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Modelling of nitrate
particles

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Modelling of nitrate particles: importance of sea salt

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Abstract

A thermo dynamical model for treatment of gas/aerosol partitioning of semi volatile inorganic aerosols has been implemented in a global chemistry and aerosol transport model (Oslo CTM2). The sulphur cycle and sea salt particles have been implemented earlier in the Oslo CTM2 and the focus of this study is on whether nitrate particles are formed as fine mode ammonium nitrate or react on existing sea salt particles. The model results show that ammonium nitrate particles play a non-negligible role in the total aerosol composition in certain industrialized regions and therefore have a significant local radiative forcing. On a global scale the aerosol optical depth of ammonium nitrate is relatively small due to limited availability of ammonia and reaction with sea salt particles. Inclusion of sea salt in the calculations reduces the aerosol optical depth and burden of ammonium nitrate particles by 25% on a global scale but with large regional variations.

1 Introduction

The chemical composition of aerosols has been investigated strongly over the last decade (Jaffe et al., 2005; Lelieveld et al., 2002; Malm et al., 2004; Novakov et al., 1997; Park et al., 2004; Putaud et al., 2004; Ramanathan et al., 2001) and given significant improvements in the insight in their complex variations. The focus on anthropogenic effects from aerosols started with sulphate aerosols and thereafter carbonaceous aerosols. Inorganic nitrate aerosols constituent a non-negligible fraction of the total aerosol mass in industrialized regions (Adams and Seinfeld, 2002; Hueglin et al., 2005; Jaffe et al., 2005; Malm et al., 2004; Marinoni et al., 2005; Park et al., 2004; Putaud et al., 2004). Over Europe nitrate particles accounts for about 10–20% of the total aerosol mass (Putaud et al., 2004). Nitrate particles shows a substantial larger fraction in urban areas than in remote areas (Hueglin et al., 2005; Malm et al., 2004; Putaud et al., 2004). The measurements show also a significant seasonal variation in

the nitrate abundance (Hueglin et al., 2005; Jaffe et al., 2005). There are also indications that nitrate has increased over the last years or at least the relative importance of nitrate, based on deposition data (Evans et al., 2001; Sopauskiene et al., 2001; Stoddard et al., 1999). The large reduction of SO_2 over Europe and thus sulphate aerosols over Europe allows more ammonia to react with nitrate instead of sulphate (Myhre et al., 2002).

In the global aerosol intercomparison study AEROCOM (http://nansen.ipsl.jussieu.fr/AEROCOM/) altogether 16 aerosol models participate having the 5 major aerosol components included (sea salt, mineral dust, sulphate, black carbon, and organic carbon). A more difficult task than modelling these non volatile aerosol components in the atmosphere is to model semi-volatile aerosols, such as organics and nitrate aerosols. For nitrate aerosols some few model studies of their global distribution exists (Adams et al., 2001, 1999; Jacobson, 2001; Martin et al., 2004; Metzger et al., 2002). A large problem when modelling aerosol concentration of semi volatile compounds is that their saturation vapour pressure is dependent on aerosol composition and temperature. A further complicating factor is that aerosols are size distributed. It is therefore not sufficient to find equilibrium vapour pressures for semi volatile gases over a total aerosol mixture, but a model for semi volatile aerosols should accurately predict partition between different size classes and mass transfer through condensation to each aerosol size. Therefore, to model nitrate aerosol a detailed description of the tropospheric chemistry, sulphate, ammonia, and sea salt is important. The saturation vapour pressure of HNO_3 is also strongly dependent on temperature. The issue of the importance of sea salt for the size of the nitrate particles has been neglected in earlier studies. In global radiative forcing simulations of nitrate aerosol the studies of Adams et al. (2001), Jacobson (2001), Liao and Seinfeld (2005), Liao et al. (2004), van Dorland et al. (1997) found very different results (-0.22, -0.07, -0.16, -0.14, -0.03 Wm^{-2}, respectively). The difference can be illustrated by (Jacobson, 2001) placing more than 90% of his 0.38 Tg of nitrate in the coarse mode based on observation data (his Table 2b) whereas Adams et al. (2001) puts all of their 0.45 Tg of nitrate in the accumulation mode based

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on absence of a coarse mode in their model. These differences show that physically based estimates of the size distribution of aerosol nitrate are important for calculating its radiative forcing. These initial studies regard to chemical and particle microphysical treatment of nitrate aerosol shows that depending on whether nitrate is in the fine

5   or coarse mode of aerosols are important for whether nitrate aerosols have a negligible radiative forcing or not. In a recent and more detailed study regard to aerosol thermodynamics Martin et al. (2004) found that burden and radiative forcing of nitrate aerosols was about 10–15% of the sulphate aerosols. Martin et al. (2004) also take into account formation of solids. They find that the combined radiative forcing of the

10 nitrate/ammonia/sulfate aerosol changed by approximately 25% depending on whether the upper side or the lower side of the hysteresis loop controlling water uptake of the aerosol is used. In two other detailed studies (Liao and Seinfeld, 2005; Liao et al., 2004) sea salt was included in the simulations, but the effect of sea salt was only to some extent quantified. In their simulations sea salt over land may contribute to accu-

15 mulation type (fine mode) particles, since equilibrium calculations are made for sulfate, ammonium, nitrate, sea salt, and water in an internal mixture. Further in Liao and Seinfeld (2005) it was found that heterogeneous reaction on varies aerosol strengthen the radiative forcing by 33%, mainly as a result of lower sulphate amount.

In this study we use a global aerosol and chemistry transport model to calculate the

20 distribution of nitrate aerosols. Calculations of chemical equilibrium are used to partition ammonia and nitrate between the gas phase and the aerosol phase. A simple concept is introduced which allows estimations of the mass transfer between accumulation mode and coarse mode nitrate. The advantage with this type of modelling is that for the radiative forcing of nitrate it is the fine mode of this compound that has an impact

25 and thus a separation of fine and coarse mode nitrate particles are of importance.

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2 Methods

2.1 Aerosol model description

We use the Oslo CTM2 model to describe transport and photochemical reactions in the global atmosphere. This model is an off-line chemical transport model that uses pre-
5 calculated meteorological fields to simulate tracer distributions in the atmosphere. In this study forecast data from European Centre for Medium-Range Weather Forecasts (ECMWF) have been used. For this study the meteorological data for 2000 in T42 (2.8×2.8°) and 40 layers are used. The advection of chemical species is calculated by the second order moment method, which is able to maintain large gradients in the
10 distribution of species (Prather, 1986). Vertical mixing by convection is based on the surplus and deficit of mass flux in a column (Tiedtke, 1989). Turbulent mixing in the boundary layer is treated according to the Holtslag K-profile scheme (Holtslag et al., 1990).

The original chemistry scheme (Berntsen and Isaksen, 1997) calculates the mixing
15 ratios of 51 tracers in the global atmosphere including OH, Ozone, NO_x and hydrocarbons. The model has recently been expanded to include the sulphur cycle (Berglen et al., 2004). The sulphur scheme includes five species and is coupled with the oxidant chemistry in the global model. Sea salt is simulated using the scheme of Grini et al. (2002). Chemical equilibrium between the inorganic compounds is simulated us-
20 ing the model of Metzger et al. (2002). The model predicts gas/aerosol partitioning at chemical equilibrium in the NH_4^+/Na^+/SO_4^{2-}/NO_3^-/H_2O/Cl^- system. The aerosols are assumed to be metastable in our study, thus we do not take into account formation of solids. This assumption was tested by Metzger et al. (2002) who compared full hysteresis calculations to metastable calculations and found small differences between the
25 two calculations due to the fact that most aerosols actually were on the upper side of the hysteresis loop.

Emissions of ammonia follow Bouwman et al. (1997) where the most important sources for ammonia are excreta from domestic animals and use of synthetic fertil-

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izers. The total global emission for 1990 is estimated to 54 Tg N yr^{-1}. The emissions from biogenic sources are weighted by number of sunlight hours. Adams et al. (2001) propose to weight the emissions by number of sunlight hours to the power of four. Given the very strong seasonal cycle this would give, and that this anyway has to be
5 interpreted as tuning, we chose to only weight the emissions linearly. Emissions of SO$_2$ and NO$_x$ follow Berglen et al. (2004) and use anthropogenic emission with reference to year 1996 of 67 Tg S yr^{-1} and 34 Tg N yr^{-1}, respectively.

2.2 Implementation of nitrate aerosols

In global models of aerosol chemical equilibrium of semi volatile gases, mass transfer
10 has often been ignored since it is too expensive (Adams et al., 2001). However, when calculating aerosol nitrate, it has to be taken into account since nitrate can condense both on fine and coarse aerosols, and we need some estimation of this process. To simplify, we chose to represent the aerosol by a fine mode, controlled by sulphate, and a coarse mode controlled by sea salt, and sulphate and sea salt do not interact
15 through chemical equilibrium. From here on we will refer to the mode controlled by sulphate as "fine mode" and the mode controlled by sea salt as "coarse mode". This is a great simplification since sulphate can exist in coarse mode, and sea salt in fine mode. However, we chose to separate the aerosols this way to allow for the simple treatment of mass transfer introduced later. This solution is an intermediate solution
20 between treating all aerosols (independent of size) in the same equilibrium calculation and calculating detailed mass transfer through condensation to several size sections as done for example by Pilinis et al. (2000).

Gaseous HNO$_3$ and NH$_3$ can condense on any of the two modes. In each time step photochemistry is first solved and H$_2$SO$_4$ and HNO$_3$ are generated. All H$_2$SO$_4$
25 is assumed to exist in the fine mode. After photochemistry, the thermodynamic equilibrium is solved twice by the package of Metzger et al. (2002). First, the fine mode has a chance of reaching chemical equilibrium. This mode is solved first because

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the smaller particles reach equilibrium faster than the larger ones (e.g. Capaldo et al., 2000; Wexler and Seinfeld, 1990). This first equilibrium in principle neutralizes sulphuric acid with ammonium. Any excess ammonia will be able to condense on the small particles together with HNO_3 as NH_4NO_3. After the small particles are in equilibrium, equilibrium is calculated a second time. This is done for any excess of HNO_3 (and NH_3) to react with sea salt. HNO_3 reacts with sea salt following the equation

$$NaCl(aq) + HNO_3(g) \rightarrow NaNO_3(aq) + HCl(g) \tag{1}$$
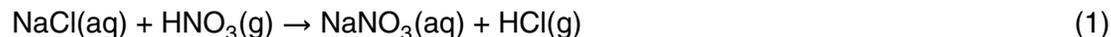
Thus sea salt depletes the HNO_3 gas concentrations. Our formulation avoids the formation of stable Na_2SO_4 salts followed by evaporation of HCl and change of particle acidity which could be the result if sea salt and sulphate would be treated in equilibrium together.

Fine mode NO_3^- is then controlled by the following factors:

– Production of HNO_3 in the photochemistry

– Availability of NH_3/NH_4 since HNO_3 will only condense if we have excess NH_4

– Temperature since the equilibrium vapour pressure of HNO_3 is only sufficiently low at low temperatures.

– Depletion of HNO_3 on the sea salt aerosols. $NaNO_3$ is assumed to be more stable than NaCl so that any excess HNO_3 available after fine mode equilibrium is lost to sea salt.

All aerosol species follow the loss processes of their "controlling species" in their aerosol mode, that is: All "fine mode" species have the same wet and dry deposition as sulphate (Berglen et al., 2004) whereas all coarse species have the same loss rates due to wet and dry deposition as the mass-averaged loss rate of sea salt (Grini et al., 2002).

Dry deposition of NH_3 follows Sorteberg and Hov (1996). Due to the large Henry's law coefficient of NH_3, NH_3 is taken up at the ground more easily than NH_4.

Washout of NH_3 is done according to it's Henry's law coefficient.

2.3 Optical properties of nitrate particles and radiative transfer calculations

The fine mode ammonium nitrate particles are assumed to have a lognormal size distribution with geometric radius of $0.05\,\mu m$ and standard deviation of 2.0. Refractive indexes are in lack of wavelength dependent available data assumed to be the same as for ammonium sulphate. The hygroscopic growth of ammonium nitrate is larger than for ammonium sulphate and is modelled according to Fitzgerald (1975). The coarse mode ammonium nitrate gives an additional mass since the molecular weight of NO_3^- is larger than the molecular weight of Cl^- (see Eq. 1). However, also the density, refractive indices, and hygroscopic growth changes with $NaNO_3$ instead of NaCl. To investigate the potential impact of coarse mode nitrate aerosols we have adopted density and refractive indices (scaled by information at one wavelength) for $NaNO_3$ (Lide, 1991) and NaCl. The hygroscopic growth for $NaNO_3$ is modelled based on Fitzgerald (1975).

The radiative transfer calculations are performed with a multi-stream model using the discrete ordinate method (Stamnes et al., 1988). In addition to the aerosols it is taken into account absorption by gases, Rayleigh scattering, and scattering of the clouds in the simulations (Myhre et al., 2002). The meteorological data used is the same as in the global aerosol and chemistry model.

3 Results

3.1 Surface concentrations

The annual mean surface concentration of fine mode ammonium nitrate particles are shown in Fig. 1. Maximum concentrations are found over China, with also high concentrations in other industrialized regions. Outside the industrialized regions the nitrate

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aerosol concentration at the surface is very low. In the Oslo CTM2 the difference in surface concentration compounds between the highly industrialized and background land areas or oceanic region is smaller for other aerosol than for ammonium nitrate particles. Nitrate particles have thus a larger difference in concentration from urban to rural and natural areas than other aerosols of industrial origin, which is in accordance with measurements (Putaud et al., 2004). Also in accordance with measurements (Hueglin et al., 2005; Jaffe et al., 2005) the seasonal variation show larger concentrations during the winter than during the summer due to the increase in saturation pressure of HNO_3 with temperature. The pattern of the surface concentration of fine mode nitrate particles is in very good agreement with observation distribution established in Malm et al. (2004) based on observations from 143 sites, except that the model has a very weak secondary maximum in the southern California. The general high values in the north east of USA with a maximum south of Chicago and the general low values in the north western USA are well reproduced. The magnitude of the modelled annual mean surface concentrations are also in good agreement with the measurements, note here that results in Malm et al. (2004) is given for NH_4NO_3. Over Europe surface concentration of fine mode nitrate of up to $2\,\mu g\ m^{-3}$ is modelled. In Putaud et al. (2004) results from urban and kerbsite locations shows mostly concentrations of 2–$3\,\mu g\ m^{-3}$ and in one case up to $8\,\mu g\ m^{-3}$. Note here that these results are point measurements inside large cities and with the global model with a resolution of 2–$3°$ are not able to reproduce these variations. Measurements in rural and natural site for nitrate are much lower than in the urban areas. The coarse mode surface nitrate (taken as difference between PM_{10} and $PM_{2.5}$ and correspond not directly to coarse mode) in Putaud et al. (2004) is mostly around $1\,\mu g\ m^{-3}$ in reasonable agreement with the model results (see Fig. 2).

The overall concentrations seem in good agreement with observations. A more detailed comparison could be of interested with specific stations. However, note that the large spatial variation in nitrate aerosols and the horizontal resolution in the state of the art global model of 2–$3°$ do not allow such detailed comparisons to be of large value.

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The distribution of the surface concentration of ammonium and sulphate (Fig. 3) has many similarities but differ mostly over remote oceanic region (due to DMS) and in regions with volcanoes. The pattern of these 2 surface concentrations has similarities to the fine mode ammonium nitrate surface concentration over the industrialized regions.

5 However, over ocean the model gives almost no ammonium nitrate, so all ammonium react with sulphate. Measurements over polluted areas in Europe (Putaud et al., 2004) shows ammonium surface concentration mostly around 2–3 μg m^{-3}, slightly larger than in the model. Whereas sulphate concentrations in the same polluted areas of typically 5–7 μg m^{-3} are found. A more detailed comparison is made in Berglen et al. (2004) for

10 sulphate with reasonably good agreement.

3.2 Vertical distribution of nitrate aerosols and limitations in production of nitrate aerosols

There are two necessary conditions for formation of fine mode aerosol nitrate in our model: 1) Temperature has to be low enough for HNO_3 to condense to the aerosol

15 phase, and 2) There has to be excess ammonia (i.e. NH_4+NH_3 has to be larger than twice the sulphate concentration – Seinfeld and Pandis, 1998). In the model, the first condition is satisfied in the free troposphere where temperatures are low. The second is most often satisfied near the ground. The reason for this is that sulphate is produced in the free troposphere through both aqueous and gaseous reactions whereas

20 the only source for NH_3/NH_4 is ground based emissions. Therefore, a condition for efficient production of fine mode aerosol NO_3 is efficient transport to the free troposphere. Adams et al. (2001) calculated 3 times higher burden of nitrate than their earlier study (Adams et al., 1999), and that the main part of the increase was in the free troposphere. The main difference between the two studies was 20% higher HNO_3 concentrations in

25 Adams et al. (2001). This indicates that their model is not limited by ammonia at high altitudes. In climate models with coarse vertical resolution, this is an expected result since the coarser resolution will make it more difficult to maintain strong gradient across the boundary layer. Figure 4 shows mixing ratios of zonal mean vertical profile of fine

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mode ammonium, total ammonia (NH_3+NH_4), sulphate, and fine mode nitrate. The figure reveals that these species have a quite different vertical distribution and that the vertical profile is a critical issue for formation of fine mode ammonium nitrate particles. Whereas the fine mode of ammonium particles decreases regularly with altitude the to-
tal ammonia has a very strong decrease with altitude near the surface. This indicates that a large fraction of ammonia reacts to ammonium above 900 hPa and that the model gives a strong gradient with altitude for NH_3. The vertical distribution of sulphate has an almost constant mixing ratio from surface to 800 hPa and with an actual increase with altitude closest to the surface. Also above 800 hPa the sulphate concentration is
significant. For nitrate (as fine mode ammonium nitrate) the mixing ratio is decreasing rapidly with altitude above 950 hPa as a result of its dependence of the availability of ammonium.

Adams et al. (1999) defined DON (degree of neutralization) as ($NH_4/2SO_4$) which equals 1 when sulphuric acid is totally neutralized by ammonia. Analogous to this
definition, we define NP (nitrate production) as the excess ammonia which is in the aerosols when all sulphuric acid is neutralized. NP is zero when all sulphuric acid is neutralized by ammonia (DON=1) and NP is larger than zero when there is excess ammonia in the aerosols. This excess ammonia is then associated with aerosol nitrate. Figures 5a and b show zonally averaged values for NP for January and July in our
model. The figures show that NP is larger than zero in the winter close to the ground in the northern hemisphere. In summer NP is rarely larger than zero anywhere meaning that the aerosols are rarely neutralized in summer.

To interpret these results, we plot the potential values of NP. We define this as Nitrate Production Potential (NPP) and assume that all gaseous ammonia can be used to
formed nitrate. Areas with NPP larger than zero are areas where fine mode nitrate can be formed given that the temperature is low enough. Whenever NPP is smaller than zero, fine mode nitrate can not be formed even if the temperature is very low. Figures 5c and d show NPP zonally averaged for January and July in our model. NPP is largest near the surface and decrease relatively fast with altitude. A secondary maximum

is found in the middle troposphere at equator. The strong reduction with altitude on the total ammonia is a main reason for the strong vertical gradient of the potential for ammonium nitrate fine mode particles to be produced, but also the increase or near constant sulphate with altitude in the lower troposphere contributes. The seasonal variation is significant and maximum in July in the southern hemisphere is partly due to very low sulphate.

There are two important reasons for the strong vertical gradients in NPP in our model. First, All ammonia emissions are on the ground. The Oslo CTM2 boundary layer scheme preserves a quite strong gradient from the lowest layer and to the top of the boundary layer. Second: There are significant secondary sources for sulphate in the free troposphere. Sulphuric acid is produced in the free troposphere both trough gaseous reactions and aqueous reactions. Figures 5c and d indicate that the Oslo CTM2 model can not produce significant nitrate in the free troposphere since it is limited by access to ammonia.

3.3 Nitrate aerosol optical depth and radiative effect

The global and annual mean burden of fine mode ammonium nitrate is $0.068 \, mg(NO_3)m^{-2}$ ($0.086 \, mg(NO_3)m^{-2}$ without sea salt) and the geographical distribution is shown in Fig. 6. The burden varies substantially from being significant in industrialized region to very low over all oceanic regions. Figure 6 shows also that it is significant distinction in the relative and absolute difference in the effect of sea salt on the burden. In absolute terms the difference in burden due to inclusion of sea salt in the simulation is largest over Europe and with some impact over India and China as well. Over Europe the reduction in the burden from sea salt attain almost 50%. Over ocean the burden of fine mode ammonium nitrate is very low due to low availability in ammonia. However, in several regions over ocean sea salt can eliminate the burden of fine mode nitrate.

Similar to the surface concentration and burden of nitrate the aerosol optical depth (AOD) of nitrate has rather localized maxima (Fig. 7). The results show peak values in

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India above 0.1, over Europe slightly larger than 0.05, and over USA of 0.04. The effect of sea salt to reduce the AOD of ammonium nitrate is mostly in the western part of the continents and most prominent near the coast (difference between Figs. 7a and b). In the industrial areas with local maxima in the nitrate AOD of ammonium nitrate account for around 10% of the total AOD based on simulation with multi component aerosol simulations with the Oslo CTM2. The global mean AOD of fine mode ammonium nitrate is 0.0010 (0.0013 without sea salt). To put this is context with other aerosol component from industrial sources the AOD from nitrate is around 5% of the AOD from sulphate, 30% lower than the AOD from organic carbon, and rather similar to the AOD from black carbon based on simulations with Oslo CTM2 for AEROCOM calculations with predefined emissions (Experiment B). The column burden of nitrate associate with sea salt particles is substantial 1.44 mg(NO_3)m^{-2}. Of the sea salt particles about 7% has a radius smaller than 0.5 μm and about 50% smaller than 1.25 μm. Therefore there is a substantial burden of fine mode $NaNO_3$, but these particles do only to some extent change the mass compared to the original sea salt particles (see Eq. 1). The mass increases slightly due to larger molecular mass of NO_3 compared to Cl, but this occurs without the number of moles increases. Figure 8 shows the annual mean aerosol optical depth from the $NaNO_3$ particles that are additional to original sea salt particles. The distribution shown in Fig. 8 has also maxima near industrialized regions, but much smaller land ocean gradients than in Fig. 7. The AOD values are also much lower in Fig. 8 than in Fig. 7. The global mean AOD from the $NaNO_3$ particles (compared to having the particles as NaCl) is 0.0006. The effect of changing the optical properties (through density, refractive index, hygroscopic growth) was to reduce the AOD by about a factor of 2.

The global and annual mean radiative forcing due to nitrate aerosols is −0.016 Wm^{-2} with a distribution similar to the distribution of the AOD. Annual mean peak values of −1.5 Wm^{-2} are simulated. Note that the simulations of the AOD and radiative forcing include anthropogenic as well as natural emission of ammonium nitrate precursors. The normalized radiative forcing is −230 Wg^{-1}, which is close to typical values for

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sulphate aerosols.

4 Summary

A scheme for gas/aerosol partitioning has been implemented into a multi component global aerosol transport model to simulate nitrate particles. Results show that nitrate particles have local maxima of significant concentration in industrialized region in accordance with measurements. A total radiative forcing of −0.02 Wm^{-2} (including anthropogenic and natural emission) indicate based on this study that the forcing of nitrate particles is on the weaker side of earlier estimates. The radiative forcing due to nitrate aerosols on a global scale is almost negligible, but on a regional scale the radiative forcing can be −1.5 Wm^{-2} for the annual mean and −3 Wm^{-2} at certain months. Also the regional surface concentration and aerosol optical depth of nitrate can be a significant fraction of the total aerosol composition. The modelled surface concentration is in reasonable agreement with measurements. Further, the vertical profile of total ammonia and sulphate will reduce the impact of fine mode ammonium nitrate with altitude in the lower troposphere. Therefore, we find it unlikely that the AOD and radiative forcing due to nitrate can be of significance on a global scale. Uptake of nitrate on sea salt particles reduces the AOD of ammonium nitrate by about 25%. However, the major limitation for higher production of ammonium nitrate is the availability of ammonium at levels where temperature (and therefore saturation vapor pressure of HNO$_3$) is low enough to produce nitrate aerosols. The burden of nitrate in sea salt particles is found to be large but do not give significant AOD and a radiative forcing.

The distribution and amount of nitrate particles may be dependent on the spatial resolution (M. Krol, personal communication). This will be investigated further in the Oslo CTM2 when meteorological data on 1 degree become available. Nitrate can also react on mineral dust particles in particular with dissolved calcium or magnesium carbonates (Derwent et al., 2003; Wolff, 1984). Further investigation with these reactions should be performed to investigate the influence on the coarse mode nitrate distribution.

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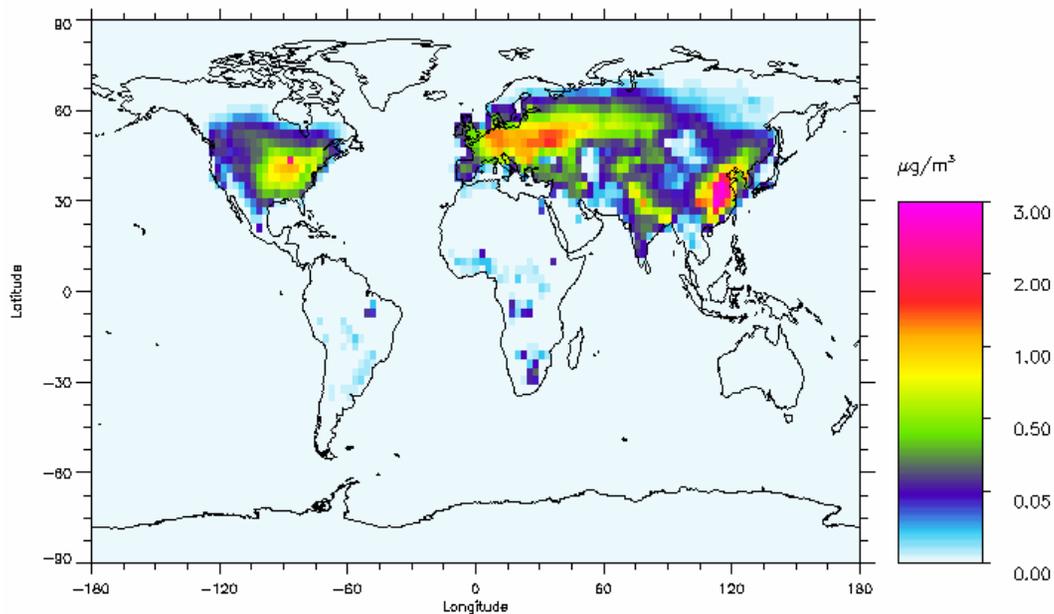
Fig. 1. Global distribution of annual mean fine mode surface concentration of nitrate (μg(NO_3)m^{-3}). Note that the scale is nonlinear.

Fig. 2. Global distribution of annual mean coarse mode surface concentration of nitrate (μg(NO$_3$)m^{-3}). Note that the scale is nonlinear.

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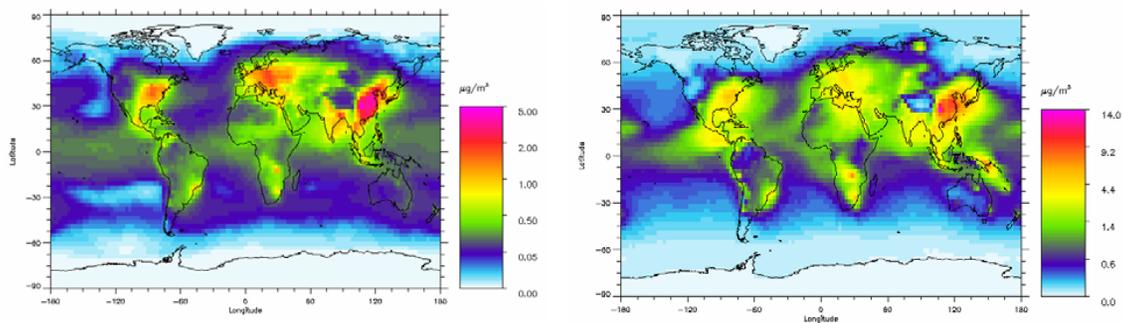
Fig. 3. Global distribution of annual mean fine mode surface concentration of ammonium (μg(NH$_4$)m^{-3}) in the left panel and the annual mean fine mode surface concentration of sulphate (μg(SO$_4$)m^{-3}) in the right panel. Note that the scales are nonlinear.

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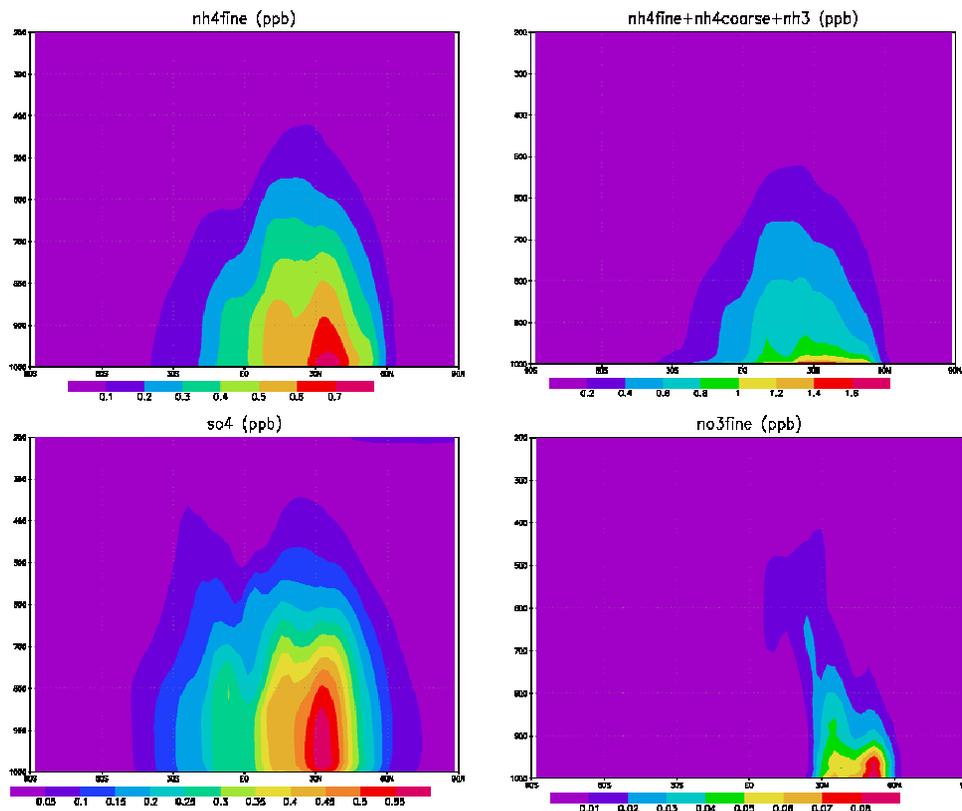
Fig. 4. Annual mean vertical and zonal mean mixing ratios (ppb) **(a)** fine mode NH$_4$ **(b)** total ammonia (NH$_3$+NH$_4$) **(c)** sulphate **(d)** fine mode nitrate.
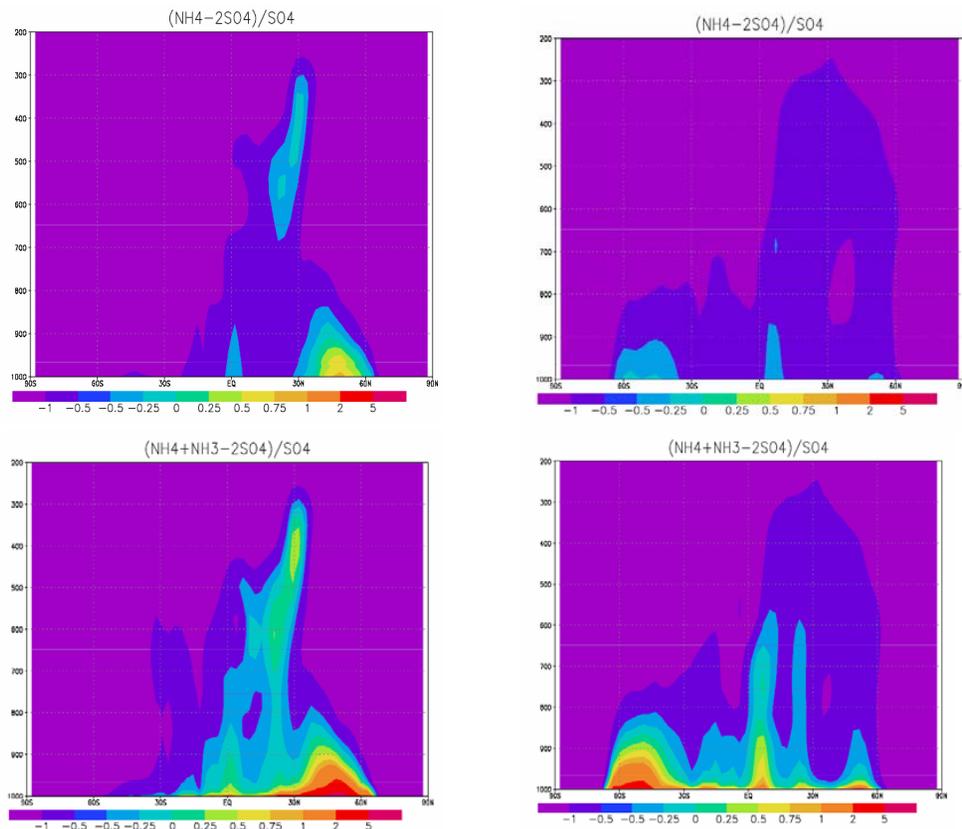
Fig. 5. In the upper panels the nitrate production (NP) is shown as the ratio of fine mode ammonium nitrate aerosol to sulfate aerosols (left January and right July). Nitrate production potential (NPP) is shown as the ratio of all available ammonia not used for neutralization of sulphate to sulfate aerosols in the lower panels (left January and right July).

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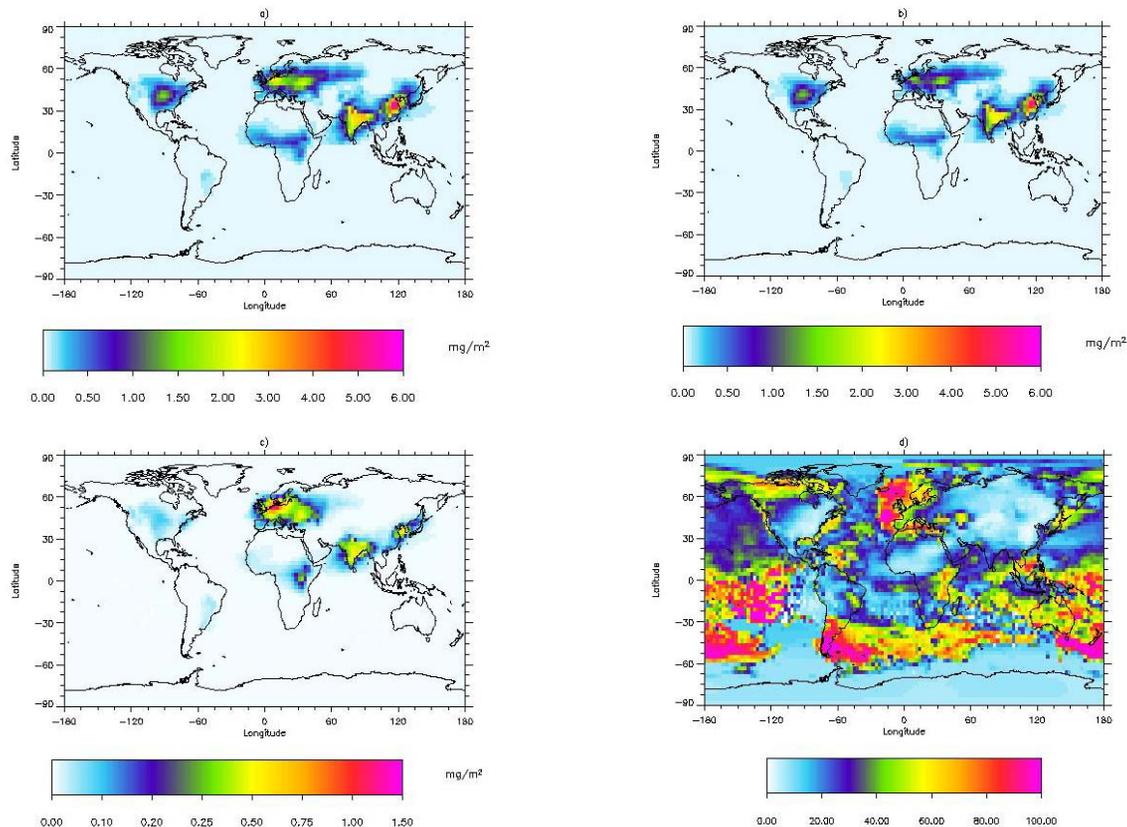
Fig. 6. Annual mean burden of nitrate (mg/m^2) **(a)** without sea salt, **(b)** with sea salt, **(c)** absolute difference between the simulation with and without sea salt, **(d)** percentage difference between the simulation with and without sea salt.
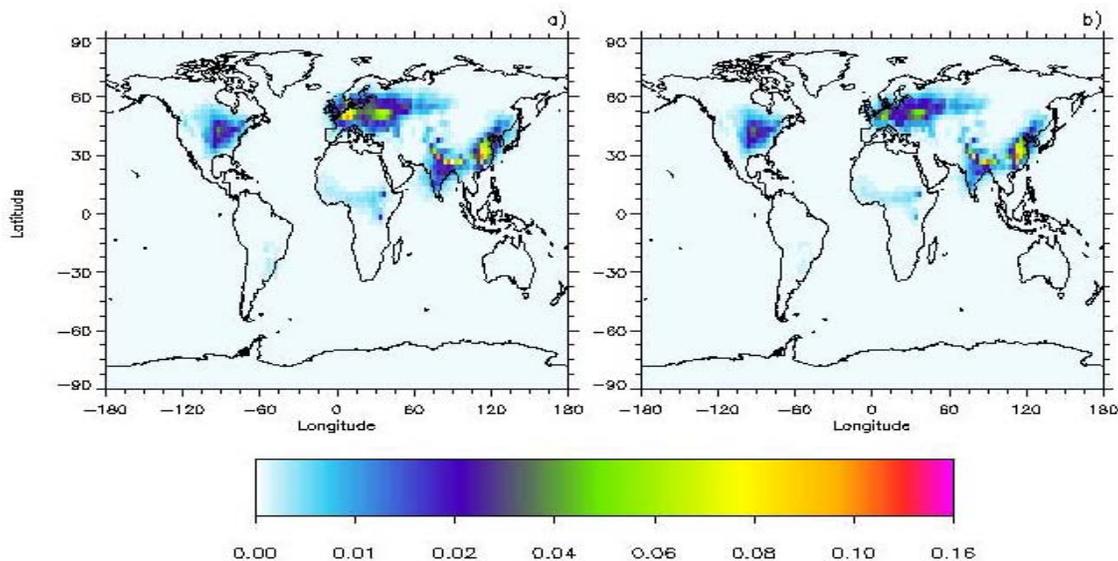
Fig. 7. Annual mean aerosol optical depth of fine mode ammonium nitrate aerosols **(a)** without sea salt, **(b)** with sea salt. Note that the scale is not linear.
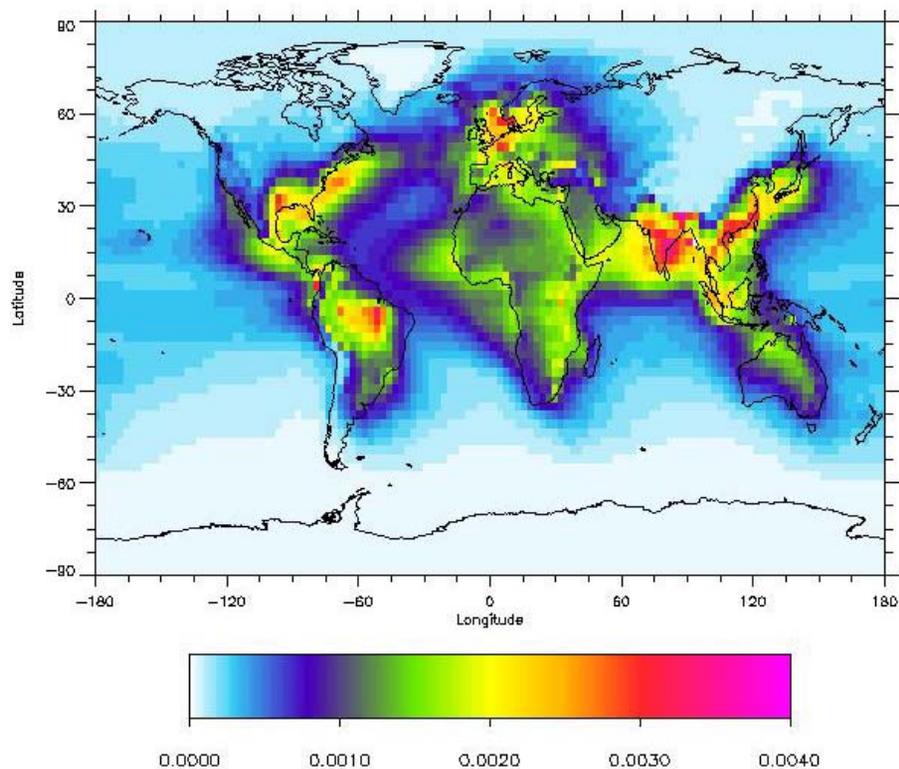
Fig. 8. Annual mean aerosol optical depth of coarse mode nitrate aerosols.