Modeling the Annual Cycle of Sea Salt in the Global 3D Model Oslo CTM2: Concentrations, Fluxes, and Radiative Impact

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ABSTRACT

A global three-dimensional chemical transport model (CTM) is used to model the yearly cycle of sea salt. Sea salt particles are produced by wind acting on the sea surface, and they are removed by wet and dry deposition. In this study, forecast meteorological data are taken from the ECMWF. The modeled concentrations are compared to measured concentrations at sea level, and both absolute values and monthly variations compare well with measurements. Radiation calculations have been performed using the same meteorological input data as the CTM calculations. The global, yearly average burden of sea salt is found to be 12 mg m$^{-2}$. This is within the range of earlier estimates that vary between 11 and 22 mg m$^{-2}$. The radiative impact of sea salt is calculated to be $\pm 1.1 \text{W m}^{-2}$. The total, yearly flux of sea salt is estimated to be 6500 Tg yr$^{-1}$.

1. Introduction

There has been an increased focus on the climate effect of tropospheric aerosols during the last few years. These aerosols consist of natural and anthropogenic species in particle form with radii from several nm to $\mu$m. Typical anthropogenic sources can be emissions of sulphuric gases leading to the formation of sulphate particles, or combustion to give carbon particles. The most important natural particles are sea salt and mineral dust that are emitted into the atmosphere because of wind stress at the ocean surface and arid land areas, respectively.

Several studies have quantified the flux and concentration of sea salt particles. Estimates of the total, global flux range from 1000 to 10 000 Tg yr$^{-1}$ (Blanchard 1985; Seinfeld and Pandis 1998). These estimates have often used empirical relations for the surface concentration, assumed a relationship between dry and wet deposition, and calculated the dry deposition. Tegen et al. (1997) found the global average burden of sea salt to be 22 mg m$^{-2}$ (with a source strength of 5900 Tg yr$^{-1}$) using a 3D tracer model whereas, Takemura et al. (2000) calculated the source strength to be 3321 Tg yr$^{-1}$ and the global burden to be 11 mg m$^{-2}$ using a general circulation model. Both studies calculate the sea salt concentration directly from an empirical dependence on surface wind.

Sea salt particles are important for the radiative balance, both directly, as a reflector of radiation, and indirectly as cloud condensation nuclei. Large uncertainties are associated with estimates of climate effect of natural and anthropogenic aerosol.

Earlier estimates of direct radiative effects of various aerosols have mainly been derived from models (Haywood and Boucher 2000). Satellite retrievals for aerosol purposes have advanced considerably over the last few years and will in the near future be an important tool for constraining the radiative forcing due to the direct aerosol effect, in particular, the anthropogenic part (King et al. 1999). Combinations of satellite observations and radiative transfer models have been used to estimate the clear-sky direct aerosol effect (Bergstrom and Russel 1999; Boucher and Tanre 2000; Haywood et al. 1999). Haywood et al. (1999) found a substantial difference between the top of the atmosphere clear-sky flux from the Earth Radiation Budget Experiment (ERBE) and a GCM. Including several aerosol components resulted in a much better agreement between the model and observation. Boucher and Tanre (2000) used retrieved aerosol optical properties and a radiative transfer model to estimate the direct radiative effect of aerosols. Both studies indicate a direct clear-sky radiative effect of aerosols of several W m$^{-2}$. The studies by Haywood et al. (1999) and Boucher and Tanre (2000) were limited to over-ocean and mostly clear-sky conditions. Uncertainties are large regarding natural versus anthropogenic aerosols contributions. Haywood et al. (1999) estimated a range for the global mean clear-sky radiative impact of sea salt from $-1.0$ to $-3.5$ W m$^{-2}$ ($-1.5$ to $-5.0$ W m$^{-2}$ over the ocean).

In this study, a 3D chemical transport model (CTM)
has been used to calculate the global concentration of sea salt particles. A radiative transfer model is used to estimate the radiative impact of sea salt aerosols in the atmosphere. The CTM is driven by forecast meteorological data from the European Centre for Medium-Range Weather Forecasts (ECMWF). The meteorological data are used in a consistent way so that both the radiation study and the transport study use the same data regarding wind-driven sources, transport, and radiative effects. Previous studies have either used less-sophisticated models or calculated boundary layer concentrations directly from wind speed correlations.

2. Modeling

a. CTM modeling

The processes taken into account in the model are production, transport by advection and convection, particle growth (by absorption of humidity from the air), dry deposition, and wet deposition. Each of these processes are described in detail below. Coagulation of particles has not been taken into account in this study.

b. Oslo CTM2

The model used in the calculations is the 3D Oslo CTM2 model described in Sundet (1997). It is an offline CTM that uses precalculated transport and physical fields to simulate chemical turnover and distribution in the atmosphere. The model is valid for the global troposphere with 19 vertical layers and the model domain reaching from the ground up to 10 hPa. Horizontal resolution is 5.625° (T21). The model uses data from ECMWF. Advection is done conserving second-order moments (Prather 1986) and convection is based on the Tiedtke mass flux scheme (Tiedtke 1989), where vertical transport of species is determined by the surplus/deficit of mass flux in a column. Meteorological data for 1996 are used.

1) Sea Salt Production

Sea spray is generated by the wind stress on the ocean surface. Air bubbles, which constitute the whitecaps resulting from breaking waves, burst at the water surface and produce small droplets by means of two mechanisms. Film drops are produced when the thin liquid film that separates the air within a bubble from the atmosphere ruptures. The remaining surface energy of the bubble, after bursting, results in a liquid jet that becomes unstable and breaks into a number of jet drops (Smith et al. 1993). The formation of film and jet drops can be called indirect mechanism. At wind speeds greater than 10–12 m s⁻¹, spume drops torn directly from the wave crests by the strong turbulence make an increasing contribution to the sea salt and dominate the concentration at larger particle sizes. The formation of spume drops is called the direct mechanism.

Production is described empirically by Monahan et al. (1986) from laboratory experiments:

$$\frac{\partial F}{\partial r} = 1.373 U_{10}^{1.51} r^{-3}(1 + 0.057 r^{1.05}) 10^{1.19\text{e}^{-0.057r}},$$

where $B$ is

$$B = \frac{0.380 - \log r}{0.650},$$

for the indirect mechanism (bubbles bursting) and

$$\frac{\partial F}{\partial r} = 8.60 \times 10^{-6} e^{2.08 U_{10}} r^{-2}$$

for direct mechanism (spume).

Here, $F$ is flux in particles m⁻² s⁻¹, $U_{10}$ is wind speed at 10-m height in m s⁻¹, and $r$ is particle radius in μm.

A similar production mechanism has been used in other works (Gong et al. 1997a; Pryor and Sorensen 2000). Equation (1) is strictly for particles bigger than radius of 0.8 μm at 80% RH, but since we do not have any better expression for smaller particles, Eq. (1) is used generally in this study, as was done by Gong et al. (1997a).

Smith et al. (1993), Gong et al. (1997a), and Pryor and Sorensen (2000) point out that the fluxes given by Monahan et al. (1986) were too big for the spume mechanism. Based on measurements, Smith et al. (1993) proposed another expression that gives a more correct representation of the fluxes for bigger particles. The expression given by Smith et al. (1993) is used in this work for production of sea salt by the spume mechanism.

The expression proposed by Smith et al. (1993) for the sea salt flux, is

$$\frac{\partial F}{\partial r} = \sum_{i=1}^{2} A_i e^{-f_i (\text{ln}(R/r_{01}))},$$

where $f_1$, $f_2$, $R_{01}$, $R_{02}$, have the values 3.1, 3.3, 2.1, and 9.2 μm, and

$$\log A_1 = 0.0676U + 2.43$$

$$\log A_2 = 0.959U^{1/2} - 1.476,$$

where index 1 means the indirect mechanism and index 2 means the direct mechanism.

Since the equations given by Monahan et al. (1986) seem to give reasonable fluxes for small particles, but not for large ones, the production of particles smaller than 7 μm was calculated using these equations. Larger particles were calculated according to Smith et al. (1993). At a radius of 7 μm these two schemes give approximately the same flux for different wind speeds.

Sea salt aerosols up to about 20 μm are found in the atmosphere (Erickson and Duce 1988). We assume that the 16 size bins given in Table 1 are sufficient to estimate the sea salt mass balance.

For dry particles, we assume a density of 2200 kg m⁻³,
the same density as pure and dry NaCl (Hess et al. 1998). Sea salt production is calculated assuming a relative humidity of 80%. At this relative humidity, the particle radius will be twice the dry radius (Fitzgerald 1975), and the density used in the production is thus 1150 kg m$^{-3}$.

The number of particles produced is converted to mass of particles according to

$$M = N \times \rho_p \times \frac{4}{3} \pi r^3,$$

where $M$ is total mass produced in one grid cell (kg), $N$ is total number produced, $\rho_p$ is particle density, and $r$ is radius.

The sea salt mass and radius are corrected for water using the formulas described in section 2b(2). Only the dry mass is added to the bin and transported. Thus we do not transport any water. Mass is not transferred between the bins because of growth. However, we do calculate wet density and radius for all the size bins as this will influence the dry deposition.

2) PARTICLE GROWTH

The particles will absorb water and grow as a function of relative humidity in the air. Ideally, a chemical equilibrium approach should be used. A chemical equilibrium model would calculate composition of aerosols with respect to water, and aerosol species based on thermodynamics (Zhang et al. 2000).

Since chemical equilibrium is not taken into account, the composition of the particles is constant during transport. The chemical composition of the dry particles is assumed to be 30.6% sodium, 55.04% chlorine, and 14.4% of other inorganic components as given by, for example, Seinfeld and Pandis (1998).

A simplified approach, described by Fitzgerald (1975), was used here. The radius of the aerosol after absorbing water vapor is given as

$$r = \alpha r_d^\beta,$$

where $r_d$ is dry radius, and $\alpha$ and $\beta$ are coefficients depending on chemical composition of aerosol and on relative humidity.

When calculating the density of the particles, it is assumed that the volume of water and the volume of dry particle can be added together to give the total volume. Thus it is assumed that the dry particle still occupies the same volume in the wet particle as when dry.

The equations from Fitzgerald (1975) are valid for particles with dry radius up to 3 $\mu$m. However, they are used generally for all particle sizes in this work and will probably introduce some errors for the larger particles.

3) DRY DEPOSITION

The dry deposition velocity is the velocity at which the particles are transported to the ground. The flux toward the ground is given as

$$\text{flux} = [(C(z) - C(0)) v_{\text{dep}},$$

where flux is given as $\mu g (m^2 s)^{-1}$, $z$ is the height (m), $C$ is concentration ($\mu g m^{-3}$), and $v_{\text{dep}}$ is the dry deposition velocity ($m s^{-1}$). The concentration on the ground is assumed to be 0. (That is, the particles are taken up completely if they reach the ground.)

The dry deposition velocity is calculated from the scheme given by Seinfeld and Pandis (1998). The dry deposition velocity is given by

$$v_{\text{dep}} = \frac{1}{r_a + r_g + r_g r_a v_f} + v_f,$$

where $r_a$ is aerodynamic resistance, $r_g$ is resistance in the quasi-laminar sublayer, and $v_f$ is the falling velocity. The expressions used to find these parameters are shown below.

The aerodynamic resistance is given by

$$r_a = \begin{cases} \frac{1}{\kappa H_0} \ln \frac{z}{z_0} + 4.7(\xi - \xi_0) & \text{(stable)} \\ \frac{1}{\kappa H_0} \ln \frac{z}{z_0} & \text{(neutral)} \\ \ln \frac{z}{z_0} + \ln \frac{(\eta_0^2 + 1)(\eta_0 + 1)^2}{(\eta^2 + 1)(\eta + 1)^2} + 2(\arctan \eta_r - \arctan \eta_a) & \text{(unstable)} \end{cases},$$

where $\xi$ is $z/L$ and $\eta = (1 - 15\xi)^{1/4}$. Here $L$ is the Monin–Obukhov length (the height above the ground where the production of turbulence by mechanical and buoyancy forces are equal). The subscript 0 denotes the roughness length and $r$ denotes the reference height (which is the middle of the grid box). The equations apply to different states of the atmosphere (stable, unstable, and neutral).
Cloud fraction and cloud liquid water are diagnosed in the ECMWF model as standard fields available from the prognostic cloud scheme. Rainfall is not a standard parameter and is diagnosed as the sum of all precipitation processes in the ECMWF model; thus, rainfall is the sum of large-scale precipitation and convective precipitation. These fields provide an accurate three-dimensional description of the rainfall as it is estimated in the ECMWF model.

c. Radiation transfer model and optical properties of sea salt

In the radiative transfer calculations a multistream model using the discrete ordinate method is used (Stamnes et al. 1988). In this study, eight streams are used. Rayleigh scattering, scattering and absorption by aerosols and clouds, and absorption by ozone and water vapor are included in the radiative transfer scheme. Absorption by ozone and water vapor are included using the exponential sum fitting method (Wiscombe and Evans 1977). Four spectral regions are used [see Myhre and Stordal (2001) for further details]. In the radiative transfer scheme meteorological fields (temperature, water vapor, clouds, and surface albedo) from ECMWF data are used, so that it is consistent with the sea salt calculations from the Oslo CTM2.

Clouds are included on the basis of cloud liquid water content and cloud amount from the ECMWF data. Based on radar observations (Hogan and Illingsworth 2000), the random cloud overlap assumption is used. The optical properties of the clouds are based on Slingo (1989) with an effective radius of 10 \( \mu \text{m} \) for clouds at pressure levels higher than about 300 hPa and 18 \( \mu \text{m} \) for clouds at lower pressures (Stephens 1978; Stephens and Platt 1987). Radiative transfer calculations are performed every 3 h, taking into account the zenith angle variations, updated meteorological fields, and sea salt concentrations.

Optical properties (specific extinction coefficient, single-scattering albedo, and asymmetry factor) of the sea salt aerosols are calculated using Mie theory (Wiscombe 1987). Eight streams are used. In this study, eight streams are used. Rayleigh scattering, scattering and absorption by aerosols and clouds, and absorption by ozone and water vapor are included in the radiative transfer scheme. Absorption by ozone and water vapor are included in the radiative transfer scheme. Optical properties of the clouds are based on Slingo (1989) with an effective radius of 10 \( \mu \text{m} \) for clouds at pressure levels higher than about 300 hPa and 18 \( \mu \text{m} \) for clouds at lower pressures (Stephens 1978; Stephens and Platt 1987). Radiative transfer calculations are performed every 3 h, taking into account the zenith angle variations, updated meteorological fields, and sea salt concentrations.

Optical properties (specific extinction coefficient, single-scattering albedo, and asymmetry factor) of the sea salt aerosols are calculated using Mie theory (Wiscombe 1987). Eight streams are used.
TABLE 2. Some earlier estimates for the global flux.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Total flux estimate (Tg yr⁻¹)</th>
<th>Comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Petrenchuk (1980)</td>
<td>1000</td>
<td>Assumes that the sea salt is homogeneously distributed and that dry deposition accounts for 10% of the deposition.</td>
</tr>
<tr>
<td>Erickson and Duce (1988)</td>
<td>10 000–30 000</td>
<td>Calculates concentrations from empirical equations. Uses one scavenging coefficient for the whole world.</td>
</tr>
<tr>
<td>Gong et al. (1997b)</td>
<td>10 000</td>
<td>Does not take into account spume production. Averages local production rates to obtain global production rate by using a 1D model at different sites of the world.</td>
</tr>
<tr>
<td>Tegen et al. (1997)</td>
<td>5900</td>
<td>Calculates surface layer concentrations from empirical functions, and not from production loss terms. Neglecting growth of particles from absorption of water vapor in the air.</td>
</tr>
<tr>
<td>Takemura et al. (2000)</td>
<td>3300</td>
<td>Calculating first layer concentrations from empirical functions, and not from production loss terms.</td>
</tr>
<tr>
<td>This study</td>
<td>6500</td>
<td>Uses global wind data from ECMWF together with equations from Monahan et al. (1986) and Smith et al. (1993) to give global production.</td>
</tr>
</tbody>
</table>

3. Results

a. Global results/budgets

1) Total flux

Most estimates of sea salt fluxes are between 1000 and 10000 Tg yr⁻¹ (Blanchard 1985; Seinfeld and Pandis 1998). In Table 2, some earlier studies in are listed. The obtained flux of 6500 Tg yr⁻¹ is within the expected range.

Fields for the total production are given in Figs. 1a and 1b. It can be seen that the production is largest when the wind is high, for instance at midlatitudes.

2) Removal mechanisms

Removal occurs by wet deposition and dry deposition as described above.

In this study, we find that dry deposition is the dominant removal mechanism (see Table 3).

Erickson and Duce (1988) have given flux fields in the same way as above for dry and wet deposition. The fields from this study and the fields from Erickson and Duce (1988) do not differ much [even though Erickson and Duce (1988) get a yearly flux of 10 000–30 000 Tg yr⁻¹ as opposed to 6500 Tg yr⁻¹ found in this study].

3) Concentration fields

The distribution of sea salt particles near the ground is shown in Figs. 1g and 1h. It can be seen that concentrations over land are very small. It can be seen from Figs. 1g and 1h that the concentrations are largest where the winds are large, for instance at midlatitudes. The areas with high concentrations are the same as the areas with high production (see Fig. 1).

4) Burden

At the surface the geographical distribution of the annual mean sea salt concentration is compared to a compilation by Koepke et al. (1997). The global aerosol dataset (GADS) in Koepke et al. (1997) is a climatological compilation based on observations and models. GADS is often used in satellite retrievals. In the modeled distribution in Fig. 2a, the highest concentrations are found at midlatitudes, particularly in the Southern Hemisphere. High concentrations are also found near the equator.

In Fig. 2b, the annual mean sea salt concentration in the lowest layer from Koepke et al. (1997) is shown. The geographical pattern is generally very similar in the two distributions, with the largest difference near the equator associated with higher values in the modeled distribution. At midlatitudes the modeled concentrations are substantially higher in the Southern Hemisphere than in the Northern Hemisphere, whereas in Koepke et al. (1997), the maximum concentrations in the Northern Hemisphere are slightly higher than in the Southern Hemisphere.

The geographical distribution of the annual mean at-
The atmospheric burden of sea salt particles from our model and from Koepke et al. (1997) is shown in Figs. 2c and 2d. In Koepke et al. (1997), the surface concentration is given and the vertical distribution is calculated using exponential profiles with a scale height (Hess et al. 1998), which is the same over all maritime regions.

Surface concentration depicted in Fig. 2 show good agreement between GADS and our model. However, the geographical distribution of the total sea salt burden differ substantially between the model and GADS. In the model, burden maximum is at low latitudes, whereas the burden from the Koepke et al. (1997) dataset has a maximum at midlatitudes. In the latter pattern, the distribution of the burden is actually reflecting the surface concentration since the scale height is the same over ocean. At midlatitudes in the Southern Hemisphere the sea salt concentration is about 50% higher in the model than in the Koepke dataset at the surface level.
other hand the burden of sea salt is about 50% higher in the Koepke dataset than in the model at midlatitudes in the Southern Hemisphere.

Transport and deposition processes are important for the calculated vertical profiles. In particular wet deposition is important at midlatitudes. This shows that the use of scale heights to estimate the vertical distribution of sea salt particles is a strong simplification.

Global and annual mean burden of sea salt is 12 and 15 mg m$^{-2}$ in our model and in the Koepke et al. (1997) dataset, respectively. In Tegen et al. (1997) the sea salt burden was calculated to 22.4 mg m$^{-2}$, whereas in Haywood et al. (1999), the burden in the high case was 36.8 and in the low case 7.5 mg m$^{-2}$. Takemura et al. (2000) calculated a sea salt burden of 11.0 mg m$^{-2}$.

In comparison, global average burden of anthropogenic sulfate aerosols range from 1.7 to 3.2 mg m$^{-2}$ (Myhre et al. 1998) and total burden of mineral dust for two datasets were 35 mg m$^{-2}$ and 110 mg m$^{-2}$, respectively (Myhre and Stordal 2001).

<table>
<thead>
<tr>
<th>Removal mechanism</th>
<th>Gong et al. (1997b) (%)</th>
<th>Erickson and Duce (1998) (%)</th>
<th>This study (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dry deposition</td>
<td>66</td>
<td>70</td>
<td>80</td>
</tr>
<tr>
<td>Subcloud wet deposition</td>
<td>33</td>
<td></td>
<td></td>
</tr>
<tr>
<td>In cloud wet deposition</td>
<td>1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Total wet deposition</td>
<td>34</td>
<td>30</td>
<td>20</td>
</tr>
</tbody>
</table>

5) RIVER RUNOFF

Petrenchuk (1980) estimates that the river runoff of sea salt is 300–400 Tg yr$^{-1}$. This should equal what is deposited over land (as nothing is assumed accumulated on land). Using yearly river data of $3.56 \times 10^9$ km$^3$ and an average chlorine concentration in rivers of 6.4–7.8 mg L$^{-1}$, Petrenchuk (1980) finds 230–280 Tg yr$^{-1}$ of chlorine. This should be approximately the same as the chlorine deposited over land from sea salt aerosols. Our model deposits 161 Tg yr$^{-1}$ of sea salt over land, corresponding to 88 Tg yr$^{-1}$ of chlorine. Given the uncertainties and assumptions made in such a comparison the estimates compare reasonably well. The comparison assumes that river concentration of chlorine is constant, that we have a steady state in chlorine at land, that no chlorine evaporates from the aerosols, and that the only way the chlorine can escape from land to ocean is by rivers.

b. Local concentrations and distributions

In this section, local concentrations and size distributions are shown. Some concentrations have been compared to measurements, and the size distributions are discussed based on data found in literature. The concentrations [in $\mu$g(Na) m$^{-3}$] are calculated by assuming that the weight percent of Na in sea salt is 0.3061 (Seinfeld and Pandis 1998).
Fig. 3. Concentrations in $\mu$g(Na) m$^{-3}$ at (left) Barbados and (right) Bermuda for 1996.

Fig. 4. Concentrations in $\mu$g(Na) m$^{-3}$ at Heimaey, Iceland, for 1996 and concentrations in grid box to the east of Heimaey. The grid box in the Oslo CTM2 containing the Heimaey station has a large percentage of land.

1) TIME SERIES 1996

Monthly average measurements for 1996, were obtained from D. Savoie at the University of Miami Rosenstiel School of Marine and Atmospheric Science (2000, personal communication). The CTM is run for the same year with 2-month spinup.

Figures 3 and 4 show the concentrations at several sites of the world.

The measurements for Bermuda and Barbados fit very well with measured data for the mass of sea salt aerosols. Both of these stations lie in areas where neither the production or any of the removal processes have large gradients (see Fig. 1). Both Bermuda and Barbados have an “average” production, dry and wet deposition, which means that the gridbox mean would not be an unrealistic approximation for the actual concentration at the station.

The fit is not so good for Heimaey, Iceland. There can be several reasons for the discrepancy. First, the grid containing Heimaey consists of 50% land in the model. This means that if the measurements are done close to the sea, the modeled concentration is an average of sea–land concentration that will be too low for a good fit. The grid cell will have emissions only half of which it would have had if the whole cell was covered by sea. Local conditions that will not be captured by the model can also play a part. It can be seen that the argument of 50% of the grid box being land plays a part as concentrations are significantly higher in the grid box to the east of Heimaey with 100% sea cover (see Fig. 4). In both cases, the model fails to reproduce the maximum measured in December. This maximum could be due to local conditions at Heimaey.

2) TIME SERIES 5-YR AVERAGE

Gong et al. (1997b) give time series at some more stations as 5-yr averages. It is useful to compare time series modeled for 1996 with these measurements to verify that the model can predict the right order of magnitude and the right seasonal variations for the concentrations. Figure 5 shows the modeled concentration compared to measured 5-yr averages.

The size order and seasonal variations are reproduced well for these two stations. It is important to notice that the Oslo CTM2 model manages to reproduce the minimum for the Northern Hemisphere summer at Hawaii. Gong et al. (1997b) point out that since they have a wind maximum (high production) and a precipitation minimum (low scavenging) for this period, their model should have problems reproducing the concentration minimum. Hawaii is situated in the Pacific in an area with large gradients in wind speed. Using winds from ECMWF, we do not get a production minimum in summer for this station, but a rather constant production throughout the year. This underlines the need for realistic meteorological data in combination with realistic production, loss, and transport in the study of sea salt particles.

The Oslo CTM2 fails to reproduce the winter maximum over Mace Head, Ireland. The model underestimates the concentrations by 24% in January, 44% in
February, and 48% in March. This could be a result of local conditions similar to those discussed above for Heimaey. It can also be seen that the modeled results (1996) show more variation than the 5-yr mean curve that is smoother. It is normal that calculations for one particular year deviate from a 5-yr average.

In Table 4, some average standard deviations for the monthly mean time series are shown. The deviation is calculated for each month and then averaged for the whole year. Heimaey is not included for reasons discussed above.

3) WEEKLY AVERAGES

At some stations, we have compared weekly averages measured from 1996 to weekly averages from the model. This should give an indication if the dependence on meteorology (e.g., wind, rainfall) is correct. The variations should correspond not only seasonally, but also on shorter timescales.

Figure 6 shows weekly averages for two stations during the first few weeks of 1996. The modeled variations seem to follow the observations nicely. The model concentrations covary with the measured concentrations.

At the Reunion Island site we have quite large weekly variations, something that is reproduced by the model. The model generally overestimates the concentrations at this site with a minimum overestimation of 18% in week 6 and a maximum overestimation of 470% in week 5.

At the Chatman Island site where the measurements show low variance, the model shows low variance as well. At this site, the model generally overestimates the concentrations. The model overestimates by 52% in week 1, 60% in week 4, and 76% in week 8, but for some weeks, the model underestimates the concentrations, such as in week 3 (12%) and week 5 (31%).

4) SIZE DISTRIBUTIONS

The mass size distributions at selected stations are shown in Fig. 7.

The size distribution is expected to be lognormal. In this case, we get a bimodal distribution with another maximum at about \( r = 10 \mu m \) because of the contribution of spume particles to the production.

Erickson and Duce (1988) propose that the distribution fitted to lognormality should be distributed about the radius \( r = 0.422u^{2.12} \), where \( u \) is surface wind speed. This would mean that for a surface wind speed of 10 m s\(^{-1}\), the distribution would be lognormally distributed around \( r = 6 \mu m \). The actual measured mass median radius from the work of Erickson and Duce (1988) varied between 3.5 and 7.5 \( \mu m \). They do not give any standard variation. Seinfeld and Pandis (1998) also propose that marine background aerosols are lognormally distributed around \( r = 6 \mu m \). The Oslo CTM2 approximately reproduces this size distribution.

5) VARIATION WITH HEIGHT

Mass concentrations decrease with height. The loss terms that lead to this are the falling velocity and the scavenging. The variation of total mass with height is shown in Fig. 8.

We do not get significant transport of sea salt mass to higher altitudes in the troposphere. The zonal mean or aerosol mass is high at latitudes with high wind and high ocean fraction. The washout process removes any sea salt that is transported to higher altitudes, therefore
no significant amount of sea salt mass is found over 750 hPa.

The high concentrations at 60°S is a consequence of high wind speeds, and small land fraction at this latitude.

The mass of different size bins vary differently with height. The larger particles have a higher falling velocity, and the concentration of these particles decrease more rapidly with altitude than the mass of the smaller particles. Figure 9 shows the concentration as a function of height at two stations.

At Heimaey in January, where washout is expected to be efficient, we see a more rapid decrease in concentrations with height than at Hawaii where washout is expected to be less efficient. The vertical profile is also influenced by different production (of different sizes of particles) or differences in transport.

We have made a global average column of the modeled results. The concentrations have been approximated to a formula of the form \( C = C_0 e^{-Z/H} \) where \( C \) is concentration in \( \mu g \) m\(^{-3} \), \( Z \) is altitude, and \( H \) is scale height (both measured in km). There are two distinguished regimes with \( H = 1 \) km below approximately 2 km and \( H = 5.5 \) km above approximately 2 km. For comparison, Koepke et al. (1997) used scale heights of 1 and 8 km for the two regions. This means that our model contains less sea salt mass in the free troposphere.

c. Radiative impact

In this section we will use the term radiative forcing of the radiative impact of sea salt. Usually radiative forcing is used for an external perturbation (anthropogenic or natural perturbation such as solar irradiation variation or aerosols from volcanic eruptions; Houghton et al. 1996); however, here we use the term radiative forcing for a natural component.

Results are performed with clouds included and for clear sky (clouds excluded in the calculations). Results for clear sky are presented first as these are most comparable to the results from Haywood et al. (1999) and Boucher and Tanre (2000), which are combinations of satellite observation and modeling. However, note that satellite observations are only performed for clear sky conditions.
when clouds are not present and therefore not identical to our clear-sky assumption.

Figure 10a shows the annual mean geographical distribution of the clear-sky radiative forcing due to sea salt. In general, the pattern of the clear-sky radiative forcing is very similar to the aerosol optical depth at 500 nm (not shown). The maximum forcing is around 60°S, with a secondary maximum at 20° in each hemisphere. The aerosol optical depth shows a somewhat smaller difference between these maxima than the forcing. This can be explained by noting that the radiative forcing due to particles is largest at high solar zenith angles (Haywood and Shine 1997).

A similar pattern as that shown in Fig. 10a is found in Haywood et al. (1999) for sea salt; however, in their analysis, few observations around 60°S exist. In Boucher and Tanre (2000) all aerosol components are included; however, over ocean in non-mineral regions as well as far from land, similar radiative impact of aerosols can be found. The radiative effect of sea salt over land is very small in accordance with the low concentrations estimated.

In Fig. 10b, clouds are included. This reduces the magnitude of the forcing as for other scattering aerosols. At mid-latitudes the radiative forcing due to sea salt is substantially weakened as the cloud cover in these regions is very high. In the more cloud-free regions around 20° the impact of clouds is smaller, therefore the strongest radiative forcing is in these regions. Clouds at mid-latitudes therefore strongly reduce the radiative impact of sea salt both by wet deposition and scattering of sunlight.

The global mean radiative forcing due to sea salt is $-1.1 \text{ W m}^{-2}$ for a cloudy atmosphere, and $-2.2 \text{ W m}^{-2}$ for clear sky. The clear-sky estimate is between the high and low estimates of sea salt in Haywood et al. (1999). Boucher and Tanre (2000) estimated a global mean clear-sky radiative forcing over ocean of about $-5.5 \text{ W m}^{-2}$ including natural and anthropogenic aerosols. To investigate the forcing due to sea salt within the clouds we used the procedure described in Haywood and Shine (1997). Neglecting sea salt aerosols in regions with clouds (in the whole column) gives a forcing of $-0.7 \text{ W m}^{-2}$. This estimate is one-third of the clear-sky ra-
diative forcing, a result that shows that clouds are very often present in regions with sea salt. We find a forcing of $-0.4$ W m$^{-2}$ inside cloudy regions or 35% of the radiative forcing.

Normalized forcing (global mean radiative forcing divided by the global mean burden) has been calculated by many investigators for sulfate aerosols (see Myhre et al. 1998, and references therein). It ranges from about $-550$ to $-125$ W g$^{-1}$. For mineral dust the normalized forcing range from $-11$ to $+4$ W g$^{-1}$ in Myhre and Stordal (2001). In the calculations performed in this study the normalized forcing for sea salt is $-88$ W g$^{-1}$. The lower normalized forcing for sea salt aerosols than for sulfate is mainly due to larger sizes of the sea salt aerosols. Mineral aerosols also have large sizes like the sea salt particles. In addition they absorb solar radiation and have a significant thermal infrared radiative forcing leading to small normalized forcing for mineral aerosols.

We have performed two additional calculations in which we have reduced the number of size bins from 16 to 8 and 4. Differences in results when the number of size bins are reduced may have several origins. This will influence the dry deposition as the dry deposition shows a strong, nonlinear variation with the radius (Seinfeld and Pandis 1998), and the total burden is changed. Further, the optical properties may change either as a result of change in the calculated size distribution or simply in the averaging procedure in the calculation of the optical properties. Reducing the number of size bins from 16 to 8 decreases the radiative forcing only by 4%. However, as the total burden is slightly increased (2%), the normalized forcing is reduced by 6%. When four size bins are used, larger changes occur. The radiative forcing is 7% stronger, whereas the total burden is 23% higher, giving a normalized forcing that is 13% lower. The scattering efficiency for aerosols is strongly dependent on the size of the aerosols. In the visible region the scattering efficiency has a strong maximum for aerosols with radius around 0.5 μm (see, e.g., Seinfeld and Pandis 1998). This nonlinear variation in the scattering efficiency leads to an underestimation of the radiative forcing for 4 and 8 size classes compared to 16 size classes. However, larger differences in the burden is found, mainly because of averaging the dry deposition over larger size intervals.

FIG. 10. Yearly averaged direct radiative forcing due to sea salt in (a) clear-sky conditions and (b) taking clouds into account.
4. Summary

A global 3D CTM (the Oslo CTM2) has been used to simulate the concentration of sea salt particles in the atmosphere. The distribution is determined by production (generation by wind), transport (advection and convection), wet and dry deposition, and growth by condensation. This study calculates the fluxes of sea salt as a function of wind speed. Earlier studies have either calculated sea salt concentrations in the boundary layer directly from empirical wind speed correlations or used less-sophisticated models (e.g., 1D models).

Monthly averaged distributions have been compared with measurements from Hawaii, Barbados, Bermuda, Ireland, and Iceland, and with weekly averages from Chatman Island and the Reunion Island. The calculated surface concentrations vary between 0 and 16 µg(Na) m⁻³ and fit well with the measured values.

The global flux of sea salt is calculated to be 6500 Tg yr⁻¹. Earlier estimates lie between 1000 and 10 000 Tg yr⁻¹. Average burden of sea salt is calculated to 12 mg m⁻², which is within the range of earlier estimates that are between 7.5 and 36 mg m⁻². Global average radiative forcing of sea salt is estimated to −1.1 W m⁻² when the effect of clouds is taken into account and −2.2 W m⁻² when the effect of clouds is ignored. It is important to take the effect of clouds into account in a consistent way using the same meteorology to calculate emissions, transport, and radiative forcing, since the sea salt concentrations often are high in areas with extensive cloud cover.

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