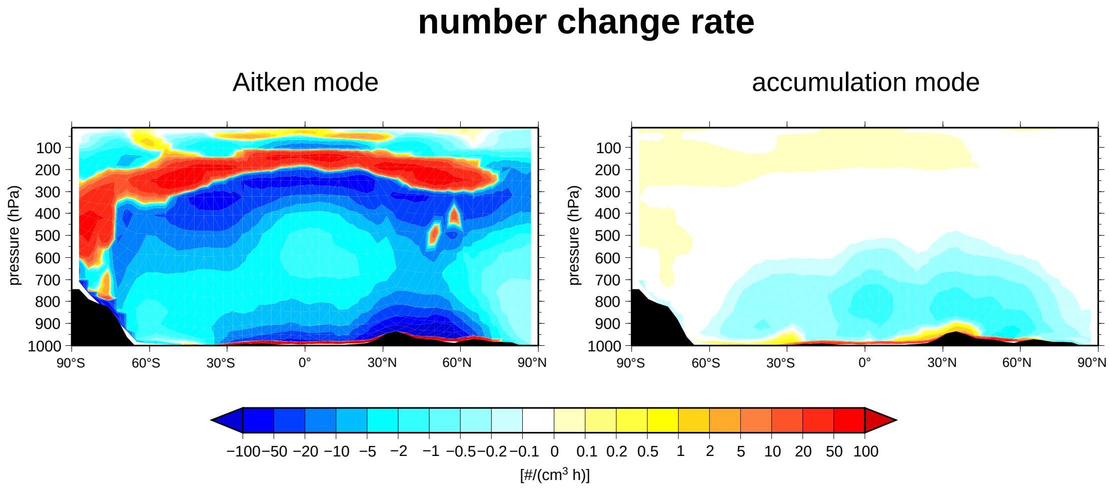


Fig. 11. Zonal mean annual averages of the net microphysical change rate (ambient values, not STP) of particle number in the Aitken (left) and the accumulation mode (right). Positive values (red/yellow) correspond to a net production of new particles, negative values (blue) to a net depletion of particles.

of particle number by wet deposition is most efficient. Only small net depletion occurs above about 600 hPa. Hence, in contrast to the Aitken mode, the particle number concentration of the accumulation mode experiences only very small changes due to microphysical processes at these altitudes. This results in a comparatively small altitudinal variation of the accumulation mode particle number concentration (see also Fig. 6).

4.4.2 Sulfate mass

Aitken mode sulfate is very efficiently removed by dry deposition at the surface resulting in a strong net depletion in the near surface layer (Fig. 12). In the lower troposphere above the surface layer, net production of sulfate takes place. In this region, large amounts of sulfuric acid vapor condense on the surfaces of the abundantly existing Aitken particles. In addition, a small net production of Aitken mode sulfate takes place in the tropopause region due to new particle formation by nucleation, which compensates the weak sinks in this altitude. In most other regions, a net depletion of Aitken mode sulfate occurs. The maximum of the net depletion occurs between about 700 and 900 hPa. Here the transfer of Aitken mode sulfate into the accumulation mode due to continued growth of the particles is very efficient.

Sulfate in the accumulation mode is produced particularly in the lower troposphere. Here, condensation of sulfuric acid vapor and sulfate production by oxidation of SO_2 in cloud droplets are most efficient. About 100 hPa above this layer of net production, net depletion reaches its maximum. The most important sink in this region is removal of sulfate by wet deposition. With increasing height, the net depletion decreases.

In the lower troposphere over Antarctica, a net production of accumulation mode sulfate is simulated, which is mainly caused by in-cloud oxidation of SO_2 during summer. The cloud formation in this region is driven by the meridional circulation called polar cell. SO_2 oxidized in cloud droplets is likely produced from DMS emitted from the ocean, in particular during Antarctic summer.

4.5 Black carbon mass

In contrast to sulfate, black carbon is released exclusively by emissions of primary particles at the surface. The net production of BC is limited to the boundary layer above the source regions (Fig. 13). In the Aitken mode, net depletion takes place in all layers above the lowermost model level, which decreases with height due to decreasing particle concentrations. The vertical structure of the accumulation mode change rate features a slightly thicker layer of net production in the lower boundary layer, which is related to emissions at the surface in the lowermost model level and Aitken particles growing into the size-range of the accumulation mode. In the tropics and in the northern mid-latitudes, strong net depletion takes place at about 750–800 hPa and about 800-950 hPa, respectively. These maximum net depletion rates are related to efficient wet deposition. Because of the lower scavenging efficiency of Aitken particles, these features are only prominent in the vertical distribution of the accumulation mode. With increasing height, the net depletion rates in the accumulation mode decrease due to decreasing particle concentrations.

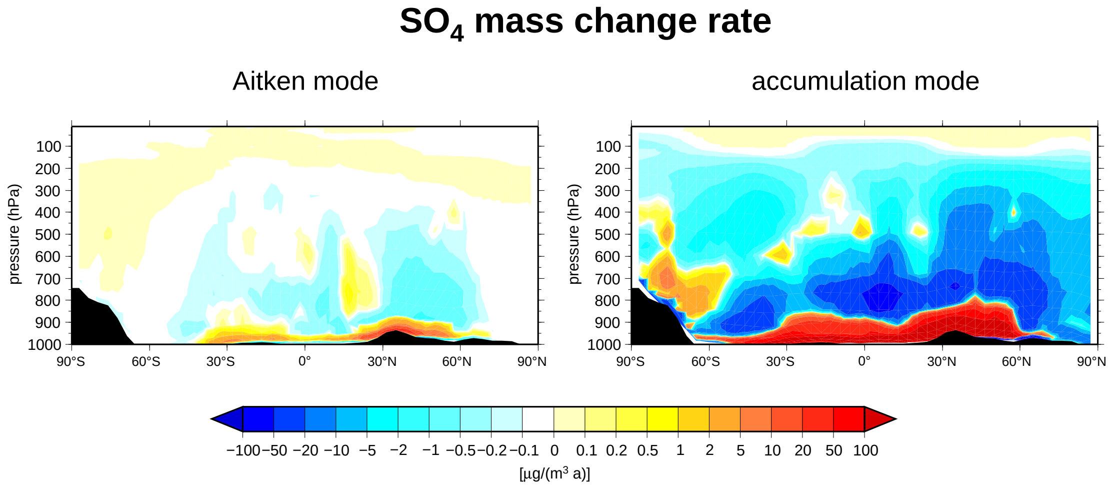


Fig. 12. As Fig. 11, but for sulfate mass concentration.

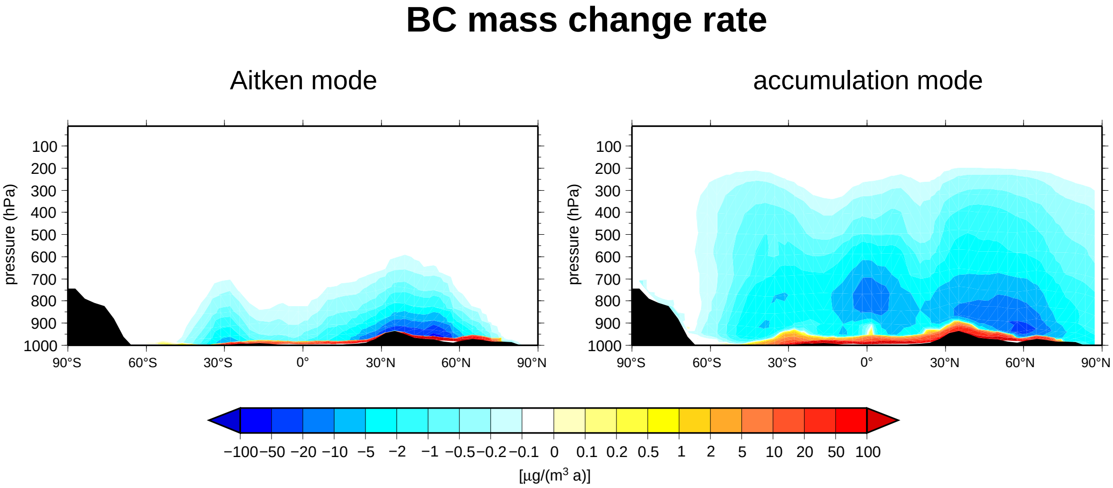


Fig. 13. As Fig. 11, but for black carbon mass concentration.

5 Conclusions

In this study, we analyzed a multiannual simulation of the global submicrometer aerosol performed with the new model system ECHAM4/MADE. Detailed aerosol physics such as coagulation, condensation and nucleation of sulfuric acid vapor, size-dependent wet and dry deposition and cloud processing of aerosols have been taken into account. Thus, in contrast to most current climate models using aerosol climatologies or simulating particle mass only, ECHAM4/MADE allows the calculation of the particle number concentration and aerosol size-distribution. This allows a more detailed insight into the global aerosol cycles than previous model studies. The climatological model data have been used to characterize the properties of submicrometer particles in terms of a global assessment. Aerosol properties such as chemical composition, number concentration, size-distribution and atmospheric residence time have been assessed. The model results reveal the following major features of the global submicrometer aerosol:

High aerosol mass and number concentrations are found in particular over the highly populated continental areas in the northern hemisphere where aerosol sources are dominated by emissions from fossil fuel combustion. In addition, high aerosol loadings occur over Africa and South America where biomass burning is the major source of submicrometer particles. Also mineral dust and sea salt particles have an important contribution to the aerosol mass loading of submicrometer particles. Nevertheless, due to large particle sizes, their impact on the number concentration of the submicrometer aerosol is comparably small. Overall, the simulated geographical patterns of aerosol mass corroborate the results of previous studies, including those considering aerosol mass only. The simulation of different particle size-ranges suggests, that predominantly accumulation mode particles are subject to long-range transport, whereas Aitken mode particles tend to reside near the source regions. This is caused by the shorter average residence time of Aitken mode particles compared to the accumulation mode.

At the Earth's surface, the aerosol-size-distribution is strongly influenced by the size of emitted primary particles. With increasing height, the mean size of the Aitken mode particles increases due to coagulation, whereas wet deposition reduces the mean size of the accumulation mode aerosols. In the middle and upper troposphere, the mean size of the Aitken mode aerosol decreases due to formation of small new particles by nucleation. On the global scale, nucleation is most efficient in the tropopause region. In the boundary layer, nucleation is of less importance.

In order to assess the role of aerosol microphysics (i.e., nucleation and condensation of precursor gases, coagulation of particles) in the global atmospheric cycles of the submicrometer aerosol, a detailed analysis of the contribution of different aerosol microphysical processes to the global sources and sinks of particulate matter has been performed. The analysis shows that aerosol microphysical processes are essential sources and sinks of particles on the global scale and are, therefore, of principal importance for the global simulation of aerosol particle number concentration and size distribution. For the simulation of total particle mass, aerosol microphysics turned out to be important but not vital. Aerosol microphysics contribute significantly to sources and sinks of Aitken mode particle mass. However, for the accumulation mode mass concentration, processes such as emissions of primary particles and wet deposition are dominant. Although wet deposition is size-dependent, aerosol microphysics showed not to be essential for the simulation of global accumulation mode particle mass burden. This is due to the fact that the larger accumulation mode particles which carry the major fraction of accumulation mode mass can easily be activated to cloud droplets and can therefore be removed by wet deposition. Nevertheless, the size-distribution information is required for a physically based description of activation scavenging and therefore for the description of accumulation mode particles processed by clouds and precipitation.

In the present study, the aerosol size-distribution is represented by lognormal modes with constant width. This is a widely used technique applied in global aerosol simulations. Nevertheless, this approach implies simplifications of complex aerosol size-distributions. It may cause uncertainties, for instance, in the case of fresh aerosol generated by nucleation or emissions which can perturb the size-distribution by well pronounced bursts. Inaccuracies may also occur in representing aerosol size-distributions perturbed by cloud processing. Nevertheless, the appropriateness of lognormal modes to represent observed aerosol size-distributions under many different atmospheric conditions has been documented in many publications (e.g. Whitby, 1978). Furthermore, severe perturbations of the aerosol size-distribution as discussed above frequently occur on scales smaller than those of current GCM. Hence we think that representing the aerosol size-distribution by a more detailed approach as, for instance, a sectional model would not fundamentally change the main findings of the present study.

We conclude from this study, that simulations of the global submicrometer aerosol should include aerosol microphysical processes whenever particle number concentrations and size-distributions are to be calculated. Since the climate effects of aerosols, e.g. due to absorption and scattering of solar radiation and their impacts on cloud formation are size-dependent, it is essential to calculate the size-distribution to make further progress for future predictions of climate change. This is also the case when assessing health effects of aerosols. On the other hand, if only particle mass concentrations are to be calculated, which are dominated by the accumulation and coarse modes, processes such as emissions, wet and dry deposition are most important. However, since also dry and wet deposition are size-dependent also the simulations of the global particle mass cycles will benefit from consideration of aerosol microphysics.

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