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# **DMS cycle in the marine ocean-atmosphere system – a global model study**

**S. Kloster, J. Feichter, E. Maier-Reimer, K. D. Six, P. Stier, and P. Wetzel**

Max Planck Institute for Meteorology, Hamburg, Germany

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Correspondence to: S. Kloster (kloster@dkrz.de)

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## Abstract

A global coupled ocean-atmosphere modeling system is established to study the production of Dimethylsulfide (DMS) in the ocean, the DMS flux to the atmosphere, and the resulting sulfur concentrations in the atmosphere. The DMS production and consumption processes in the ocean are simulated in the marine biogeochemistry model HAMOCC5, embedded in a ocean general circulation model (MPI-OM). The atmospheric model ECHAM5 is extended by the microphysical aerosol model HAM, treating the sulfur chemistry in the atmosphere and the evolution of the microphysically interacting internally- and externally mixed aerosol populations.

We simulate a global annual mean DMS sea surface concentration of 1.8 nmol/l, a DMS emission of 28 Tg(S)/yr, a DMS burden in the atmosphere of 0.077 Tg(S), and a DMS lifetime of 1.0 days. To quantify the role of DMS in the atmospheric sulfur cycle we simulate the relative contribution of DMS-derived  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  to the total atmospheric sulfur concentrations. DMS contributes 25% to the global annually averaged  $\text{SO}_2$  column burden. For  $\text{SO}_4^{2-}$  the contribution is 27%.

The coupled model setup allows the evaluation of the simulated DMS quantities with measurements taken in the ocean and in the atmosphere. The simulated global distribution of DMS sea surface concentrations compares reasonably well with measurements. The comparison to  $\text{SO}_4^{2-}$  surface concentration measurements in regions with a high DMS contribution to  $\text{SO}_4^{2-}$  shows an overestimation by the model. This overestimation is most pronounced in the biologically active season with high DMS emissions and most likely caused by a too high simulated  $\text{SO}_4^{2-}$  yield from DMS oxidation.

## 1. Introduction

Major uncertainties remain in the quantitative assessment of the climate response to anthropogenic forcing. Biogeochemical feedbacks, which may reduce or amplify the net impact of the anthropogenic forcing, are extremely important to the understanding

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and prediction of climate change (Lovelock et al., 1972). One possible negative biogeochemical feedback proposed by Charlson et al. (1987) involves the marine biosphere, the ocean and the atmosphere coupled through the marine biogenic sulfur compound Dimethylsulfide (DMS).

DMS produced from phytoplankton is the most abundant form in which the ocean releases gaseous sulfur. In the atmosphere, DMS is oxidized to sulfate particles that alter the amount of solar radiation reaching the Earth's surface both by directly scattering solar energy and indirectly by acting as cloud condensation nuclei (CCN), thereby affecting the cloud albedo. The change in cloud albedo results in global temperature perturbations potentially affecting the productivity of the marine biosphere and hence the concentration of oceanic DMS. The oceanic and atmospheric processes involved in this multistep feedback mechanism are complex. Fundamental gaps remain in our understanding of key issues in this feedback process, in particular with regard to the processes that regulate the DMS seawater concentration (Andreae and Crutzen, 1997; Liss et al., 1997). Traditionally, the DMS cycle in the atmosphere and in the ocean have been assessed independently. As a consequence, it has not been possible to assess the strength of the proposed feedback or even to anticipate if global warming will result in an increase or decrease of DMS emissions. However, significant progress has been made to understand many of the included mechanisms.

Enzymatic decomposition of Dimethylsulfonium Propionate (DMSP) is the primary source of DMS in seawater with DMSP being released from phytoplankton by a variety of mechanisms. The biological function of DMSP, and especially of its degradation to DMS, is still not clear. It has been suggested that the physiological function of DMSP is related to maintaining intercellular osmotic pressure (Vairavamurthy et al., 1985). Other suggested physiological functions of DMSP in marine algae are that it may act as a cryoprotectant (Kirst et al., 1991; Stefels, 2000) and serve as an antioxidant system (Sunda et al., 2002).

DMSP is widespread among taxa but seems to be particularly abundant in specific groups, including coccolithophorids like *Emiliania huxleyi*. Other groups, like diatoms,

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are generally poor producers of DMSP (Keller et al., 1989). Among other factors, DMSP release into the water is assumed to be controlled by phytoplankton senescence (Nguyen et al., 1990; Kwint and Kramer, 1995), viral lysis of phytoplankton cells (Malin et al., 1998) and zooplankton grazing (Dacey and Wakeham, 1986).

5 Consumption by bacteria is the major sink for DMS in seawater (Kiene and Bates, 1990; Dacey and Wakeham, 1986). Chemical oxidation of DMS to Dimethyl sulfoxide (DMSO) in seawater and ventilation to the atmosphere contribute only a minor part to the total DMS removal in seawater (e.g. Shooter and Brimblecombe, 1989; Kieber et al., 1996; Bates et al., 1994; Gabric et al., 1993).

10 DMS sea-air fluxes may vary by orders of magnitude in space and time. Although there are no direct means of measuring DMS sea-air flux, it can be estimated from the DMS sea surface concentration combined with an empirically determined exchange rate. Parameterization of DMS sea-air exchange rates have been investigated in several studies (e.g. Wanninkhof, 1992; Wanninkhof and McGillis, 1999; Liss and Merlivat, 15 1986; Nightingale et al., 2000; McGillis et al., 2000; Zemmeling et al., 2002). According to Nightingale et al. (2000), the uncertainties associated with the exchange rate are approximately 50%.

Many aspects of the DMS oxidation in the atmosphere are poorly understood (e.g. Campolongo et al., 1999; Andreae and Crutzen, 1997). It is well-established that OH 20 reacts with DMS and that SO<sub>2</sub> and methane sulfonic acid (MSA) are among the major reaction products (e.g. Capaldo and Pandis, 1997). It is also known that NO<sub>3</sub> and halogen radicals (e.g. BrO, IO, ClO) react with DMS in the atmosphere, but the importance of these reactions is even less well known (Sayin and McKee, 2004).

25 A key link in the proposed feedback between DMS and climate is the nucleation of DMS-derived sulfuric acid into new particles and eventually the formation of new cloud-forming particles (Charlson et al., 1987). The nucleation ability of DMS-derived sulfuric acid is still under debate (Yoon and Brimblecombe, 2002). Measurements over the South Atlantic, at Cape Grim, and at Amsterdam Island show a strong correlation between DMS emission and the concentration of total aerosol particles and CCN

(Putaud et al., 1993; Andreae et al., 1995, 1999; Ayers and Gillett, 2000). However, such findings are by no means generally applicable. Several investigators found no or only sporadic correlation between DMS and non-sea-salt sulfate, CN or CCN (Bates et al., 1992; Berresheim et al., 1993).

5 DMS and its oxidation products in the atmosphere are short-lived species. Therefore, it is necessary to resolve the temporal and spatial distribution of the DMS sea surface concentration on a global scale to investigate its impact on the climate system. Several studies attempted to build climatologies of the global distribution of DMS in the sea surface water. Belviso et al. (2004a) recently compared seven global DMS monthly climatologies (Kettle et al., 1999; Kettle and Andreae, 2000; Anderson et al., 2001; Aumont et al., 2002; Simo and Dachs, 2002; Chu et al., 2003; Belviso et al., 2004b). For the zonal and annual mean they found differences ranging from 50% in the tropics to 100% in high latitudes. The studies of Kettle et al. (1999); Kettle and Andreae (2000); Anderson et al. (2001); Simo and Dachs (2002) all rely on the Kettle et al. (1999) database which consists of almost 16 000 DMS sea surface measurements. Kettle et al. (1999) and Kettle and Andreae (2000) derived monthly mean maps by a compilation of the measurements included in the database. Anderson et al. (2001) and Simo and Dachs (2002) used only the data points from the Kettle et al. (1999) database with concurrent chlorophyll  $\alpha$  and DMS sea surface measurements and extended the resulting database by climatological information about incoming light and nutrient abundance or by information about the mixed layer depth (MLD), respectively. From the extended database they derived nonlinear relationships to predict DMS sea surface concentrations. Aumont et al. (2002) and Belviso et al. (2004b) used a prognostic nonlinear parameterization to compute DMS sea surface concentrations from chlorophyll  $\alpha$  concentrations together with an index of community structure of marine phytoplankton derived from measurements taken at several DMS surveys. Only the approach of Chu et al. (2003) uses a prognostic biogeochemical formulation for DMS production and DMS removal in the ocean based on the regional work of Gabric et al. (1993).

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Several atmospheric modeling studies simulate the DMS gas exchange between atmosphere and ocean based on prescribed DMS sea surface concentrations. The Kettle and Andreae (2000) climatology of the DMS seawater concentration is widely used in global atmospheric models (e.g. Boucher and Pham, 2002; Jones et al., 2001; Berglen et al., 2004; Gondwe et al., 2003; Rotstayn and Lohmann, 2002). Up to now, none of the global model studies include a description of the DMS cycle in the ocean. The response of the DMS emission to climate change could therefore only be assessed through changes in the sea-air exchange rate which varies with wind speed and temperature. Penner et al. (2001) showed a small increase in simulated DMS emissions between 2000 and 2100 (from 26.0 Tg(S)/yr to 27.7 Tg(S)/yr) using a constant DMS sea surface concentration field (Kettle et al., 1999) combined with a constant monthly climatological ice cover. The gas exchange rates were calculated interactively in the simulation based on wind speed and sea surface temperature. However, DMS sea surface concentrations are controlled by marine biology which is affected by climate variables such as solar irradiance, temperature and ocean physics. These variables are likely to change under changing climate conditions.

Changes in climate will lead to changes in the emission of other components that are linked to the DMS cycle, e.g. sea salt aerosols (Gong and Barrie, 2003), emissions associated with organic aerosols (O'Dowd et al., 1999) and dust emissions (Tegen et al., 2004). Stier et al. (2004)<sup>1</sup> showed in a global microphysical aerosol modeling study that specific emission changes cause changes in aerosol cycles of other components confirming a microphysical coupling between the different aerosol cycles. To account for these effects, the DMS-climate feedback has to be studied as a part of the complex global aerosol system.

The aim of this work is to study the DMS cycle in the ocean and in the atmosphere by setting up a comprehensive global model system including the ocean, the marine

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<sup>1</sup>Stier, P., Feichter, J., Kloster, S., Vignati, E., and Wilson, J.: Emission-Induced Nonlinearities in the Global Aerosol System – Results From the ECHAM5-HAM Aerosol-Climate Model, J. Clim., submitted, 2004.

biogeochemistry, the atmosphere and the aerosol system. The marine biogeochemistry model has been extended by a formulation of the DMS cycle in the ocean. The simulated DMS flux into the atmosphere is passed to the aerosol model included in the atmosphere model to calculate concentration of DMS, SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> in the atmosphere. To our knowledge, this is the first simulation of the DMS cycle in the ocean and in the atmosphere in a consistent coupled model setup. To assess the role of DMS in the climate system, it is essential to treat the DMS cycle interactively in the ocean-atmosphere system, as the proposed DMS-climate feedback is a multi-compartment feedback. Furthermore, the treatment of the DMS cycle in the ocean and in the atmosphere allows an evaluation using measurements conducted in the atmosphere and in the ocean. The lack of DMS measurements is one of the major limitations for the understanding of the DMS-climate feedback. The combination of oceanic and atmospheric measurements will give further insights to the shortcomings of the model.

## 2. Model description

The model used in the experiment is a coupled atmosphere-ocean general circulation model (AOGCM). The AOGCM consists of sub-models which correspond to the atmosphere (ECHAM5) and the ocean (MPI-OM). The atmospheric model includes a microphysical aerosol model (HAM) which simulates the evolution of an ensemble of microphysically interacting internally- and externally mixed aerosol populations as well as their size-distribution and composition. Embedded in the ocean model is a marine biogeochemistry model (HAMOCC5) which has been extended by a formulation of the DMS cycle in the ocean. The single model components are briefly described in the following sections.

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## 2.1. The MPI-OM general circulation model

The ocean component is the Max-Planck-Institute ocean model (MPI-OM) (Marsland et al., 2003). MPI-OM is a z-coordinate global general circulation model based on the primitive equations for a Boussinesq-fluid on a rotating sphere. The transport is computed with a total variation diminishing (TVD) scheme (Sweby, 1984). It includes parameterizations of sub grid scale mixing processes like isopycnal diffusion of the thermohaline fields, eddy induced tracer transport following Gent et al. (1995), and a bottom boundary slope convection scheme. The model contains a free surface and a state of the art sea ice model with viscous-plastic rheology and snow (Hibler, 1979). The model works on a curvilinear orthogonal C-grid. In this study, we use a nominal resolution of 1.5° at the equator with one pole located over Greenland and the other over Antarctica. In the vertical, the model has 40 levels with level thickness increasing with depth. 8 layers are located within the upper 90 m and 20 layers within the upper 600 m.

## 2.2. The marine biogeochemistry model HAMOCC5

The marine biogeochemistry component is the Hamburg Oceanic Carbon Cycle Model (HAMOCC5) (Maier-Reimer, 1993; Six and Maier-Reimer, 1996; Wetzel, 2004; Maier-Reimer et al., 2005). HAMOCC5 simulates the biogeochemical tracers in the oceanic water column and the sediment. The model is coupled online to the circulation and diffusion of the MPI-OM ocean model and runs with the same time step and resolution. The eco-system model is based on nutrients, phytoplankton, zooplankton and detritus (NPZD-type) as described in Six and Maier-Reimer (1996). In addition, new elements like nitrogen, dissolved iron and dust are accounted for and new processes like denitrification, nitrogen fixation, dissolved iron uptake and release by biogenic particles, and dust deposition and sinking are implemented as described in detail in Wetzel (2004). The dust deposition to the ocean surface is calculated online in the ECHAM5-HAM submodel and passed to the marine biogeochemistry model HAMOCC5 once per day.

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Bioavailable iron is released in the surface layer immediately from the freshly deposited dust.

### 2.2.1. DMS formulation in the marine biogeochemistry module HAMOCC5

The DMS formulation in HAMOCC5 is derived from the formulation originally developed for a former version of the marine biogeochemistry module (HAMOCC3.1 Six and Maier-Reimer, 1996) (Six and Maier-Reimer, in prep.).

The formulation for DMS production in the ocean assumes that DMS is produced ( $DMS_{prod}$ ) when phytoplankton cells are destroyed due to senescence or grazing processes. The DMS decay occurs via consumption by bacteria ( $DMS_{bac}$ ), chemical oxidation to DMSO ( $DMS_{UV}$ ) and ventilation to the atmosphere ( $DMS_{flux}$ ).

$$\frac{d[DMS]}{dt} = DMS_{prod} - DMS_{bac} - DMS_{UV} - DMS_{flux} \quad (1)$$

Here only dissolved DMS is considered. DMSP as the precursor of DMS in the ocean is not taken into account explicitly because very little is known about the actual processes that lead to the reduction of DMSP to DMS. Kiene (1992) concluded from estuarine experiments where less than 30% of the DMSP was converted to DMS that DMS is not the major transformation product of DMSP, presumably owing to an alternative demethylation pathway. Kiene and Linn (2000) hypothesized that the sulfur-demand of bacteria determines the proportion on which DMSP is processed through this alternative demethylation pathway, rather than being converted by enzymatic degradation to DMS. However, these findings are not yet satisfactorily understood to be incorporated into a global model study. Moreover, only few measurements of DMSP concentrations in the ocean are available which makes an evaluation not feasible.

HAMOCC5 distinguishes between silicate and calcium carbonate export ( $export_{sil}$  and  $export_{CaCO_3}$ , respectively). Therefore, it separates indirectly between the two phytoplankton groups the diatoms which form opal frustels, and the coccolithophorids which build skeletons made of calcium carbonate. Fast growing diatoms consume

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nutrients as long as silicate is available. After depletion of silicate the phytoplankton growth is carried out by coccolithophorids. The two phytoplankton groups are known to differ markedly in terms of their intercellular DMSP content, and hence their ability for producing DMS (Keller et al., 1989). DMS production and consumption processes are just considered in the uppermost 100 m of the water column (euphotic zone). The DMS production is formulated as follows:

$$DMS_{prod} = f(T) * (k_{psi} * export_{sil} + k_{pcc} * export_{CaCO_3}) \quad (2)$$

$k_{psi}$  and  $k_{pcc}$  are the respective scaling factors defined in Table 1. The function  $f(T)$  accounts for the observed temperature dependence of intercellular DMSP concentrations. Under low temperature conditions, e.g. in polar regions, the DMSP content in phytoplankton cells is higher than under temperate conditions (Baumann et al., 1994). This effect is parameterized as follows:

$$f(T) = \left(1 + \frac{1}{(T + k_{pt})^2}\right) \quad (3)$$

with  $T$  in °C,  $k_{pt}$  scales the temperature dependency (Table 1).

The DMS destruction processes are formulated as follows:

The destruction of DMS by photo-oxidation to DMSO depends on the solar radiation at the surface (Shooter and Brimblecombe, 1989; Brimblecombe and Shooter, 1986; Kieber et al., 1996). The incident solar radiation  $I_0$  is attenuated in HAMOCC5 by water and phytoplankton as a function of depth ( $z$ ) according to the equation:

$$I_z = I_0 * e^{(k_w + k_{chl}) * z} \quad (4)$$

The attenuation coefficient for pure water is chosen to be  $k_w = 0.04 \text{ m}^{-1}$ . Light attenuation by phytoplankton is assumed to be a linear function of the chlorophyll concentration  $k_{chl} = 0.03 [\text{CHL}] \text{ m}^{-1}$ , with the chlorophyll concentration  $[\text{CHL}]$  given in  $\text{mg l}^{-1}$ . The chlorophyll concentration is computed from the modeled phytoplankton concentration with a fixed chlorophyll to carbon ratio of 1:80. The incident surface irradiance ( $\text{Wm}^{-2}$ )

is calculated interactively, including the effects of clouds and aerosols, in the ECHAM5-HAM model. The decay of DMS by photo-oxidation is then formulated as follows:

$$DMS_{UV} = k_{IUV} * I_z * [DMS] \quad (5)$$

$k_{IUV}$  is the respective scaling factor defined in Table 1. DMS destruction due to consumption by bacteria is assumed to be temperature dependent:

$$DMS_{bac} = k_{Ib} * (T + 3.) * [DMS] * f([DMS]) \quad (6)$$

with  $T$  in °C.  $k_{Ib}$  denotes the scaling factor for the consumption process by bacteria. Observations suggest that consumption by bacteria appears to be less efficient in oligotrophic areas with low DMS concentrations. We parameterize this variation with a saturation function:

$$f([DMS]) = \left( \frac{DMS}{k_{cb} + [DMS]} \right) \quad (7)$$

$k_{cb}$  is set to 10 nmol/l which ensures an almost linear behavior for low DMS concentrations.

For the atmosphere, the most important DMS loss mechanism in seawater is the loss due to sea-air exchange. For the sea-air exchange calculation we neglect the DMS concentration in the atmosphere, as it is small compared to the DMS sea surface concentration, and formulate the DMS sea-air exchange as:

$$[DMS]_{flux} = k_{sea-air} * [DMS] \quad (8)$$

$k_{sea-air}$  denotes the sea-air exchange rate. We choose the formulation after Waninkhof (1992):

$$k_{sea-air} = 0.31 * w_{10m}^2 * \left( \frac{SC}{660} \right)^{-\frac{1}{2}} \quad (9)$$

where  $w_{10}$  is the 10 m wind speed and  $SC$  the Schmidt number for DMS which is calculated analogous to Saltzman et al. (1993).

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[Kettle and Andreae \(2000\)](#) compiled a database of almost 16 000 DMS sea surface concentration measurements. Thereby, the original database ([Kettle et al., 1999](#)) was updated by new measurements. We utilized the updated database for an optimization of the parameters in the proposed DMS formulation. Therefore, the data points are distributed onto the ocean grid cells on a monthly mean basis. The DMS grid value is taken to be the average of the individual monthly mean measurements within the grid cell. Since the data coverage is very sparse and the partitioning into monthly means is rather arbitrary, we extrapolated the resulting DMS sea surface concentration for a single grid box to the adjacent grid boxes and also took values from the adjacent months into account for the monthly splitting. Due to computational constraints it is not feasible to conduct the optimization process within the full coupled AOGCM. To derive the DMS scaling parameters, we use an offline version of the ocean model (MPI-OM/HAMOCC5) forced by NCEP/NCAR reanalysis data ([Kalnay et al., 1996](#)). The model setup is described in detail in [Wetzel et al. \(2005\)](#). From this simulation we arbitrarily choose the year 1995 for the optimization process of the DMS scaling parameters. Periodically repeating the simulation with NCEP/NCAR forcing fields for the year 1995, we calculate global value deviation fields of the modeled DMS sea surface concentration and the DMS sea surface map, generated from the [Kettle and Andreae \(2000\)](#) database. Thereby, we take only ocean grid boxes into account with an ocean depth greater than 300 m. Regions with a shallower depth, like the North Sea, are not well captured by the model. In these regions the [Kettle and Andreae \(2000\)](#) database includes a disproportional high number of measurements. To avoid a bias in the optimization process towards these measurements, these grid boxes are excluded. The DMS scaling parameters ( $k_{psi}$ ,  $k_{pcc}$ ,  $k_{pt}$ ,  $k_{luv}$ ,  $k_{lb}$ ) are systematically adjusted after every second model year to minimize the global deviation from the [Kettle and Andreae \(2000\)](#) database. The resulting parameters are compiled in Table 1.

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### 2.3. The ECHAM5 general circulation model

The atmospheric component is the ECHAM5 model (Roeckner et al., 2003) with the current standard resolution of 31 vertical levels on a hybrid sigma-pressure coordinate system up to a pressure level of 10 hPa. Prognostic variables are vorticity, divergence, surface pressure, temperature, water vapor, cloud water and cloud ice. Except for the water and chemical components, the prognostic variables are represented by spherical harmonics with triangular truncation at wavenumber 63 (T63). Physical processes and nonlinear terms are calculated on a Gaussian grid with a nominal resolution of 1.8° in longitude and latitude. For the advection of water vapor, cloud liquid water, cloud ice and tracer components, a flux form semi-Lagrangian transport scheme (Lin and Rood, 1996) is applied. Cumulus convection is based on the mass flux scheme after Tiedtke (1989) with modifications according to Nordeng (1994). The cloud microphysical scheme (Lohmann and Roeckner, 1996) consists of prognostic equations for cloud liquid water and cloud ice. The cloud cover is predicted with a statistical scheme including prognostic equations for the distribution moments (Tompkins, 2002). The transfer of solar radiation is parameterized after Fouquart and Bonnel (1980) and the transfer of longwave radiation after Morcrette et al. (1998).

### 2.4. The HAM aerosol model

The ECHAM5 model has been extended by a complex microphysical aerosol model HAM, described in detail in Stier et al. (2005). The aerosol spectrum is represented by the superposition of seven log-normal distributions. These seven aerosol modes can be described by three moments: the aerosol number, the number median radius, and the standard deviation. The seven modes are divided into four geometrical size classes (nucleation, Aitken, accumulation and coarse mode). Three of the modes include only insoluble compounds, four of the modes contain at least one soluble compound. Aerosol compounds considered are sulfate (SU), black carbon (BC), particulate organic mass (POM), sea salt (SSA), and mineral dust (DU). HAM consists of a

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microphysical core M7, an emission module, a sulfur chemistry scheme, a deposition module, and a radiation module defining the aerosol radiative properties.

The microphysical core M7 (Vignati et al., 2004) treats the aerosol dynamics and thermodynamics. Processes considered are coagulation among the modes, condensation of gas-phase sulfuric acid on the aerosol surface, the binary nucleation of sulfate, and water uptake.

The emission of mineral dust and sea salt is calculated interactively according to the scheme of Tegen et al. (2002) and Schulz et al. (2004), respectively. DMS emissions are calculated online in the marine biosphere submodel HAMOCC5. For the other aerosol compounds, emission strengths, emission size distribution and emission height are based on the AEROCOM (Aerosol Model Inter-Comparison, <http://nansen.ipsl.jussieu.fr/AEROCOM>) emission inventory for the year 2000 (Frank Dentener, in prep.). The emission strength for all aerosol compounds is summarized in Table 2.

The sulfur chemistry module (Feichter et al., 1996) treats DMS, SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> as prognostic variables. In the gas phase, SO<sub>2</sub> and DMS are oxidized by hydroxyl (OH) during the day. Additionally, DMS reacts with nitrate radicals (NO<sub>3</sub>) at night. Reaction products are SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>. Dissolution of SO<sub>2</sub> within cloud water is calculated according to Henry's law. In the aqueous phase, the oxidation of SO<sub>2</sub> by hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and ozone (O<sub>3</sub>) are considered. The oxidant concentrations are prescribed as three dimensional monthly mean fields from calculations of the MOZART chemical transport model (Horowitz et al., 2003). Gas phase produced sulfate is allowed to condensate onto pre-existing particles or to nucleate to new particles, calculated by the aerosol microphysical module M7. In-cloud produced sulfate is distributed to the available pre-existing accumulation mode and coarse mode aerosol particles according to their respective number concentration. The deposition processes, i.e. wet deposition, dry deposition, and sedimentation, are calculated online in dependence of aerosol size and composition.

The radiation module calculates the aerosol optical properties explicitly from the

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prognosed size distribution, composition and mixing state which are then passed to the radiation scheme of ECHAM5. Only the effects of the aerosols on the solar part of the spectrum are considered.

For this study the ECHAM5-HAM model has been extended by a technique to mark  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  attributable to DMS. This allows to isolate the fraction of DMS-derived  $\text{SO}_2$  and  $\text{SO}_4^{2-}$ . Such a quantification facilitates to assess the importance of aerosols of DMS origin. Additionally, the knowledge of the contribution of DMS to  $\text{SO}_4^{2-}$  enables us to use  $\text{SO}_4^{2-}$  concentration measurements at sites with a relatively high contribution of DMS to  $\text{SO}_4^{2-}$  for an evaluation of the atmospheric DMS cycle in the model.

## 2.5. Model setup

The ocean and the atmosphere models are coupled quasi-synchronously with the OASIS coupler (Valcke et al., 2003) with a coupling time step of one day. The ocean model MPI-OM passes the sea surface temperature and sea ice variables to the atmosphere through OASIS. The atmosphere model ECHAM5-HAM uses these boundary conditions for one coupling timestep and transfers the surface forcing fields through OASIS back to the ocean model. Required surface forcing fields are heat, freshwater and momentum fluxes, downward solar radiation and the 10 m wind speed. Additionally the DMS flux to the atmosphere calculated in HAMOCC5 is passed to the atmosphere model and the dust deposition calculated in the HAM model is passed to HAMOCC5 through OASIS. The model does not employ flux adjustments.

In order to initialize the coupled atmosphere-ocean AOGCM, the uncoupled ocean model was integrated over thousand years to reach quasi-equilibrium state. From there on the coupled AOGCM was integrated with fixed external forcing to reach quasi-equilibrium state. From these initial conditions the simulation is started and integrated for 15 years. The results presented here are averaged over the last 10 years.

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### 3. Results

#### 3.1. Ocean

A detailed description of the simulated ocean and biochemical mean state of MPI-OM/HAMOCC5 is given in [Wetzel \(2004\)](#). In this study, the dust deposition is calculated interactively within the atmospheric model ECHAM5-HAM and differs therefore from the monthly mean offline fields used by [Wetzel \(2004\)](#). Assuming an iron content in dust of 3.5% implies an annual global mean iron deposition flux of 666 Gmol(Fe)/yr, 204 Gmol(Fe)/yr are deposited to the ocean surface. The iron deposition onto the ocean surface lies within the range used in other global marine biogeochemistry studies ([Fung et al. \(2000\)](#): 118 Gmol(Fe)/yr, [Aumont et al. \(2003\)](#): 149.7 Gmol(Fe)/yr, [Archer and Johnson \(2000\)](#): 131.7–487.4 Gmol(Fe)/yr). The wide range given in the global iron deposition rates highlights the uncertainties in the iron content of dust particles, as in the magnitude and the size variation of the dust emission and deposition in the atmosphere. However, the possibility to use interactively calculated dust depositions as input for the marine biogeochemistry provides the means to include effects of varying dust depositions caused by climate change on the marine biogeochemistry and on the DMS sea surface concentration.

In the following section we will focus on the simulated DMS sea surface concentrations and compare our results to measurements as well as to a recently developed DMS algorithm by [Simo and Dachs \(2002\)](#).

##### 3.1.1. DMS in the ocean

HAMOCC5 simulates a global total DMS production of 351 Tg(S)/yr. The loss of DMS in the ocean proceeds mainly via the consumption by bacteria (294 Tg(S)/yr). The DMS flux into the atmosphere (28 Tg(S)/yr) accounts for 8% of the global DMS removal in the ocean, the photo-oxidation (31 Tg(S)/yr) for 9%. The relative rates of the decay processes are not well constrained through measurements, in particular on a global

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scale. Archer et al. (2002) found a DMS flux equivalent to 10% of the DMS production in a six day Lagrangian experiment conducted in the northern North Sea. DMS removal by bacterial consumption accounted for the majority of the DMS removal (62–82% for surface levels and 98% for subsurface levels). Similar findings are reported for the Eastern Tropical Pacific where measurements show a 3 to 430 times faster removal of DMS by biological consumption than by the DMS flux into the atmosphere (Kiene and Bates, 1990) and for the North East Pacific where biological consumption accounted for 67% of the total DMS consumption and the DMS flux accounted for only a small fraction (1%) of the DMS loss (Bates et al., 1994). The simulated annual mean decay rates are in accordance with these findings. However, for an evaluation of the simulated production and decay processes more measurements are needed.

The simulated annual mean sea surface concentration of DMS is shown in Fig. 1. The predicted DMS concentrations show high values, generally exceeding 3–4 nmol/l, in the biological active upwelling zones like the equatorial Pacific Ocean, or in the upwellings off Peru and Angola. The subtropical gyres in both hemispheres show very low DMS sea surface concentrations. The polar oceans (North Pacific, North Atlantic and Southern Ocean) feature high DMS concentration with values up to 8 nmol/l in the Southern Ocean. In the Kettle and Andreae (2000) database measurement points in these areas are typically representative for the summer months. Therefore the model results might be biased towards too high annual mean values due to a sampling bias in these areas.

The predicted annual global mean DMS sea surface concentration lies with 1.8 nmol/l within the range of annual mean concentrations from DMS climatologies (Kettle et al., 1999: 2.1 nmol/l; Kettle and Andreae, 2000: 2.0 nmol/l; Anderson et al., 2001: 2.6 nmol/l; Aumont et al., 2002: 1.7 nmol/l; Simo and Dachs, 2002: 2.3 nmol/l; Chu et al., 2003: 1.5 nmol/l; Belviso et al., 2004b: 1.6 nmol/l, global numbers taken from Belviso et al., 2004a).

To evaluate the simulated DMS sea surface distribution, we compare the simulated DMS sea surface concentrations with the measurements given in the Kettle and An-

5 dreae (2000) database. Like Kettle et al. (1999), we utilize the concept of dividing the ocean into biogeochemical provinces as introduced by Longhurst et al. (1995). The data points of the Kettle and Andreae (2000) database were distributed onto the model grid on a monthly mean basis. The averaged values for each province together with the corresponding simulated mean values are listed in Table 5. Figure 2a shows the comparison in form of a scatter plot. For 50 out of the 57 biogeochemical provinces defined by Longhurst et al. (1995) measurements are available in the database. Out of these 50 provinces, 34 (68%) show an agreement within a factor of 2 between averaged measurements and the averaged simulation. Underestimated values are simulated for provinces in the Pacific Ocean, Atlantic Ocean and Indian Ocean, mainly in the Northern Hemisphere. For the Pacific Ocean 6 samples out of total of 20 samples differ more than a factor 2. For the Atlantic Ocean the agreement is similar, 7 out of a total of 21 provinces differ more than a factor of 2. Provinces in the Southern Ocean show a good agreement. Only a few provinces show simulated values higher than a factor of 2 compared to measurements, for example the Western Tropical Atlantic Province or the Caribbean Provinces. However, despite the large number of conducted measurements many provinces contain only few measurements, for example those in the Indian Ocean. Since the Kettle and Andreae (2000) database was used for the optimization of the model parameters the comparison might be misleading. For an independent evaluation we compare the simulation with the updated version of the Kettle and Andreae (2000) database (Global Surface Seawater Dimethylsulfide (DMS) Database, available at <http://saga.pmel.noaa.gov/dms/>) which has been extended by additional 12,866 DMS sea surface measurements by 10 different measurement campaigns. Compared to the Kettle and Andreae (2000) database the data coverage of the additional measurements is sparse. By gridding the measurement data points onto the model grid, only 418 grid boxes are assigned to an annual mean DMS sea surface concentration value, whereby the Kettle and Andreae (2000) data points cover 1641 grid boxes. Therefore only 32 provinces out of the total 57 provinces contain measurement values. Figure 2b shows the scatter plot only using the additional measurements.

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Table 6 lists the provinces together with the simulated and measured mean values. 20 provinces (63%) are within a factor of two in agreement with the measurements. There are no additional measurements in the Southern Ocean. The provinces in the Atlantic Ocean show a rather constant measured DMS sea surface concentration between 1 and 2 nmol/l. The simulation shows strong spatial variability for the Atlantic Ocean, especially in the Northern Hemisphere. The simulated mean values for the Pacific Ocean are in a good agreement with the measurements. Only 1 province out of a total of 13 provinces in the Pacific Ocean differs more than a factor of 2. In the Indian Ocean additional measurements were made in the Indian Monsoon Gyres Province which was rather undersampled in the original database. Here the simulations are in a excellent agreement with the measurements (2.93 compared to 2.92 nmol/l, respectively). For the East African Coastal Province the original database shows very high DMS sea surface concentrations (7.83 nmol/l) which were not captured by the simulation. The additional measurements do not confirm these high values. The averaged value for the province is for the additional measurements data 2.38 nmol/l which is in agreement with the simulation (2.01 nmol/l).

Monthly latitudinal profiles of the model results, the Kettle and Andreae (2000) database data and the DMS sea surface climatology from Kettle and Andreae (2000) are compared in Fig. 3. Again, only data points where the ocean depth is above 300 m are used. As DMS is a product of marine biological activity, the DMS sea surface concentration has large seasonal variations. This is especially pronounced in the high latitudes where DMS concentrations peak in the Southern Hemisphere in December and in the Northern Hemisphere in June. The amplitude of the seasonal variation is lower in the model than in the Kettle and Andreae (2000) climatology. However, the climatology is based only on a few data points in this region. Around the equator the modeled DMS sea surface concentrations stay almost constant with 2–3 nmol/l throughout the year. This value is confirmed by measurements in these latitudes and present as well in the Kettle and Andreae (2000) climatology. Overall the model simulates the observed DMS sea surface concentrations reasonably well.

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### 3.1.2. DMS concentration predicted from mixed layer depth and chlorophyll $\alpha$

[Simo and Dachs \(2002\)](#) developed a two equation algorithm to predict DMS sea surface concentrations using chlorophyll  $\alpha$  surface concentration (CHL) and the mixed layer depth (MLD). Our model setup allows to apply the proposed algorithm to the simulated MLD and sea surface chlorophyll  $\alpha$  concentration. We compare the resulting DMS sea surface concentration to the one given in [Simo and Dachs \(2002\)](#) using climatological input fields and to the one simulated within the marine biogeochemistry model. The DMS algorithm is formulated by [Simo and Dachs \(2002\)](#) as follows:

$$DMS = -\ln(MLD) + 5.7, \quad CHL/MLD < 0.02 \quad (10)$$

$$DMS = 55.8(CHL/MLD) + 0.6, \quad CHL/MLD \geq 0.02 \quad (11)$$

The units of MLD are m, of CHL are  $\text{mgm}^{-3}$ , and of DMS sea surface concentrations are  $\text{nmol/l}$ . The algorithm is based on the assumption of [Simo and Pedros-Alio \(1999\)](#) that vertical mixing plays a major role in controlling the production of DMS in the sea surface layers. They found that DMS is quantitatively related to the ratio of chlorophyll  $\alpha$  and MLD, leading to high DMS concentrations not only associated with high chlorophyll  $\alpha$  concentrations but also with moderate chlorophyll  $\alpha$  concentrations and a concurrent shallow MLD. They explained this relation with the fact that a shallow mixing tends to favor phytoplankton blooms of taxa with a high DMSP cell content. [Simo and Dachs \(2002\)](#) derived global monthly DMS sea surface concentrations using a global monthly climatology for the MLD (Samuel and Cox' GFDL Global Oceanographic Data Set Atlas, available at <http://dss.ucar.edu/datasets/ds279.0/>) and chlorophyll  $\alpha$  concentration from SeaWiFS averaged over the period September 1997 to November 2000. The MLD is equally defined by the density criterion as in our simulation (depth where  $\Delta\sigma_t=0.125$  relative to the surface). We applied the proposed relationship using the simulated MLD and chlorophyll  $\alpha$  concentration. In about 80% of the total ocean surface, the ratio CHL/MLD is  $<0.02$  and Eq. (10) applies. If the MLD exceeds 298 m, Eq. (10) results in a prediction of negative DMS values. This is the case for 9% of

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the total ocean surface. We excluded these values. A monthly mean MLD deeper than 298 m is simulated in the winter months in the North Atlantic and in the Southern Ocean. This is consistent with observations and also present in the Samuel and Cox MLD climatology, whereby here only 2% of the ocean surface shows a monthly mean MLD deeper than 298 m. Figure 4 shows the resulting zonal annual mean DMS sea surface concentrations compared to the concentration obtained from our simulation, the concentration from [Simo and Dachs \(2002\)](#) using climatological input fields and the concentration from the [Kettle and Andreae \(2000\)](#) climatology.

The DMS sea surface concentration derived from the [Simo and Dachs \(2002\)](#) algorithm with simulated MLD and chlorophyll  $\alpha$  concentration is comparable to the one derived by [Simo and Dachs \(2002\)](#) using a MLD climatology and chlorophyll  $\alpha$  concentrations from SeaWiFS. Discrepancies occur in the high latitudes where during the summer months high chlorophyll  $\alpha$  concentrations persist and predominantly Eq. (11) applies. The DMS sea surface concentration resulting from the simulated MLD and chlorophyll  $\alpha$  concentration is higher in the southern high latitudes and slightly lower in the northern high latitudes. The lower values in the northern high latitudes are caused by a simulated deeper mixed layer depth compared to the Samuel and Cox climatology. For the southern high latitudes high simulated chlorophyll  $\alpha$  concentrations lead to high DMS sea surface concentrations. The simulated chlorophyll  $\alpha$  concentrations are slightly higher than the satellite estimates and probably overestimated by the model ([Wetzel, 2004](#)). However, particularly in the high latitudes the climatological fields comprise large uncertainties which are caused by a sparse data coverage for the MLD and by frequent cloud contamination for the satellite derived chlorophyll  $\alpha$  concentration.

The simulated DMS sea surface concentration using the DMS formulation introduced in the marine biogeochemistry model HAMOCC5 results in distinct different zonal annual mean concentration variations compared to DMS sea surface concentration derived with the [Simo and Dachs \(2002\)](#) algorithm. Particularly in the Northern Hemisphere our simulation results in significantly lower DMS sea surface concentrations which are in agreement with the DMS sea surface climatology from [Kettle and An-](#)

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dreae (2000). Both show a minimum in the DMS sea surface concentration around 30° N which is not captured with the Simo and Dachs (2002) algorithm. Discrepancies are highest in the northern high latitudes. In the late summer, with moderate chlorophyll  $\alpha$  concentrations in the northern high latitudes, the Simo and Dachs (2002) algorithm predominately relies on the MLD only. This results in a broadening of the summer maximum leading to high annual mean DMS sea surface concentration in this region. The Kettle and Andreae (2000) climatology shows as the Simo and Dachs (2002) algorithm high DMS sea surface concentrations in the northern high latitudes. The high values in the climatology are probably caused by the inclusion of DMS measurements from the North Sea region. We excluded these datapoints for the optimization of our DMS formulation. The remaining measurements mainly show DMS sea surface concentrations around 1–2 nmol/l (see Fig. 3) which is consistent with our simulation.

### 3.2. DMS sea-air exchange

As described in Sect. 2.2.1 we use the DMS sea-air exchange parameterization after Wanninkhof (1992) utilizing the 10 m wind speed and the sea surface temperature provided by the atmospheric model part ECHAM5 to calculate the DMS flux interactively in the model. The resulting global annual mean DMS flux is 28 Tg(S)/yr. Estimates of the global DMS flux differ widely depending mainly on the used DMS sea surface climatology, sea-air exchange parameterization, and wind speed data, ranging from 16 Tg(S)/yr up to 54 Tg(S)/yr (see e.g. Kettle and Andreae, 2000, for a review). Kettle and Andreae (2000) calculated a global DMS flux between 27 and 32 Tg(S)/yr with their DMS sea surface climatology applying the same sea-air exchange parameterization (Wanninkhof, 1992) and four different combinations of data sets for the wind speed and sea surface temperature. Our simulated global annual mean DMS flux is in agreement with their findings. Figure 5 displays the global distribution of the annual mean DMS flux into the atmosphere. The distribution of the DMS flux is closely related to the DMS sea surface distribution (c.f. Fig. 1). High DMS fluxes persist in regions with high DMS sea surface concentrations, such as the equatorial Pacific and Atlantic Ocean

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and in high wind speed regions, e.g. the broad band with elevated DMS emissions in the Southern Ocean between 40° and 60° S.

### 3.3. The atmospheric sulfur cycle

#### 3.3.1. Global budgets

5 Table 3 summarizes the characteristics of the global DMS and sulfur budgets. Globally, the DMS emission flux is 28 Tg(S)/yr. The highest emissions are simulated in the Southern Hemisphere (17.6 Tg(S)/yr). Compared to the total SO<sub>2</sub> source resulting from SO<sub>2</sub> emissions from anthropogenic sources, wildfires and volcanic eruptions as prescribed from the AEROCOM emission scenario and SO<sub>2</sub> resulting from DMS oxidation, DMS accounts for 30% of the total sulfur source (14% in the Northern Hemisphere  
10 and 67% in the Southern Hemisphere). Globally, 84% of the DMS is removed via oxidation by OH radicals. This ratio is close to the one reported in other studies using similar DMS reaction mechanisms (Berglen et al. (2004): 73%, Chin et al. (2000): 88%, Pham et al. (1995): 86%). Oxidation by NO<sub>3</sub> is more important in the Northern Hemisphere (8.7%) than in the Southern Hemisphere (6.5%). The annual global mean DMS burden is with 0.08 Tg(S) in agreement with other studies (Chin et al., 2000: 0.07 Tg(S); Koch et al., 1999: 0.06 Tg(S); Pham et al., 1995: 0.05 Tg(S)) as is the lifetime with 1.0 days  
15 (Chin et al., 2000: 2.0 days; Koch et al., 1999: 1.9 days; Pham et al., 1995: 0.9 days). The chemical conversion is the major sink for SO<sub>2</sub> (78%). The dry deposition accounts for only 18% of the total removal. This is low compared to other studies, as already pointed out by Stier et al. (2005). The serial resistance dry deposition scheme used here results in significantly lower SO<sub>2</sub> dry deposition fluxes compared to other studies (Ganzeveld et al., 1998). The low dry deposition sink results in a high yield of SO<sub>4</sub><sup>2-</sup> from the chemical conversion of SO<sub>2</sub> into SO<sub>4</sub><sup>2-</sup>. SO<sub>4</sub><sup>2-</sup> is mainly wet deposited (95%).  
20 Dry deposition accounts for 3% and sedimentation for 2% of the total SO<sub>4</sub><sup>2-</sup> removal.

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### 3.3.2. DMS in the atmosphere

The annual global distribution of the DMS burden is displayed in Fig. 6. Highest burdens persist in the Southern Hemisphere, in particular around 60° S. This agrees with the high DMS flux simulated in this region. Additionally, DMS experiences a longer lifetime in the Southern Hemisphere compared to the Northern Hemisphere (1.15 days compared to 0.78 days, respectively). In the industrialized Northern Hemisphere, high NO<sub>3</sub> levels ensure a steady oxidation of DMS even during nighttime when OH concentrations are zero. About 88% of DMS emitted in the Southern Hemisphere is oxidized by OH and 12% by NO<sub>3</sub>. In the Northern Hemisphere, 77% is oxidized by OH and 23% by NO<sub>3</sub>. In the Southern Hemisphere, the limited removal of DMS via NO<sub>3</sub> due to the low NO<sub>3</sub> concentrations leads to an accumulation of DMS and a higher atmospheric DMS burden compared to the Northern Hemisphere (0.056 Tg(S) compared to 0.021 Tg(S), respectively).

In Fig. 7a simulated and observed seasonal variations of atmospheric DMS concentrations are compared at Amsterdam Island in the Southern Ocean (Sciare et al., 2000). Atmospheric DMS mixing ratios were measured on a daily basis from August 1990 to December 1999. Shown are monthly mean values. The simulated seasonal variation is in agreement with the observations. However, the summer maximum is overestimated and shifted by two months in the model. Highest concentrations are simulated in November (822 pptv), whereas the observations show a maximum in January (557 pptv). Figure 7b shows the simulated SO<sub>2</sub> concentration at Amsterdam Island compared to measurements reported by Putaud et al. (1992). The measurements were taken for the period March 1989 to January 1991. The SO<sub>2</sub> concentrations show a similar seasonal variation as the DMS concentrations, both in the simulation and in the measurements. The amplitude of the seasonal cycle which is defined by the ratio  $R = (\text{average concentration for December, January and February}) / (\text{average concentration for June, July and August})$ , is 8 for the measured as well as for the simulated SO<sub>2</sub> concentrations. The amplitude of the seasonal atmospheric DMS cycle is simulated

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equal to 9 and measured equal to 8 and is comparable to that of SO<sub>2</sub>. The seasonal variation of the atmospheric DMS concentration at Amsterdam Island is largely driven by variations in the DMS sea surface concentration. The seasonal amplitude of the sea surface DMS is simulated equal to 9 and fits exactly the simulated seasonal cycle for DMS in the atmosphere (data not shown).

### 3.3.3. DMS contribution to SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> column burdens

In order to quantify the importance of DMS-derived SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup>, we simulated the contribution of DMS-derived SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> to the total SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> concentration in the atmosphere. Additionally, this allows to utilize SO<sub>4</sub><sup>2-</sup> concentration measurements in regions with a high DMS contribution for an evaluation of the DMS cycle.

#### SO<sub>2</sub> column burden

The spatial distribution of the total SO<sub>2</sub> column burden, the SO<sub>2</sub> column burden resulting from DMS and the relative contribution of the DMS-derived SO<sub>2</sub> to the total SO<sub>2</sub> column burden are displayed in Fig. 8. Shown are annual averages, averages for June, July and August and for December, January and February. The same is shown for the SO<sub>4</sub><sup>2-</sup> column burden in Fig. 9. The respective global mean column burdens are summarized in Table 4.

The global distribution of SO<sub>2</sub> column burdens resulting from all sources reflects the dominant anthropogenic sulfur sources in the Northern Hemisphere, most pronounced over the industrialized areas of Europe, North America and China. The SO<sub>2</sub> column burden resulting from DMS emission alone highlights the strong seasonal variation of DMS in the atmosphere. The highest column burdens persist in the Southern Hemisphere for December, January and February with values up to 1 mg(S)m<sup>-2</sup> in high latitudes. The maximum in the Northern Hemisphere for June, July and August is less pronounced. In the equatorial regions, the SO<sub>2</sub> column burden attributable to DMS stays almost constant throughout the year. The simulated high DMS sea surface con-

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centration here causes a steady emission of DMS into the atmosphere and therefore a high load of  $\text{SO}_2$  derived from DMS integrated over the atmospheric column. The relative contribution of DMS-derived  $\text{SO}_2$  to the total  $\text{SO}_2$  shows clearly the overwhelming role of DMS in the Southern Hemisphere during the biological active season. In December, January and February the contribution is up to 90%.

The simulated global annual burden of  $\text{SO}_2$  is 0.60 Tg(S), 25 % of which can be attributed to DMS. Compared to the DMS contribution to the total sulfur emissions source, which is globally 30%, the contribution of DMS to the  $\text{SO}_2$  column burden is reduced. The contribution is greatest in the Southern Hemisphere (44%). The anthropogenic sources are dominant in the Northern Hemisphere. DMS accounts for 16% of the total  $\text{SO}_2$  column burden on the annual mean in the Northern Hemisphere. Even in the summer months (June, July and August), when maximum DMS sea surface concentrations are simulated in the Northern Hemisphere, the contribution of DMS to the total  $\text{SO}_2$  column burden is greatest in the Southern Hemisphere (32% compared to 17% in the Northern Hemisphere).

### $\text{SO}_4^{2-}$ column burden

The global distribution of the  $\text{SO}_4^{2-}$  column burden resulting from all sources shows highest burdens close to the main sources and the sources of its precursor  $\text{SO}_2$  in the Northern Hemisphere (Fig. 9). Significant export to low emission regions, for example the Middle East, North Africa, the North Pacific and the North Atlantic, occurs throughout the year. The  $\text{SO}_4^{2-}$  column burden resulting from DMS emissions alone shows almost the same distribution as the  $\text{SO}_2$  column burden resulting from DMS. A high burden persists in the Southern Hemisphere in the summer season. The export of  $\text{SO}_4^{2-}$  into low emission regions cause a slight southward shift of the areas significantly influenced by DMS emissions. While for the  $\text{SO}_2$  burden a contribution of DMS of 60 to 70% for December, January and February is simulated in the equatorial regions of the Pacific and Atlantic, the contribution for  $\text{SO}_4^{2-}$  lies only between 40 to 50%.

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The global annually averaged column burden of  $\text{SO}_4^{2-}$  is 0.73 Tg(S) with a DMS contribution of 27% globally. The greatest contribution occurs in December, January and February with up to 37% globally and 57% in the Southern Hemisphere. The DMS contribution to the  $\text{SO}_4^{2-}$  burden lies within the same range as the DMS contribution to the  $\text{SO}_2$  column burden. This is caused by the simulated strong chemical conversion rate of  $\text{SO}_2$  to  $\text{SO}_4^{2-}$ .

### 3.3.4. DMS-derived $\text{SO}_4^{2-}$ in the atmosphere

The global distribution of the fraction of the  $\text{SO}_4^{2-}$  burden attributable to DMS shows the dominant role of DMS as  $\text{SO}_4^{2-}$  precursor in the Southern Hemisphere, in particular at high latitudes (Fig. 9). A comparison of the simulated  $\text{SO}_4^{2-}$  concentrations with measurements in these remote regions therefore gives an indication of the representation of the DMS cycle in the model simulation. Several measurement networks include  $\text{SO}_4^{2-}$  surface concentration measurements. We choose the multi-annual measurements from the University of Miami network (D. Savoie, pers. comm.), as this network includes mainly measurements from remote sites. These measurements have been conducted mainly on islands or at coastal stations. The number of measurement years varies with site. The measurement locations are displayed in Fig. 10. Red points indicate stations with a simulated annual contribution of DMS to  $\text{SO}_4^{2-}$  of more than 50% within the lowest model layer. These stations are mainly located in the Southern Hemisphere.

Figure 11 shows the scatter of the measured and simulated  $\text{SO}_4^{2-}$  surface concentration. Shown are monthly, annual, December, January and February, and June, July and August mean values. In addition, Table 7 lists the annual mean values for all measurement locations together with the relative contribution of DMS to  $\text{SO}_4^{2-}$  in the lowest model layer. Out of a total of 320 monthly mean samples, 213 (67%) agree within a factor of 2 with the measurements. The model overestimates the  $\text{SO}_4^{2-}$  surface concentrations especially for low concentrations. The lowest values reported coincide with

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locations where anthropogenic influences are low and where therefore the DMS contribution to  $\text{SO}_4^{2-}$  generally exceeds 50%. On the annual mean, 15 stations show a contribution of DMS greater than 50%. 9 of these stations lie within a factor of 2 within the measurement values. At the remaining stations the concentrations are all overpredicted by the model. This becomes exceedingly evident for the Southern Hemisphere summer season (mean over December, January and February) where the DMS sea surface concentration and the DMS emission are high. DMS contributions higher than 50% appear in 17 stations. For 12 stations with a contribution higher than 50%, the simulated values are a factor of 2 higher than the measured values. During the winter months (mean over June, July and August) DMS contributions higher than 50% exist for only 5 locations, 2 of which show higher simulated than measured values. In summary, for remote measurement stations the simulated  $\text{SO}_4^{2-}$  surface concentrations are in agreement with the reported measurement values. Discrepancies from the observed values are highest for locations with a high DMS contribution, in particular in the summer season of the Southern Hemisphere. Here the model overpredicts the averaged observed concentrations by a factor of 2.3. This overestimation may be caused by too high DMS emissions in these regions which are either due to too high DMS sea surface concentrations or too high sea-air exchange rates. The simulated DMS sea surface concentrations in the Southern High Latitudes are very high compared to other regions of the ocean. Measurements reported from these regions confirm these high concentrations in the sea surface layers. The annually averaged values for provinces in the Southern Ocean show a slight overestimation in our simulation. The differences lie thereby in the range of the standard deviation (Table 5) and cannot solely explain the too high simulated atmospheric  $\text{SO}_4^{2-}$  surface concentrations. The sea-air exchange rate parameterization used (Wanninkhof, 1992) leads to the highest flux compared to other existing sea-air exchange rate parameterizations (e.g. Nightingale et al., 2000; Liss and Merlivat, 1986). Differences in the DMS flux can be up to a factor of 2 (e.g. Kettle and Andreae, 2000; Boucher et al., 2003). Despite too high simulated DMS emissions, one possible reason for the overestimated  $\text{SO}_4^{2-}$  surface concentration might be

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a missing reaction mechanism of DMS in the atmosphere preventing the formation of  $\text{SO}_4^{2-}$ . Several studies highlight the importance of the reaction of DMS with bromine oxide (BrO) radicals leading to the formation of DMSO (e.g. [von Glasow and Crutzen, 2004](#); [Boucher et al., 2003](#)). This reaction mechanism is not included in our study.

DMSO reacts with OH in the atmosphere, whereas the reaction products are uncertain. It has been postulated that dimethyl sulphone ( $\text{DMSO}_2$ ) is the main oxidation product leading to the formation of methyl sulfinic acid (MSIA) and methyl sulfonic acid (MSA) ([Yin et al., 1990](#)). A 1-d model study for marine boundary layer conditions ([von Glasow and Crutzen, 2004](#)) shows that the inclusion of halogen chemistry increases the DMS destruction by about 25% in summer and 100% in winter time. The  $\text{SO}_2$  yield from the oxidation of DMS is simulated lower when halogen chemistry is included. However, the lack of BrO measurements in the atmosphere makes a global assessment of the importance of the BrO oxidation difficult.

#### 4. Summary and conclusions

The production of marine Dimethylsulfide (DMS) and its fate in the atmosphere are simulated in a global coupled atmosphere-ocean circulation model. The processes for marine DMS production and decay are included in the representation of plankton dynamics in the marine biogeochemistry model HAMOCC5 embedded in a global ocean general circulation model (MPI-OM). The atmospheric model ECHAM5 is extended by the microphysical aerosol model HAM.

The simulated DMS sea surface concentrations generally match the observed concentrations. The seasonal variation with its high DMS sea surface concentration in the high latitudes in the summer hemispheres is captured by the model. The global annual mean DMS sea surface concentration of 1.8 nmol/l lies within the range of DMS sea surface climatologies (e.g. [Belviso et al., 2004a](#)).

We apply our simulated mixed layer depth (MLD) and chlorophyll  $\alpha$  concentration to the [Simo and Dachs \(2002\)](#) algorithm, to calculate DMS sea surface concentrations

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solely from this two quantities. The resulting zonal mean DMS sea surface distribution is comparable to the one derived by [Simo and Dachs \(2002\)](#) using climatological MLD fields and chlorophyll  $\alpha$  concentrations from SeaWiFS. However, compared to the DMS sea surface concentration simulated with the biogeochemical model the [Simo and Dachs \(2002\)](#) algorithm results in distinct different DMS sea surface distributions. This is most pronounced in the Northern Hemisphere, with simulated values lower than the ones predicted from the [Simo and Dachs \(2002\)](#) algorithm.

The treatment of DMS in a coupled atmosphere-ocean model including a microphysical aerosol scheme and an atmospheric sulfur model allows to gain additional insight into the DMS representation in the model by a comparison with atmospheric DMS related measurements. The simulated DMS flux into the atmosphere is 28 Tg(S)/yr which is in the range of current estimates (e.g. [Kettle and Andreae, 2000](#)). The resulting column integrated burden of DMS in the atmosphere is 0.08 Tg(S) and the lifetime is 1.0 days. DMS contributes 30% to the total sulfur source considered in the model (sulfur emissions from fossil- and bio- fuel use, wildfires and volcanoes and sulfur from DMS oxidation). The contribution of  $\text{SO}_2$  derived from oxidation of DMS by OH and  $\text{NO}_3$  to the total  $\text{SO}_2$  column burden is 25%.  $\text{SO}_2$  is oxidized by OH in the gas phase and by  $\text{H}_2\text{O}_2$  and  $\text{O}_3$  in the aqueous phase to form  $\text{SO}_4^{2-}$ . 27% of the produced  $\text{SO}_4^{2-}$  can be attributed to DMS oxidation. The contribution is highest in the biologically active season in remote regions of the Southern Ocean with values up to 90%.

The comparison of  $\text{SO}_4^{2-}$  measurements and simulated  $\text{SO}_4^{2-}$  concentrations at remote sites where the contribution of DMS to  $\text{SO}_4^{2-}$  is generally high shows an overestimation of the  $\text{SO}_4^{2-}$  surface concentrations by the model, most pronounced in the biologically active season. Possible explanations are an overestimation of the DMS sea surface concentration, a too high sea-air exchange rate or a missing reaction mechanism of DMS in the atmosphere. The simulated DMS sea surface concentrations are generally in agreement with the observations. A direct validation of the DMS flux is not possible because it cannot yet be measured directly. A missing reaction of DMS in the atmosphere model is the reaction with BrO. It has been shown by several investigators

that this reaction is important in remote regions (e.g. von Glasow and Crutzen, 2004; Boucher et al., 2003). However, the concentration of BrO in the atmosphere is not well known which makes a global assessment not feasible.

Future work will be to include the next step of the proposed DMS-climate feedback, i.e. the connection between aerosols and the cloud microphysics. The prognostic treatment of aerosol size distribution, composition and mixing state in the ECHAM5-HAM model provides the basis for such a microphysical coupling of the aerosol and the cloud scheme with an explicit simulation of cloud droplet and ice crystal number concentrations. For an assessment of the DMS-cloud link, it is important to consider sea salt and its role as CCN. Sea-salt emissions are high in regions with high DMS emissions and are also influenced by climate change.

The generally good agreement between model and measurements indicates that the DMS cycle in the model represents the processes governing DMS sea surface concentrations, DMS emissions and resulting atmospheric concentrations reasonably well. However, the lack of measurements of the consumption and production processes of DMS in the ocean hampers the full evaluation of the DMS formulation as a predictive tool for DMS. Nevertheless, the DMS formulation applied in a coupled ocean-atmosphere model is a step forwards in providing a model system to assess marine biosphere-climate feedbacks.

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**Table 1.** Parameters for DMS formulation in HAMOCC5. The parameters are derived from an optimization procedure of HAMOCC5 using DMS sea surface concentration measurements from the [Kettle and Andreae \(2000\)](#) database.

Symbol	constant		Process
DMS Production			
$k_{psi}$	0.0136	$(S(DMS)/(Si))$	silicate
$k_{pcc}$	0.1345	$(S(DMS)/(C))$	calcium carbonate
$k_{pt}$	10.01	(°C)	temperature dependence
DMS Decay			
$k_{Iuv}$	0.0011	(m <sup>2</sup> /Wd)	photolysis
$k_{Ib}$	0.1728	(d <sup>-1</sup> °C <sup>-1</sup> )	bacteria

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**Table 2.** Aerosol and aerosol precursor emissions used in the HAM model. Global annual mean in Tg/yr and Tg(S)/yr for sulfuric species.

Species	Source	Reference	Tg/yr
DMS	Terrestrial Biosphere	<a href="#">Pham et al. (1995)</a>	0.3
	Marine Biosphere	HAMOCC5	27.6
SO <sub>2</sub>	Volcanoes	<a href="#">Andres and Kasgnoc (1998)</a> <a href="#">Halmer et al. (2002)</a>	14.6
	Vegetation Fires	<a href="#">van der Werf et al. (2003)</a>	2.1
	Industry, Fossil-Fuel, Bio-Fuels	<a href="#">Cofala et al. (2005)</a>	54.2
Total sulfur			99.0
BC	Vegetation Fires	<a href="#">van der Werf et al. (2003)</a>	3.0
	Fossil-Fuel and Bio-Fuels	<a href="#">Bond et al. (2004)</a>	4.7
Total BC			7.7
POM	Vegetation Fires	<a href="#">van der Werf et al. (2003)</a>	34.7
	Biogenic	<a href="#">Guenther et al. (1995)</a>	19.1
	Fossil-Fuel and Bio-Fuels	<a href="#">Bond et al. (2004)</a>	12.5
Total POM			66.3
SSA	Wind driven	<a href="#">Schulz et al. (2004)</a>	5868.6
DU	Wind driven	<a href="#">Tegen et al. (2002)</a>	1060.6

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**Table 3.** Sulfur budget: global, Northern Hemisphere and Southern Hemisphere. The lifetime is calculated as the ratio of the column burden to the sum of all sources.

	SH	NH	Global
<b>DMS</b>			
Source ( Tg(S)/yr):			
total:	17.8	9.8	27.6
Sinks (Tg(S)/yr):			
oxidation with OH	15.8	7.5	23.3
oxidation with NO <sub>3</sub>	1.8	2.4	4.2
burden (Tg(S)):	0.056	0.021	0.077
lifetime (d) :	1.15	0.78	1.02
<b>SO<sub>2</sub></b>			
Source (Tg(S)/yr):			
total (Emission + DMS oxidation)	25.9	69.3	94.2
Sinks (Tg(S)/yr):			
oxidation	21.6	51.7	73.3
dry deposition	2.9	14.3	17.3
wet deposition	1.4	2.2	3.6
burden (Tg(S)):	0.19	0.41	0.60
lifetime (d) :	2.7	2.2	2.4
<b>SO<sub>4</sub><sup>2-</sup><sub>gas</sub></b>			
Source (Tg(S)/yr):			
total ( SO <sub>2</sub> gas oxidation )	8.7	18.3	27.0
Sinks (Tg(S)/yr):			
condensation	8.7	18.2	26.8
nucleation	0.05	0.06	0.11
wet deposition	0.02	0.02	0.04
dry deposition	0.002	0.004	0.006
burden (Tg(S)):	0.0003	0.0004	0.0007
lifetime (d) :	0.014	0.008	0.010
<b>SO<sub>4</sub><sup>2-</sup></b>			
Source (Tg(S)/yr):			
total (Emission + SO <sub>2</sub> in cloud oxidation + condensation + nucleation)	24.4	53.9	78.4
Sinks (Tg(S)/yr):			
dry deposition	0.6	1.8	2.5
wet deposition	23.3	50.8	74.1
sedimentation	0.56	1.29	1.85
burden (Tg(S)):	0.24	0.49	0.73
lifetime (d) :	3.64	3.32	3.42

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**Table 4.** Annual mean column burdens of DMS, SO<sub>2</sub>, SO<sub>4</sub><sup>2-</sup> resulting from all sulfur sources (total) in (Tg(S)) and resulting only from DMS in (%).

	annual mean			December/January/ February			June/July/August		
	global	NH	SH	global	NH	SH	global	NH	SH
DMS (Tg(S))	0.077	0.021	0.056	0.151	0.013	0.137	0.048	0.032	0.016
SO <sub>2</sub> total (Tg(S))	0.604	0.414	0.190	0.642	0.440	0.202	0.592	0.400	0.192
% of SO <sub>2</sub> from DMS	24.7	15.7	44.2	28.8	15.0	58.9	21.0	15.8	31.25
SO <sub>4</sub> <sup>2-</sup> total (Tg(S))	0.733	0.493	0.240	0.674	0.363	0.311	0.813	0.617	0.195
% of SO <sub>4</sub> <sup>2-</sup> from DMS	26.7	17.8	45.0	37.2	20.7	56.6	21.3	17.5	33.3

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**Table 5.** List of biogeochemical provinces defined by Longhurst et al. (1995) and corresponding DMS sea surface concentrations for data points included in the Kettle and Andreae (2000) database as used in Fig. 2a. Longitude and latitude values represent the center of the province.

Province	longitude (°)	latitude (°)	measurements (nmol/l)	sdev (nmol/l)	model (nmol/l)	sdev (nmol/l)	number of measurements	area (m <sup>2</sup> )
<b>Atlantic Ocean</b>								
Boreal Polar Province	-160	80	2.26	1.64	0.29	2.31	12	1.27E+13
Atlantic Arctic Province	-10	75	2.92	4.98	4.26	6.71	37	3.54E+12
Atlantic Subarctic Province	-15	60	9.45	9.83	2.21	1.84	28	2.23E+12
N. Atlantic Drift Province	-30	50	3.38	2.66	2.27	2.28	67	3.44E+12
Gulf Stream Province	-60	40	3.94	3.46	1.33	1.72	11	1.06E+12
N. Atlantic Subtropical Gyral Province (West)	-65	30	1.46	0.85	0.76	0.53	26	6.22E+12
N. Atlantic Tropical Gyral Province	-50	20	2.21	1.17	2.18	1.05	30	7.84E+12
Western Tropical Atlantic Province	-45	5	1.56	1.34	3.21	1.45	39	5.64E+12
Eastern Tropical Atlantic Province	0	-5	1.82	1.55	3.42	1.49	34	5.37E+12
South Atlantic Gyral Province	-20	-20	1.76	1.37	2.31	11.07	23	1.74E+13
NE Atlantic Shelves Province	5	55	6.26	3.78	3.89	2.97	10	1.44E+12
Canary Coastal Province	-20	15	5.93	4.63	4.54	2.34	13	7.39E+11
Guinea Current Coastal Province	5	5	3.06	1.35	2.67	1.10	8	1.33E+12
Guianas Coastal Province	-65	3	1.59	0.00	2.45	0.00	1	1.33E+12
NW Atlantic Shelves Province	-70	45	4.33	2.84	3.30	5.70	15	1.95E+12
Mediterranean Sea, Black Sea Province	10	40	5.02	4.29	1.57	0.59	16	2.96E+12
Caribbean Province	-80	15	1.25	0.00	3.31	0.00	1	4.31E+12
N. Atlantic Subtropical Gyral Province (East)	-25	40	5.17	6.51	2.23	1.76	79	4.67E+12
Cheasapeake Bay Province	-75	38	-	-	-	-	0	9.50E+11
Brazil Current Coastal Province	-55	-25	3.06	3.33	2.21	0.57	14	1.02E+12
SW Atlantic Shelves Province	-73	-50	5.77	3.38	5.89	6.27	6	1.49E+12
Benguela Current Coastal Province	8	-25	4.14	2.30	2.42	1.04	15	1.20E+12
<b>Indian Ocean</b>								
Indian Monsoon Gyres Province	65	-5	1.59	0.92	2.19	0.99	12	1.44E+13
Indian S. Subtropical Gyre Province	65	-20	3.66	3.71	1.67	0.93	13	1.88E+13
E. Africa Coastal Province	30	-20	7.83	4.00	1.39	0.21	2	3.51E+12
Red Sea, Persian Gulf Province	35	35	1.20	0.22	2.50	0.70	3	5.63E+11
NW Arabian Upwelling Province	50	50	2.76	0.87	2.99	1.40	14	2.90E+12
E. India Coastal Province	90	90	-	-	-	-	0	7.87E+11
W. India Coastal Province	70	70	-	-	-	-	0	8.32E+11
Australia-Indonesia Coastal Province	110	110	-	-	-	-	0	2.79E+12

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**Table 5.** Continued.

Province	longitude (°)	latitude (°)	measurements (nmol/l)	sdev (nmol/l)	model (nmol/l)	sdev (nmol/l)	number of measurements	area (m <sup>2</sup> )
<b>Southern Ocean</b>								
S. Subtropical Convergence Province	70	-40	3.16	2.43	4.25	3.51	42	1.96E+13
Subantarctic Province	70	-50	3.43	3.95	3.37	3.81	74	2.82E+13
Antarctic Province	70	-60	5.42	7.04	7.47	7.08	79	2.23E+13
Austral Polar Province	70	-68	12.31	27.01	15.25	14.38	61	9.10E+12
<b>Pacific Ocean</b>								
N. Pacific Epicontinental Province	-175	60	3.51	2.86	2.72	1.10	4	3.88E+12
Pacific Subarctic Gyres Province (East)	-170	50	6.48	4.31	1.30	1.53	32	3.00E+12
Pacific Subarctic Gyres Province (West)	150	50	4.27	1.78	2.12	0.79	10	2.71E+12
Kuroshio Current Province	145	37	1.41	0.47	0.93	0.19	10	4.07E+12
N. Pacific Polar Front Province	-170	40	2.20	1.67	1.64	1.37	10	3.63E+12
N. Pacific Subtropical Gyre Province (East)	-170	30	4.12	4.10	1.67	1.03	30	6.69E+12
N. Pacific Subtropical Gyre Province (West)	150	30	2.60	1.31	0.89	0.25	6	3.68E+12
Offshore California Current Province	-135	45	2.33	1.19	2.43	1.44	15	2.29E+12
Tasman Sea Province	150	-40	2.32	1.33	5.71	2.46	18	1.66E+12
S. Pacific Subtropical Gyre Province	-140	-20	2.66	1.49	1.71	0.78	93	3.71E+13
N. Pacific Tropical Gyre Province	-170	20	1.40	0.61	1.54	0.44	78	2.22E+13
N. Pacific Equatorial Countercurrent Province	-120	8	2.45	1.47	2.16	0.99	26	7.98E+12
Pacific Equatorial Divergence Province	-120	-5	3.06	1.00	2.61	1.06	61	9.85E+12
W. Pacific Warm Pool Province	150	5	2.44	1.17	2.40	0.81	75	1.60E+13
Archipelagic Deep Basins Province	160	-20	2.48	1.04	1.37	0.27	15	8.63E+12
Alaska Downwelling Coastal Province	-140	60	2.35	3.50	0.84	1.61	4	5.88E+11
California Upwelling Coastal Province	-125	35	2.49	1.62	3.85	1.00	17	9.01E+11
Central American Coastal Province	-110	25	2.38	0.25	2.44	0.49	3	1.46E+12
Chile-Peru Current Coastal Province	-78	-30	5.75	2.20	3.69	1.98	7	2.73E+12
China Sea Coastal Province	120	35	-	-	-	-	0	9.59E+11
Sunda-Arafura Shelves Province	130	-10	1.28	0.92	1.97	0.84	4	6.51E+12
East Australian Coastal Province	140	-25	-	-	-	-	0	1.15E+12
New Zealand Coastal Province	155	-50	-	-	-	-	0	1.00E+12

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**Table 6.** List of biogeochemical provinces defined by Longhurst et al. (1995) and corresponding DMS sea surface concentrations for data points not included in the Kettle and Andreae (2000) database as used in Fig. 2b. Longitude and latitude values represent the center of the province.

Province	longitude (°)	latitude (°)	measurements (nmol/l)	sdev [nmol/l]	model (nmol/l)	sdev (nmol/l)	number of measurements	area (m <sup>2</sup> )
<b>Atlantic Ocean</b>								
Boreal Polar Province	-160	80	2.31	1.03	0.02	0.01	31	1.27E+13
Atlantic Arctic Province	-10	75	1.38	0.43	0.02	0.02	3	3.54E+12
Atlantic Subarctic Province	-15	60	1.47	0.18	2.20	0.62	3	2.23E+12
N. Atlantic Drift Province	-30	50	1.54	0.49	3.62	2.90	8	3.44E+12
Gulf Stream Province	-60	40	0.82	0.00	0.05	0.00	1	1.06E+12
N. Atlantic Subtropical Gyral Province (West)	-65	30	1.11	0.56	0.35	0.20	9	6.22E+12
N. Atlantic Tropical Gyral Province	-50	20	1.37	0.23	0.77	0.24	12	7.84E+12
Western Tropical Atlantic Province	-45	5	1.66	0.38	2.50	0.99	10	5.64E+12
Eastern Tropical Atlantic Province	0	-5	1.65	0.25	4.14	0.76	9	5.37E+12
South Atlantic Gyral Province	-20	-20	3.89	1.64	2.92	0.52	17	1.74E+13
NE Atlantic Shelves Province	5	55	1.55	0.24	1.37	0.96	5	1.44E+12
Canary Coastal Province	-20	15	1.56	0.23	4.91	2.59	7	7.39E+11
Guinea Current Coastal Province	5	5	-	-	-	-	0	1.33E+12
Guianas Coastal Province	-65	3	-	-	-	-	0	1.33E+12
NW Atlantic Shelves Province	-70	45	1.34	0.00	0.27	0.00	1	1.95E+12
Mediterranean Sea, Black Sea Province	10	40	-	-	-	-	0	2.96E+12
Caribbean Province	-80	15	-	-	-	-	0	4.31E+12
N. Atlantic Subtropical Gyral Province (East)	-25	40	1.45	0.25	3.02	1.60	15	4.67E+12
Cheasapeake Bay Province	-75	38	-	-	-	-	0	9.50E+11
Brazil Current Coastal Province	-55	-25	-	-	-	-	0	1.02E+12
SW Atlantic Shelves Province	-73	-50	-	-	-	-	0	1.49E+12
Benguela Current Coastal Province	8	-25	3.96	0.77	2.18	1.27	8	1.20E+12
<b>Indian Ocean</b>								
Indian Monsoon Gyres Province	65	-5	2.92	0.99	2.93	0.99	31	1.44E+13
Indian S. Subtropical Gyre Province	65	-20	4.70	2.21	1.83	2.21	13	1.88E+13
E. Africa Coastal Province	30	-20	2.38	0.84	2.01	0.84	15	3.51E+12
Red Sea, Persian Gulf Province	35	35	-	-	-	-	0	5.63E+11
NW Arabian Upwelling Province	50	50	-	-	-	-	0	2.90E+12
E. India Coastal Province	90	90	-	-	-	-	0	7.87E+11
W. India Coastal Province	70	70	-	-	-	-	0	8.32E+11
Australia-Indonesia Coastal Province	110	110	-	-	-	-	0	2.79E+12

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**Table 6.** Continued.

Province	longitude (°)	latitude (°)	measurements (nmol/l)	sdev [nmol/l]	model (nmol/l)	sdev (nmol/l)	number of measurements	area (m <sup>2</sup> )
<b>Pacific Ocean</b>								
N. Pacific Epicontinental Province	-175	60	2.62	3.29	1.64	2.08	19	3.88E+12
Pacific Subarctic Gyres Province (East)	-170	50	2.26	2.11	2.23	1.97	13	3.00E+12
Pacific Subarctic Gyres Province (West)	150	50	4.23	6.31	2.43	3.14	5	2.71E+12
Kuroshio Current Province	145	37	2.37	0.72	1.69	0.65	12	4.07E+12
N. Pacific Polar Front Province	-170	40	-	-	-	-	0	3.63E+12
N. Pacific Subtropical Gyre Province (East)	-170	30	4.05	3.40	2.16	1.03	72	6.69E+12
N. Pacific Subtropical Gyre Province (West)	150	30	1.17	0.32	0.09	0.10	15	3.68E+12
Offshore California Current Province	-135	45	3.07	1.36	1.70	0.19	4	2.29E+12
Tasman Sea Province	150	-40	-	-	-	-	0	1.66E+12
S. Pacific Subtropical Gyre Province	-140	-20	2.09	0.26	1.45	0.14	3	3.71E+13
N. Pacific Tropical Gyre Province	-170	20	0.97	0.04	1.52	0.21	3	2.22E+13
N. Pacific Equatorial Countercurrent Province	-120	8	2.11	0.98	2.13	0.54	18	7.98E+12
Pacific Equatorial Divergence Province	-120	-5	2.65	0.55	2.36	0.84	20	9.85E+12
W. Pacific Warm Pool Province	150	5	-	-	-	-	0	1.60E+13
Archipelagic Deep Basins Province	160	-20	-	-	-	-	0	8.63E+12
Alaska Downwelling Coastal Province	-140	60	5.52	4.87	2.66	3.26	13	5.88E+11
California Upwelling Coastal Province	-125	35	1.42	0.74	0.88	0.48	10	9.01E+11
Central American Coastal Province	-110	25	4.44	0.84	2.23	0.13	2	1.46E+12
Chile-Peru Current Coastal Province	-78	-30	-	-	-	-	0	2.73E+12
China Sea Coastal Province	120	35	-	-	-	-	0	9.59E+11
Sunda-Arafura Shelves Province	130	-10	-	-	-	-	0	6.51E+12
East Australian Coastal Province	140	-25	-	-	-	-	0	1.15E+12
New Zealand Coastal Province	155	-50	-	-	-	-	0	1.00E+12

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**Table 7.** List of measurements sites from the University of Miami Network used in Fig. 11.

Location	Longitude	Latitude	Model annual mean ( $\mu\text{g m}^{-3}$ )	Measurements annual mean ( $\mu\text{g m}^{-3}$ )	DMS contribution annual mean (%)
Chatham Island – New Zealand	–176.5	–43.9	0.52	0.27	59
Cape Point – South Africa	18.5	–34.3	1.08	0.60	37
Cape Grim – Tasmania	144.7	–40.7	0.89	0.30	51
Linvergill – New Zealand	168.4	–46.4	0.49	0.44	60
Marsh – King George Island	–58.3	–62.2	0.38	0.27	52
Marion Island	37.8	–46.9	0.44	0.08	61
Mawson – Antarctica	62.5	–67.6	0.08	0.11	73
Palmer Station – Antarctica	–64.1	–64.8	0.24	0.09	63
Reunion Island	55.8	–21.2	0.40	0.35	54
Wellington – New Zealand	174.9	–41.3	0.75	0.43	52
Yate – New Caledonia	167.0	–22.1	0.59	0.43	45
Funafuti – Tuvalu	–179.2	–8.5	0.58	0.17	60
Nauru	166.9	–0.5	1.22	0.15	58
Norfolk Island	168.0	–29.1	0.51	0.27	55
Rarotonga – Cook Islands	–159.8	–21.2	0.19	0.11	65
American Samoa	–170.6	–14.2	0.25	0.34	64
Midway Island	–177.4	28.2	0.46	0.52	33
Oahu Hawaii	–157.7	21.3	0.73	0.51	33
Cheju – Korea	126.5	33.5	6.14	7.21	6
Hedo Okinawa – Japan	128.2	26.9	3.55	4.28	15
Fanning Island	–159.3	3.9	1.17	0.64	48
Enewetak Atoll	162.3	11.3	0.37	0.08	55
Barbados	–59.4	13.2	0.75	0.67	38
Izana Tenerife	–16.5	29.3	2.40	0.96	22
Bermuda	–64.9	32.3	1.03	2.09	22
Heimaey Iceland	–20.3	63.4	0.47	0.69	26
Mace Head – Ireland	–9.9	53.3	1.70	1.27	22
Miami	–80.2	25.8	1.82	2.17	18

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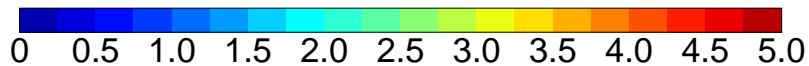
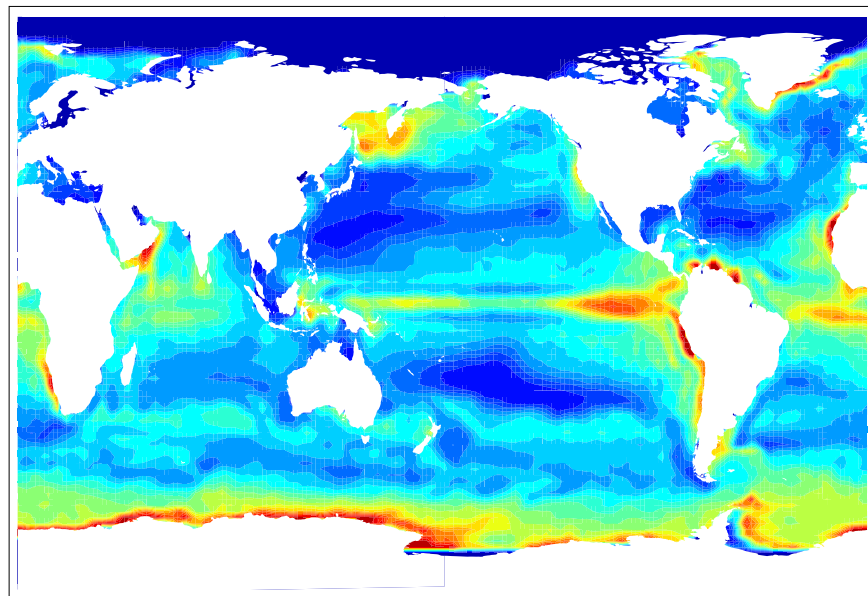
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**Fig. 1.** Modeled annual mean DMS sea surface concentration. Units are nmol/l.

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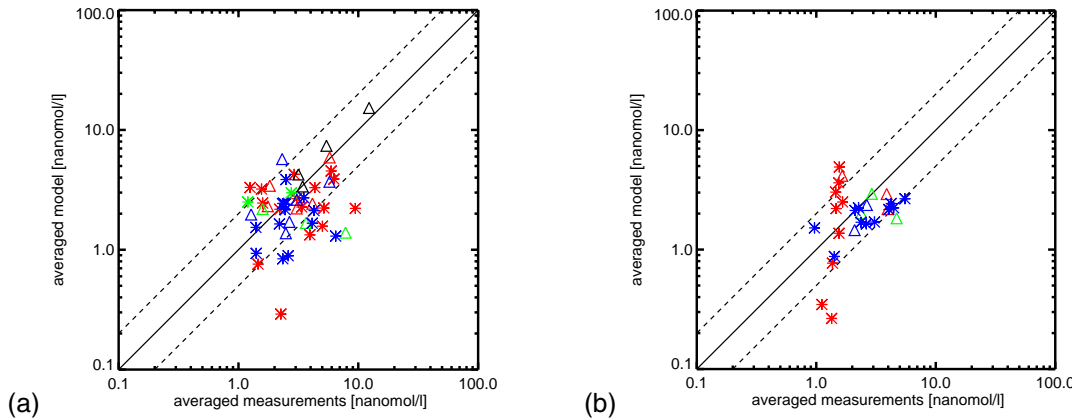
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**Fig. 2.** Scatter plot of simulated and measured annual mean DMS sea surface concentrations averaged over the biogeochemical provinces defined by Longhurst et al. (1995). Provinces for the Atlantic Ocean in red, the Indian Ocean in green, the Pacific Ocean in blue, and the Southern Ocean in black. Provinces in the Southern Hemisphere are indicated by a triangle, in the Northern Hemisphere by an asterisk. Exact values are listed in Tables 5 and 6. **(a)** Measurements from the Kettle and Andreae (2000) database. **(b)** Measurements obtained from <http://saga.pmel.noaa.gov/dms/>, excluding the measurements of the Kettle and Andreae (2000) database. The solid line indicates the 1:1 ratio, the dashed lines the 1:2 and 2:1 ratios. Units are nmol/l.

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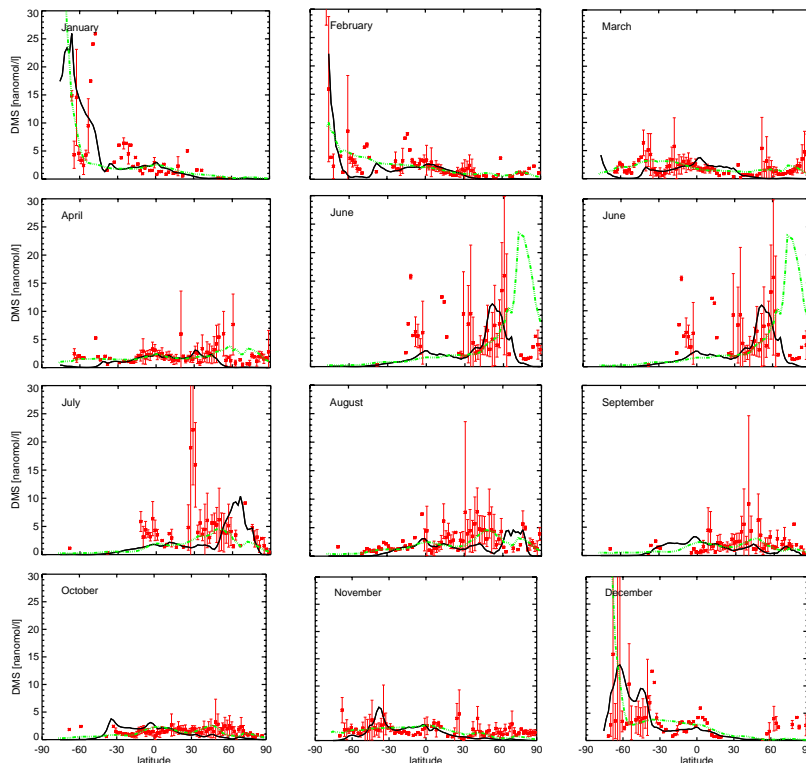
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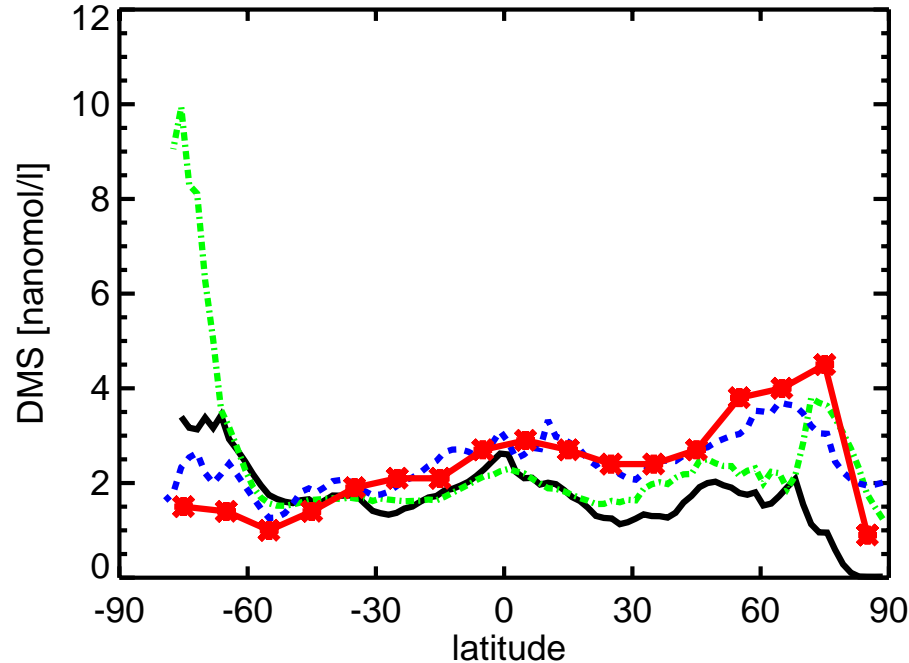


**Fig. 3.** Zonally averaged profiles of DMS sea surface concentrations for all months. The black line represents the zonal average of the modeled DMS sea surface concentration, the green line the zonal average of the [Kettle and Andreae \(2000\)](#) DMS sea surface climatology. The red symbols represent the zonally averaged ocean data points given in the [Kettle and Andreae \(2000\)](#) database (data points where the ocean depth is below 300 m are excluded) gridded onto the model grid. Where more than one ocean grid box is present, the standard deviation is given by the red vertical line. Units are nmol/l.

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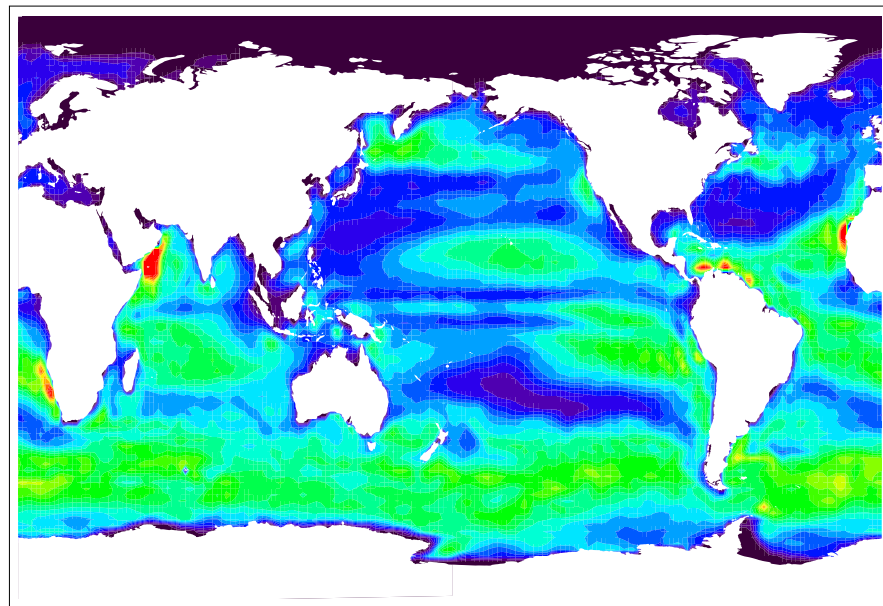


**Fig. 4.** Zonal annual means of sea surface DMS for the [Simo and Dachs \(2002\)](#) algorithm using the simulated MLD and chlorophyll  $\alpha$  concentration (dashed blue line), resulting concentrations from the simulation using the marine biogeochemistry model HAMOCC5 (black line), annual mean DMS sea surface concentration averaged over  $10^\circ$  latitudinal bands given in [Simo and Dachs \(2002\)](#) using climatological MLD and chlorophyll  $\alpha$  concentration from SeaWiFS (red stars) and for the [Kettle and Andreae \(2000\)](#) climatology (dotted green line). Units are nmol/l.

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**Fig. 5.** Annual mean DMS flux into the atmosphere. Units are  $\text{mg(S) m}^{-2} \text{ yr}^{-1}$ .

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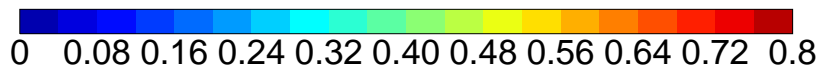
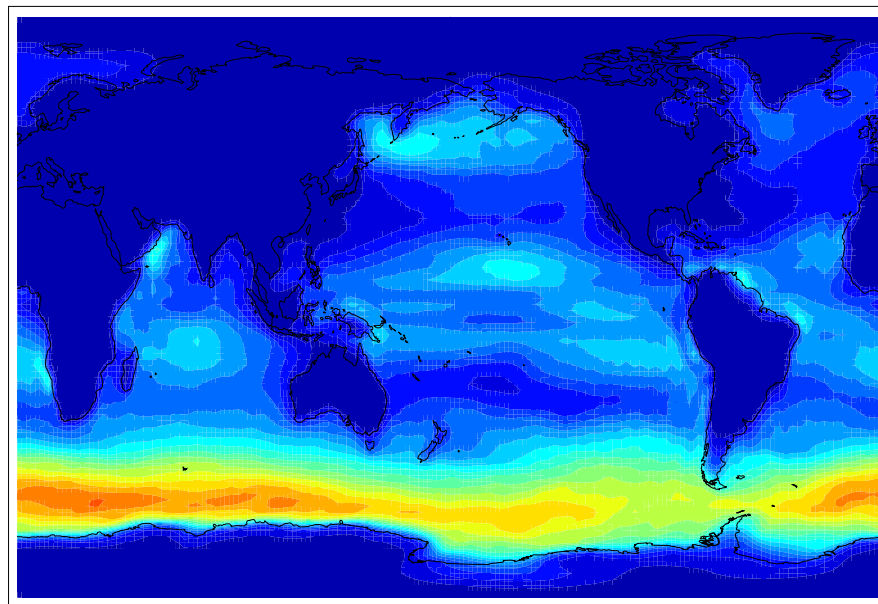
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**Fig. 6.** Annual mean DMS column burden. Units are mg(S) m<sup>-2</sup>.

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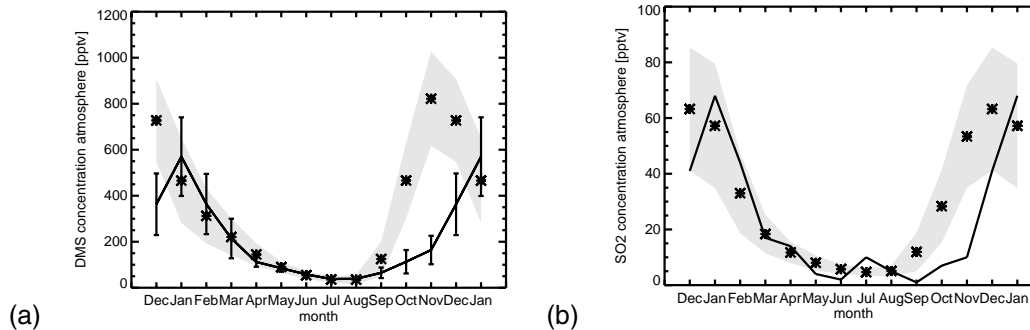
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**Fig. 7.** (a) Atmospheric DMS at Amsterdam Island, measurements and standard deviation after (Sciare et al., 2000). (b) Atmospheric SO<sub>2</sub> at Amsterdam Island, the measurements are reported by Putaud et al. (1992). The line represents the measurements, stars the model results. The grey shading indicates the monthly mean simulated standard deviation. Units are pptv.

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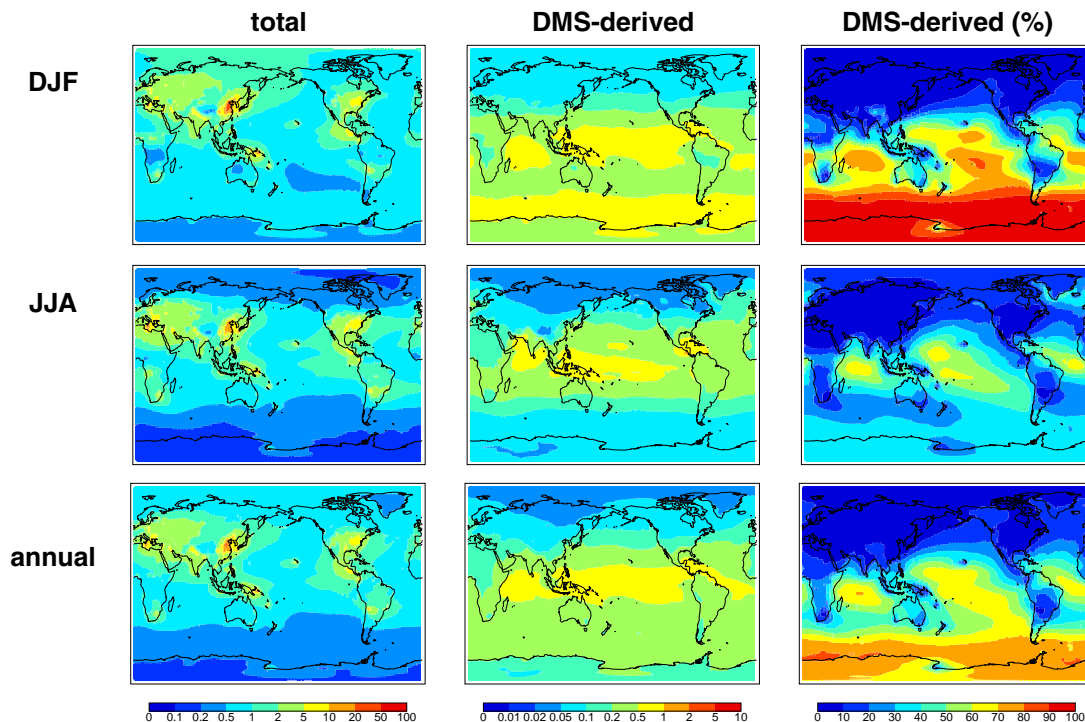
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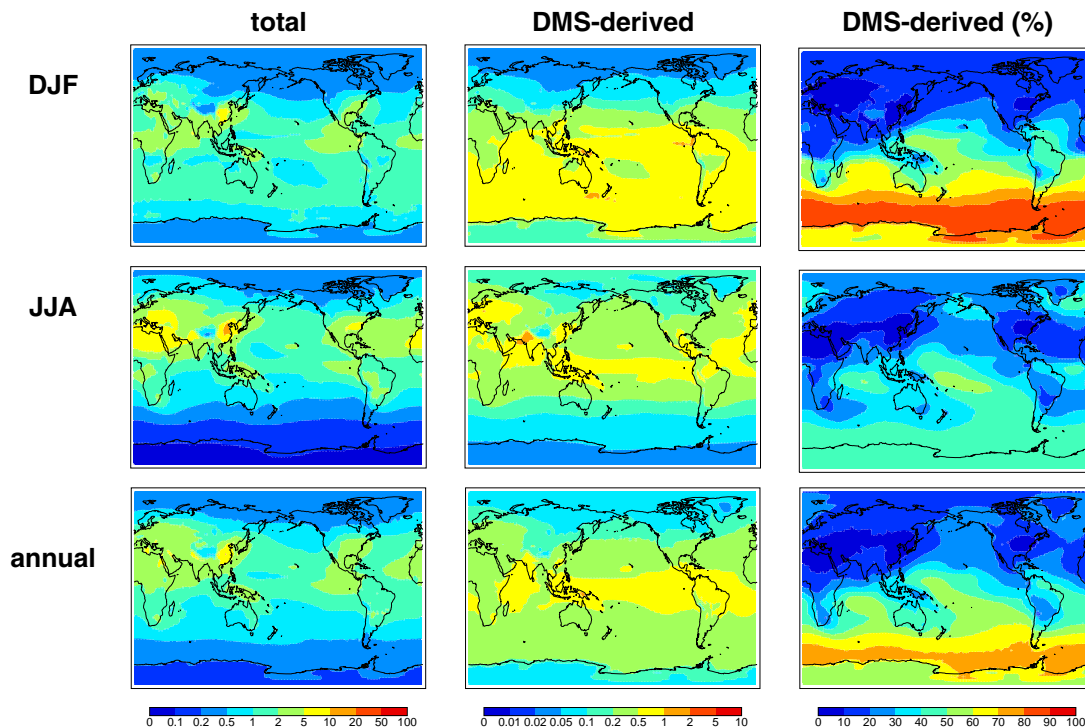
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**Fig. 8.** Mean column burdens of  $\text{SO}_2$  averaged for December, January and February (DJF) and June, July and August (JJA) and annual mean values resulting from all sources (total), resulting solely from DMS (DMS-derived) and percentage of  $\text{SO}_2$  attributable to DMS (DMS-derived (%)), respectively. Units are  $\text{mg(S) m}^{-2}$  and %, respectively.

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**Fig. 9.** Mean column burdens of  $\text{SO}_4^{2-}$  averaged for December, January and February (DJF) and June, July and August (JJA) and annual mean values resulting from all sources (total), resulting solely from DMS (DMS-derived) and percentage of  $\text{SO}_4^{2-}$  attributable to DMS (DMS-derived (%)), respectively. Units are  $\text{mg}(\text{S}) \text{m}^{-2}$  and %, respectively.

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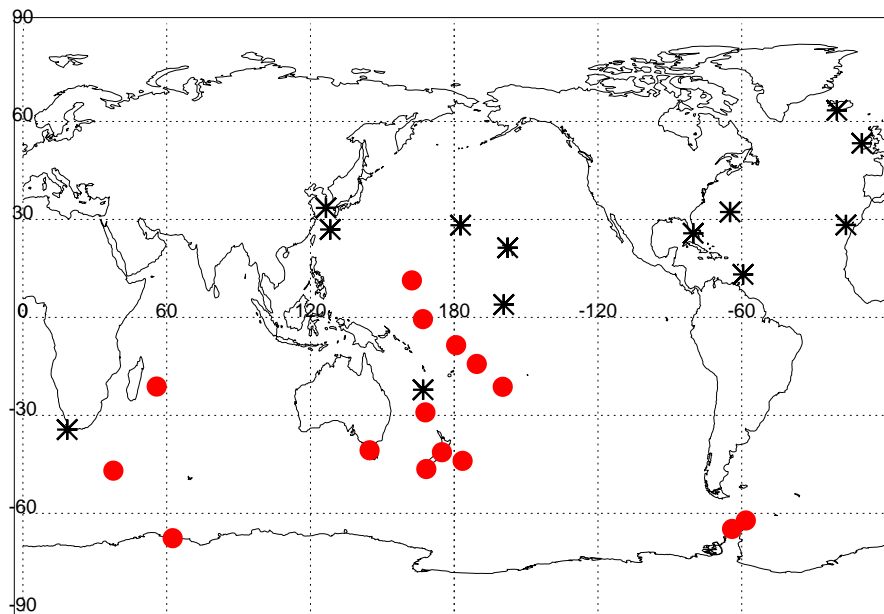
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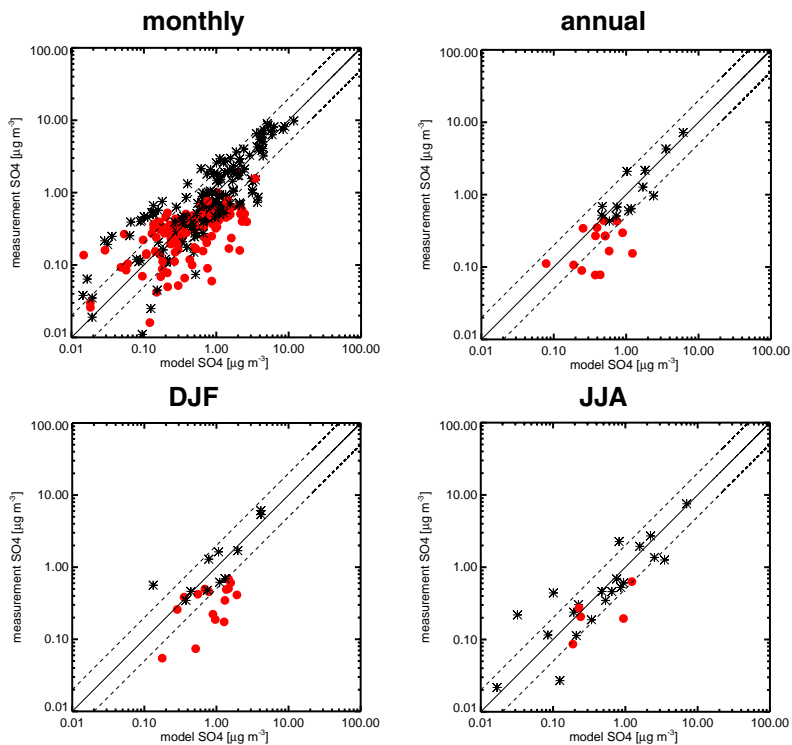


**Fig. 10.** Location of the measurement sites included in the University of Miami network. Red points indicate stations with a simulated annual mean DMS contribution to  $\text{SO}_4^{2-}$  at the surface higher than 50%.

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**Fig. 11.** Scatter plot of measured and simulated surface aerosol mass concentration of  $\text{SO}_4^{2-}$ . Measurements are from the University of Miami network. **(a)** Monthly mean, **(b)** annual mean, **(c)** mean for December, January and February, **(d)** mean for June, July and August. Red symbols indicate a contribution of DMS to  $\text{SO}_4^{2-}$  higher than 50%. The solid line indicates the 1:1 ratio, the dashed lines the 1:2 and 2:1 ratios. Units are  $\mu\text{g}(\text{S}) \text{m}^{-3}$ .

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