

Will marine dimethylsulfide emissions amplify or alleviate global warming? – A model study.

Laurent Bopp, Olivier Boucher, Olivier Aumont, Sauveur Belviso, Jean-Louis Dufresne, Mai Pham, and Patrick Monfray

Abstract:

Dimethylsulfide (DMS) is a biogenic sulfur compound produced in sea-surface water and outgassed to the atmosphere. Once in the atmosphere, it is oxidized and contributes to form sulfate aerosol particles that affect the radiative budget as cloud condensation nuclei (CCN). More than 15 years ago, Charlson et al. (1987) have postulated that climate may be partly modulated by variations in DMS production. We test this hypothesis in the context of anthropogenic climate change and present here a first modeled estimate of the radiative impact due to changes in DMS air-sea fluxes caused by global warming. Following previous work on modeling the marine DMS response to climate change (Bopp et al. 2003), we use an atmospheric model of the global sulfur cycle to simulate the responses of DMS sea-to-air flux and its associated radiative impact to increased greenhouse gas concentration (a 1% increase per year in atmospheric CO₂ until the present-day concentration has doubled). At 2xCO₂, our model estimates a small increase (3%) in the global DMS flux to the atmosphere but with large spatial heterogeneities (from -15% to 30% in the zonal mean). The radiative perturbation due to the DMS-induced change in cloud albedo is estimated to be -0.05 Wm⁻², which represents only a small negative climate feedback on global warming. However there are large regional changes, such as a perturbation of up to -1.5 Wm⁻² in summer between 40°S and 50°S, which can significantly impact the regional climate. In the Southern Ocean, the radiative impact due to changes in the DMS cycle may partly alleviate the radiative forcing due to anthropogenic CO₂ (which is estimated between +2.5 and +3 Wm⁻² at 2xCO₂ compared to 1xCO₂ conditions).

1. Introduction

The sea-to-air flux of DMS is controlled by the DMS concentration at the sea surface and by the magnitude of the DMS transfer velocity across the air-sea interface, which both depend on climate variables. Sea-to-air transfer velocity mainly varies with sea-surface temperature and wind velocity (Liss and Merlivat 1986, Wanninkhof 1992, Nightingale 2000). Sea-surface DMS concentration is also regulated by climate variables such as solar irradiance, sea-surface temperature and ocean physics, through their control on the marine biology.

It has been postulated that the Earth's climate is partly regulated by variations in DMS emissions through a DMS-CCN-cloud albedo feedback (Charlson et al. 1987). However, there are large uncertainties both on the sign and on the magnitude of this feedback (Liss et al. 1994) and it is still not clear whether this mechanism can play a role in future climate change.

A first step has been accomplished by Gabric et al. (1998) and Gabric et al. (2001) with the coupling of a general cir-

ulation model together with a DMS production model to investigate the response of DMS emissions to climate change. Based on a 10°×20° area in the Subantarctic Southern Ocean, south of Australia, Gabric et al. (2001) estimate an increase in DMS emissions of 5% by 2080 (corresponding to an equivalent CO₂ tripling relative to pre-industrial levels). With this modest increased percentage, they find a small negative radiative impact, which, they conclude, confirms the minor role for DMS-derived aerosols in climate regulation. But this study is essentially a regional one and it is difficult to extend such results to the world ocean. Penner et al. (2001) estimated a small increase in global DMS flux between the years 2000 and 2100, with global DMS fluxes of 26.0 and 27.7 TgS yr⁻¹, respectively. This estimate is for constant marine DMS concentrations and thus includes only the effects of changing wind speed and sea surface temperature which were estimated from the Climate System Model of NCAR. Moreover, Penner et al. (2001) did not estimate the impact of such changes on the radiation budget.

In a previous paper (Bopp et al. 2003), a model of the global distribution of sea-surface DMS concentrations (Aumont et al. 2002) was coupled to an atmosphere-ocean general circulation model (Barthelet et al. 1998) to predict the evolution of DMS sea-water concentrations and air-sea fluxes in the context of global warming. Here, we propose to extend this previous study. To predict the impact of changes in marine DMS emissions on the atmospheric sulfur cycle and the radiative budget of the Earth, we use a global model of the atmospheric sulfur cycle. The aim of this study is to give a first quantitative assessment of the role of DMS emissions in future climate change.

As a measure of reliability, we perform additional simulations to test the robustness of our results. In particular, we use 2 different parameterizations for the air-sea gas exchange transfer function (Liss et Merlivat 1986, Nightingale 2000) and 2

Laurent Bopp,¹ **Sauveur Belviso**, and **Patrick Monfray**. Laboratoire des Sciences du Climat et de l'Environnement, CEA / CNRS, Gif sur Yvette, France.

Olivier Boucher. Laboratoire d'Optique Atmosphérique, CNRS / USTL, Villeneuve d'Ascq, France.

Olivier Aumont. Laboratoire d'Océanographie Dynamique et de Climatologie, CNRS / UPMC / IRD, Paris, France.

Jean-Louis Dufresne. Laboratoire de Météorologie Dynamique, CNRS / UPMC, Paris, France.

Mai Pham. Service d'Aéronomie, CNRS / UPMC / UVSQ, Paris, France.

¹Corresponding author: L. Bopp. e-mail: bopp@lscce.saclay.cea.fr. Phone: +33 1 69 08 32 74. Fax: +33 1 69 08 77 16.

different parameterizations for the relationship between the atmospheric sulfate mass and the cloud droplet number (CDN) concentration (Boucher and Lohmann, 1995).

2. Method

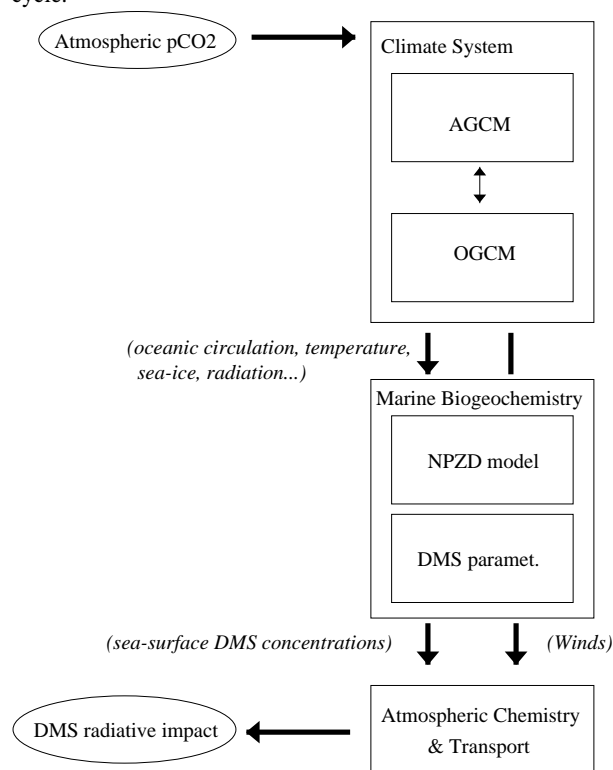
2.1. Marine DMS emissions

Sea-water DMS concentrations for $1xCO_2$ and $2xCO_2$ conditions are obtained from the work of Bopp et al. (2003). (Note that $1xCO_2$ corresponds here to 1990 (350 ppmv) rather than pre-industrial conditions.) In this previous work, the transient climate response to increased atmospheric CO_2 was obtained from simulations with the Institut Pierre-Simon Laplace Coupled Model 1 (IPSL-CM1) ocean-atmosphere general circulation model (OAGCM) (Barthelet et al. 1998). The transient climate run consists of a global warming scenario in which atmospheric pCO_2 was increased from 350 ppmv ($1xCO_2$) at a rate of 1% per year, reaching 700 ppmv ($2xCO_2$) after about 70 years. This transient climate was then used to force a marine biogeochemical scheme and to predict the evolution of marine biology (Bopp et al. 2001). The biogeochemical scheme used for this study is a Nutrient Phytoplankton Zooplankton and Detritus (NPZD) type model. Phytoplankton growth depends on the local conditions of light, temperature and turbulence, and considers PO_4^{3-} as the only limiting nutrient. Parameterizations of DMS, as non-linear functions of the phytoplankton biomass and the food web structure of the ecosystem, were applied to predict DMS sea-water concentrations (Bopp et al. 2003). Those relationships (DMS as a function of phytoplankton biomass and the food web structure of the ecosystem) have been established from datasets obtained during several cruises carried out in contrasted areas of the world oceans. The marine biogeochemical scheme and the DMS parameterizations that were used are fully described and evaluated in Aumont et al. (2002).

2.2. Sulfur cycle model

A model of the global atmospheric sulfur cycle is used to propagate the DMS fluxes into the atmosphere and to compute the associated radiative impact on clouds. The model used in this study was developed in the framework of the general circulation model of the Laboratoire de Météorologie Dynamique and is fully described in Boucher et al. (2002b). Only aspects relevant to the DMS cycle are recalled here. Once emitted in the atmosphere, DMS is oxidized in the gas phase by OH and NO_3 radicals. Oxidation of DMS by NO_3 produces SO_2 and oxidation of DMS by OH produces either SO_2 or dimethylsulfoxide (DMSO). We also included the oxidation of DMS by O_3 in the gas phase (producing SO_2 only) and aqueous phase (producing DMSO only) as introduced in Boucher et al. (2002b). DMSO is oxidized in the gas phase by OH to produce SO_2 and methanesulfonic acid (MSA). No heterogeneous sink is included for DMS and DMSO. All reaction rates are given in Boucher et al. (2002a) and Boucher et al. (2002b). The monthly concentrations of oxidants are prescribed from the IMAGES model (Pham et al. 1995), except H_2O_2 which is calculated interactively in the model.

Fig. 1. The suite of models used for this study includes an atmosphere-ocean general circulation model, an oceanic biogeochemical scheme incorporating parameterizations of air-sea DMS emissions, and a model of the global atmospheric sulfur cycle.



2.3. Experimental design

Table 1 gives the list and characteristics of the seven experiments performed with our global model of the sulfur cycle. All the experiments use the same meteorology (i.e., the radiative impact of changing DMS emissions does not feedback on the meteorology). The different experiments also use the same sources of sulfur compounds other than marine DMS. In particular, the industrial sources of SO_2 are taken from the EDGAR 2.0 database and are representative of year 1990.

The seven experiments differ (1) by their marine DMS concentrations, taken from Bopp et al. (2003), (2) by the parameterization of air-sea DMS exchange (Nightingale 2000 or Liss and Merlivat 1986) used to compute interactively the DMS fluxes and (3) by the parameterization of the sulfate mass-cloud droplet number concentration (CDNC) relationship (Boucher and Lohmann, 1995).

In the 3 different control experiments (CONTROL, CTLgex and CTLcdn), marine DMS concentrations are set to their $1xCO_2$ value. In the “global warming” experiments (GW, GWgex and GWcdn), marine DMS concentrations are set to their $2xCO_2$ value and we correct the air-sea DMS flux by applying a scaling factor on the 10-m wind speed which accounts for the changes in wind speed as simulated in the OAGCM $1xCO_2$ and $2xCO_2$ experiments. This scaling factor is applied as a two-dimensional (latitude-longitude) monthly average and is estimated from the changes in wind stress in the OAGCM experiments. We also perform an additional sensitivity experiment

Table 1. Characteristics of the experiments performed in this study. Oceanic DMS concentrations for 1xCO₂ and 2xCO₂ and the wind speed correction factor are obtained from the work of Bopp et al. (2003). The sea-air transfer function for DMS emissions are from Nightingale (2000) or Liss and Merlivat (1986). The relationship between CDNC and the sulfate mass concentration are taken from Boucher and Lohmann (1995) and correspond to relationships A and D of this paper. Oceanic DMS fluxes are given in TgS yr⁻¹, DMS burdens in TgS and radiative impacts in W m⁻².

Experiment	CONTROL	GWdms	GW	CTLgex	GWgex	CTLcdn	GWcdn
Oceanic DMS concentration	1xCO ₂	2xCO ₂	2xCO ₂	1xCO ₂	2xCO ₂	1xCO ₂	2xCO ₂
Wind speed correction factor	-	-	2xCO ₂	-	2xCO ₂	-	2xCO ₂
Sea-air transfer function	Nightingale	Nightingale	Nightingale	Liss	Liss	Nightingale	Nightingale
Sulfate-CDNC relationship	D	D	D	D	D	A	A
Oceanic DMS emission flux	29.47	29.77 (+1%)	30.23 (+3%)	19.97	20.48 (+3%)	29.47	30.23 (+3%)
DMS burden	0.077	0.078	0.079	0.053	0.054	0.077	0.079
Radiative impact (90°S-90°N)	-	-0.03	-0.052	-	-0.048	-	-0.045
Radiative impact (40°S-60°S)	-	-0.25	-0.37	-	-0.35	-	-0.29

(GWdms) similar to GW but without the correction factor on wind speed in order to separate effects of the changes in marine DMS concentrations and wind speed. We do not account in these experiments for changes in the DMS air-sea transfer velocity induced by the change in sea surface temperature and sea-ice, but these have been estimated to be negligible.

For our baseline experiments (CONTROL and GW), we use the parameterization of Nightingale (2000) to compute the air-sea DMS fluxes. We also perform two additional experiments (CTLgex and GWgex) using the less sophisticated parameterization of Liss and Merlivat (1986) instead of Nightingale (2000) in order to test the robustness of our results to the choice of the gas exchange parameterization.

For our baseline experiments (CONTROL and GW), we use the relationship D of Boucher and Lohmann (1995) to relate the cloud droplet number concentration to the atmospheric sulfate mass concentration. This calculation is only diagnostic and therefore only includes the first indirect effect (change in cloud optical properties for a fixed liquid water content). Although very uncertain, the second indirect effect (due to a reduction in precipitation efficiency) may be of similar magnitude than the first indirect effect. The radiative impact on clouds due to changes in DMS emissions is computed as the difference in top-of-atmosphere radiative fluxes between the “global warming” and the control experiments (i.e., with DMS emissions at 2xCO₂ and 1xCO₂, respectively). The results are moderately sensitive to the assumed minimum background cloud droplet number concentration (prescribed here at the rather large value of 50 cm⁻³).

To test the robustness of our results to this relationship, we perform two additional experiments (CTLcdn and GWcdn) identical to CONTROL and GW except that we use the relationship A of Boucher and Lohmann (1995) to relate the cloud droplet

number concentration to the atmospheric sulfate mass. With the relationship D, there is only one relation, identical over the continents and the oceans, whereas relationship A distinguishes between land and ocean.

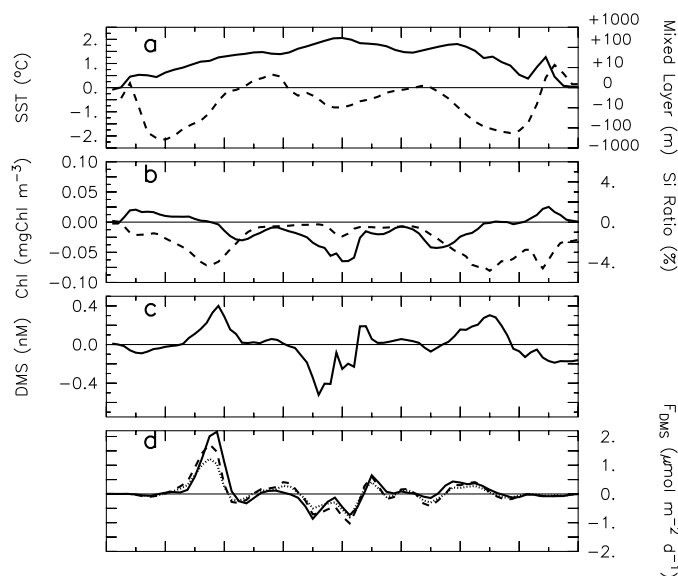
The model was run 18 months for each experiment and we present results for the last 12 months. The spin-up time of 6 months is long enough considering the short lifetime of aerosols.

3. Results and discussion

We first recall the main results of Bopp et al. (2003), in which the authors focus on the modeled marine productivity and marine DMS concentrations responses to climate change. The biogeochemical model used in this previous study predicts a 9% global decrease in the mean annual primary production and a small decrease of global sea-surface DMS concentrations ($\sim -1\%$). It also predicts opposing changes between the high and low latitudes (Figure 2b–2c). In the low latitudes, climate-induced changes (reduced nutrient supply caused by increased stratification of the upper ocean and decreased intensity of tropical upwellings) decrease marine production (-20%) and consequently lead to lower DMS concentrations. In the high latitudes, climate-induced changes (a longer growing season caused by increased stratification of the upper ocean) increase marine production (30%). In the 40°–60° band (north and south), the model also shows a poleward retreat of siliceous species (considered as low-DMS producers), and their replacement by non-siliceous species (considered as high-DMS producers). This shift in the phytoplankton ecosystem (depicted as the Si-ratio on figure 2b) is responsible for a DMS concentration increase in the mid latitudes at 2xCO₂.

Results, mechanisms, and uncertainties of the response of marine production and DMS concentrations to climate change

Fig. 2. Zonally-averaged change (global warming minus control) of (a) sea surface temperature (solid line, °C) and mixed layer depth (dashed line, m, log-scale), (b) surface chlorophyll (solid line, mg m⁻³) and relative abundance of siliceous phytoplankton species (Si-ratio, dashed line, %), (c) DMS sea-surface concentration (solid line, nM), (d) DMS flux to the atmosphere (solid line, $\mu\text{mol d}^{-1} \text{m}^{-2}$). For the last panel (d), the solid lines represent both effects of changes in marine DMS and wind speed (GW–CONTROL) while the dashed lines represent the effect of sea-surface DMS concentration changes alone (GWdms–CONTROL). On the same panel, the dotted line represents both effects (marine DMS and wind speed) but using the Liss and Merlivat (1986) parameterization (GWgex–CTLgex). Panels (a), (b) and (c) recall the main results of Bopp et al. (2003).



are described in more details in Bopp et al. (2001) and Bopp et al. (2003). Here, we investigate how these changes propagate up to sea-to-air DMS emissions, and to the atmospheric sulfur cycle.

3.1. DMS flux to the atmosphere

From our baseline experiments (CONTROL and GW) and on global average, the DMS flux to the atmosphere is increased by about 3%, from 29.47 Tg S yr⁻¹ at 1xCO₂ conditions to 30.23 Tg S yr⁻¹ at 2xCO₂ conditions (Table 1). But there is a large spatial heterogeneity in the change in DMS flux (Figure 3). In the tropical Pacific Ocean, the DMS fluxes are reduced by up to 10 $\mu\text{mol m}^{-2} \text{d}^{-1}$ (–50%). Conversely, in the subtropical and subantarctic zones, DMS fluxes are strongly enhanced by up to 6 $\mu\text{mol m}^{-2} \text{d}^{-1}$ (or 50%). These regional changes and the latitudinal opposition (Figure 2d) are driven by variations in DMS concentrations. Stronger winds in the Southern Ocean and weaker winds in the equatorial region at 2xCO₂ amplify the large heterogeneity. In the 40°S–60°S band for example, the model predicts almost a 20% increase in

the annual DMS flux. The effect of wind changes (estimated from GW-GWdms) alone accounts for one quarter of the total increase (a 5% increase) whereas the major effect is due to changes in sea-surface DMS concentrations (a 15% increase). On global average, the effects of changes in marine DMS and wind speed (estimated from GW, GWdms and CONTROL) account for 0.3 and 0.5 Tg S yr⁻¹ of this increase, respectively (Table 1). The effect of wind speed is small on zonal average compared to the effect of marine DMS, but relatively larger on global average because of the cancellation of the marine DMS effect between tropical and mid-latitude regions.

The additional experiments using the Liss and Merlivat (1986) parameterization for the air-sea gas exchange (CTLgex and GWgex) also show an increase of the global DMS flux to the atmosphere. But this increase (+0.55 TgS yr⁻¹) is only two third of the one simulated with the Nightingale (2000) parameterization (+0.78 TgS yr⁻¹) (Table 1 and figure 2d). As shown by Jones et al. (2001) and Boucher et al. (2002a), the use of Liss and Merlivat (1986) instead of Nightingale (2000) parameterization for the air-sea gas exchange also reduces the mean global DMS flux (from 29.47 to 19.97 TgS yr⁻¹) in the control experiment. Thus, the relative increase of DMS flux is almost identical in the two global warming experiments (+3%).

3.2. Atmospheric DMS and sulfate

At 2xCO₂ and for our baseline scenario (GW), the mean atmospheric DMS burden is very similar to its 1xCO₂ value (0.077 TgS at 1xCO₂ vs. 0.079 TgS at 2xCO₂). The DMS lifetime is also unchanged at about 0.9 day. The small increase in the DMS burden is accompanied by a very small decrease in the sulfate burden, probably because of a shift of sulfate production in regions where wet scavenging is more efficient.

However, the differences in the spatial distribution of DMS emissions result in large regional differences of atmospheric DMS concentrations. The changes in the distribution of DMS mixing ratios at the surface follow the changes in marine DMS emissions. In the Southern Ocean, DMS mixing ratio is increased by up to 50 pptv (or 25%). It is decreased by up to 40 pptv (or –50%) in the western Equatorial Pacific Ocean.

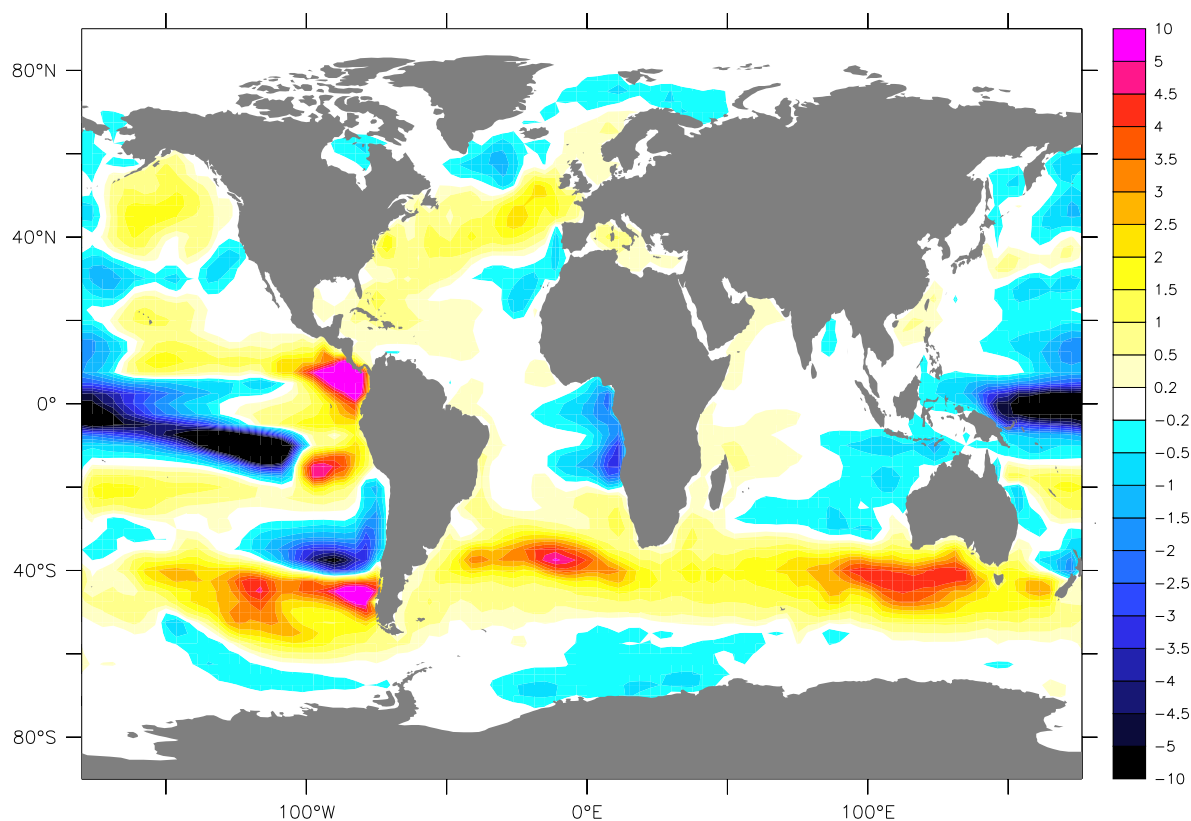
Again, changes in atmospheric DMS concentrations computed using the Liss and Merlivat (1986) parameterization (GWgex minus CTLgex) are considerably lower than in our baseline scenario, but very similar in terms of relative changes.

3.3. Indirect radiative effect

The radiative impact in clear sky (or direct effect) induced by the changes in sulfate aerosol burden is negligible. As mentioned above, we focus here on the radiative impact in cloudy sky and consider only the first indirect radiative effect (changes in cloud optical properties for fixed liquid water content).

The change in radiative flux between the GW and CONTROL experiments (with DMS emissions set to their 2xCO₂ and 1xCO₂ values, respectively) is very small at –0.052 Wm⁻². Again, this modest global figure conceals large spatial and temporal heterogeneities. In regions where DMS emissions are increased (decreased) by global warming, sulfate mass and the cloud droplet number concentration increase (decrease). Through the associated changes in cloud optical properties, this increase

Fig. 3. Change in DMS flux between the CONTROL and GW experiments. Unit is $\mu\text{mol d}^{-1} \text{m}^{-2}$.



(decrease) in DMS emission leads to an albedo increase (decrease) and a negative (positive) impact on the radiative budget. The radiative impact of DMS at $2\times\text{CO}_2$ reaches 1 Wm^{-2} in the western Equatorial Pacific Ocean and off the west coasts of Angola and Chile whereas it reaches -1 Wm^{-2} in the South Atlantic Ocean (Figure 4, upper panel).

On figure 4, we compare our estimate of the radiative impact of changes in DMS emissions to the radiative forcing of CO_2 at $2\times\text{CO}_2$, computed with the LMD atmospheric GCM (Laurent Li, personal communication). In the subantarctic Southern Ocean, the radiative impact caused by changes in DMS emissions may partly counteract the CO_2 radiative forcing (-0.5 Wm^{-2} vs 2.5 Wm^{-2} in zonal annual means). Moreover, the DMS radiative impact differs from the CO_2 radiative forcing by its seasonality (Figure 5). Whereas CO_2 radiative forcing is more or less constant over the year, the DMS radiative impact reaches its maximum in summer, with values up to -1.5 Wm^{-2} in the 40°S – 60°S band (Figure 5), because marine DMS emissions are maximum in summer.

There are several reasons why the symmetry in marine DMS

change between the northern (NH) and southern (SH) hemispheres (Figure 2c) does not translate into a similar symmetry in the radiative impact (Figure 2e). In the first place, the surface covered by the ocean is less in the NH than in SH. In the second place, the amplification of the change in DMS flux due to changes in wind speed occurs only in the SH (Figure 2d). Finally, the radiative impact is expected to be negligible in polluted regions where the cloud droplet number concentration and the indirect aerosol forcing are already large (Figure 6). The radiative impact induced by changes in DMS was estimated assuming constant anthropogenic emissions of SO_2 over the period of doubling CO_2 concentrations. These emissions and those of other aerosol types are expected to change in the future. There are observed negative trends in sulfate aerosol emissions and concentrations over Europe and North America (Boucher and Pham, 2002), while emissions are expected to continue to grow over Asia and some other regions of the world. If this trend continue, it would result in a positive radiative impact in the mid-latitudes of the NH thus strengthening the hemispheric contrast observed in Figure 4 (upper panel).

Fig. 4. Annually-averaged radiative impact on clouds (first indirect effect only) due to changes in DMS fluxes associated with a CO₂ doubling (GW minus CONTROL, upper panel). The corresponding radiative forcing due to CO₂ (from 1xCO₂ or 350 ppmv to 2xCO₂ or 700 ppmv) is also shown for comparison (lower panel). Unit is Wm⁻².

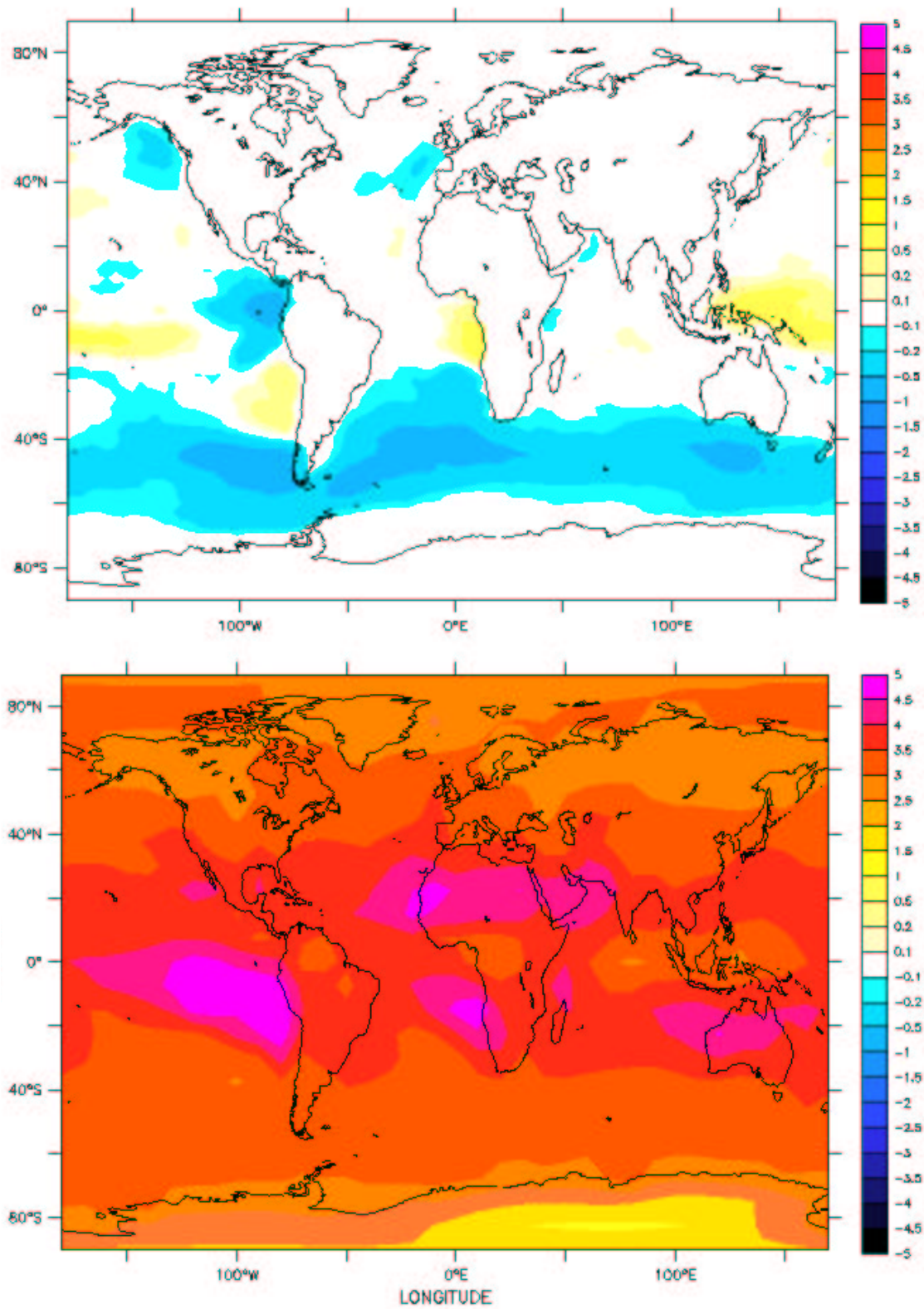


Fig. 5. Latitude-time diagram of the radiative impact (first indirect effect) induced by changes in the DMS flux (GW minus CONTROL, zonal mean, unit is Wm^{-2}).

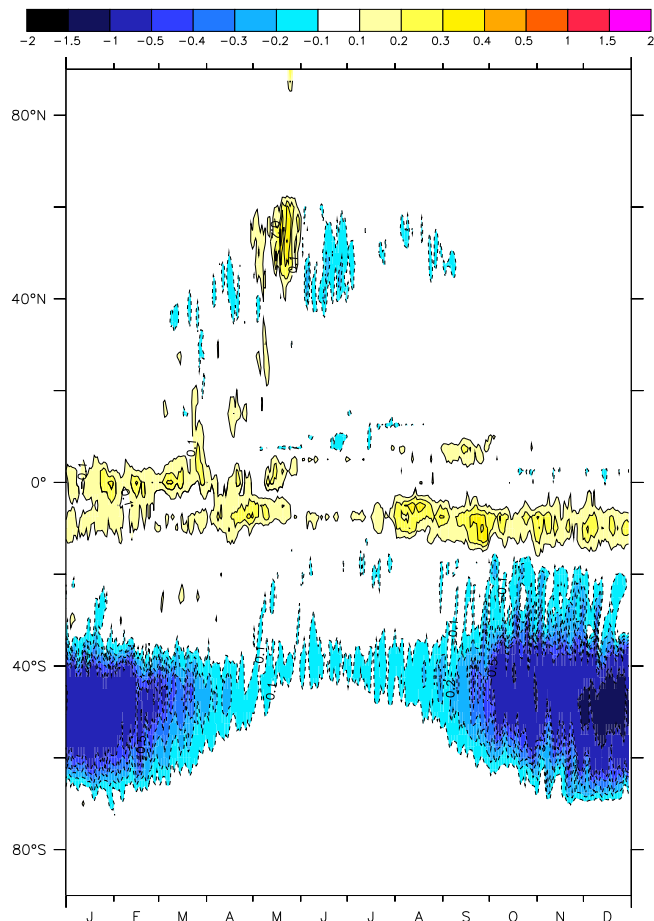


Fig. 6. Indirect radiative forcing due to anthropogenic sulfate from present-day relative to pre-industrial conditions, from Boucher and Pham (2002). Unit is W m^{-2} .

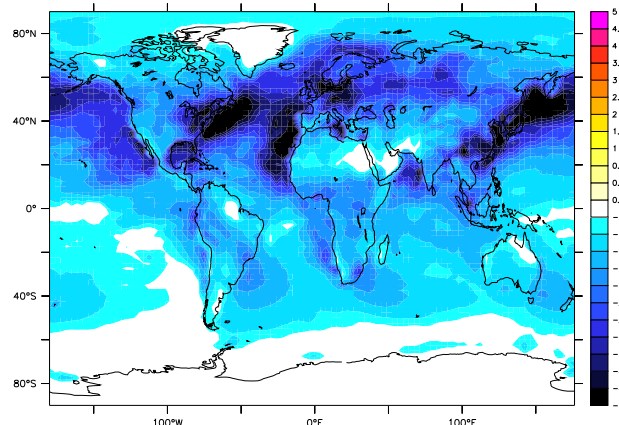
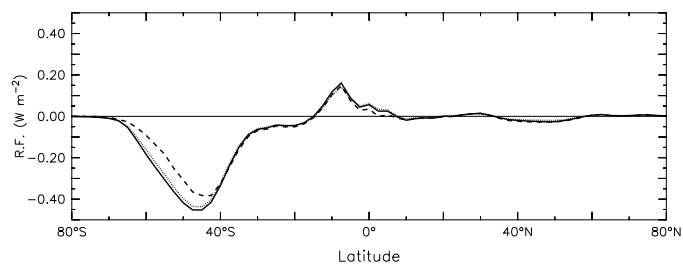


Fig. 7. Zonally-averaged change (global warming minus control) of radiative impact due to changes in DMS emissions (in Wm^{-2}). Different sensibility tests are shown: solid line is GW minus CONTROL, dashed line is GW_{gex} minus CTL_{gex} and dotted line is GW_{cdn} minus CTL_{cdn} .



The additional sensitivity experiments we performed enable us to test the robustness of our results to the choice of (1) the air-sea flux parameterization and (2) the sulfate mass–cloud droplet number relationship. Interestingly, our results shows a similar radiative impact no matter how we parameterize the air–sea exchange of DMS (Nightingale 2000 or Liss and Merlivat 1986) whereas the absolute changes in DMS emissions and atmospheric concentrations are not the same in the 2 cases. In the unpolluted marine air of the Southern Ocean, a smaller increase in DMS emissions with Liss and Merlivat (1986) parameterization than with Nightingale (2000) results in the same radiative impact at $2\times\text{CO}_2$ (around -0.35 W m^{-2}). Because DMS emissions at $1\times\text{CO}_2$ are significantly reduced when the Liss and Merlivat parameterization is used, the atmospheric sulfur cycle is more sensitive to changes in DMS emissions in the Liss and Merlivat case.

Second, our results show that using the relationship A of Boucher and Lohmann (1995) instead of relationship D significantly reduces the absolute values of the radiative impact due to changes in DMS emissions. This is particularly true in the Southern Ocean (-0.29 W m^{-2} vs. -0.37 W m^{-2} , Table 1). With the relationship A that distinguishes maritime and continental air, the slope of the log-log regression lines between the sulfate mass concentration and the cloud droplet number concentration is reduced over oceans and increased over continents. Accordingly, a change in marine DMS emissions will have less effect on the cloud droplet number concentration and the radiative budget. However, our main conclusions concerning the radiative impact of changes in DMS emissions caused by global warming remain unchanged: the global radiative impact is very small (around -0.05 W m^{-2}), but is significant (around $-0.3/-0.4 \text{ W m}^{-2}$) in the unpolluted Southern Ocean.

4. Conclusion

Following previous work on modeling the marine DMS response to climate change (Bopp et al. 2003) and using an atmospheric model of the global sulfur cycle, we estimate the impact of future climate change on DMS fluxes to the atmosphere and the associated radiative impact through changes in cloud albedo. At $2\times\text{CO}_2$, the DMS flux is similar to its $1\times\text{CO}_2$ value (at about 29 Tg S yr^{-1}), however it displays large regional contrasts: a reduction (-15%) of the DMS flux in the tropical band and an increase (30%) in the mid latitudes of the Southern hemisphere. In this paper, we focus on how those changes propagate up to the atmospheric sulfur cycle and to the Earth's radiative budget.

Changes in marine DMS emissions at $2\times\text{CO}_2$ significantly impact the regional radiative budget, up to -1.5 W m^{-2} in summer between 40°S and 50°S . In the Southern Ocean, this radiative impact may partly cancel out the radiative forcing of anthropogenic CO_2 ($2.5/3 \text{ W m}^{-2}$ at $2\times\text{CO}_2$). The strength of the DMS-CCN-cloud albedo feedback is nevertheless small at the global scale.

We have tested the robustness of our results to some of the parameterizations we employ. In particular, we show that the use of the Liss and Merlivat (1986) parameterization for the air-sea gas exchange instead of the more recent Nightingale (2000) parametrization does not affect our results in terms of

radiative impact. Second, we show that the use of an other parameterization for the relationship between the sulfate mass concentration and the cloud droplet number may slightly modify the magnitude of the estimated impact, but that our conclusions remain the same.

Our results strongly depends on the skill of our models to simulate marine productivity, on the relationships we use to predict DMS from biological variables, and on the parameterization of the aerosol indirect effect used in the atmospheric model of the sulfur cycle. Improvements in all aspects will be necessary to develop greater confidence in such future predictions. In particular, the role of sea-salt particles as CCN and the potential change in sea-salt induced by changes in wind speed should also be considered. Whereas such a study must be viewed as a sensitivity study more than a prediction, it represents a first step toward the assessment of the DMS-cloud albedo climate feedback loop.

5. Acknowledgments

This work was supported by the Environment and Climate Programme of the European Community (IRONAGE contract EVK2-1999-00227 and PHOENICS contract EVK2-CT-2001-00098) and the French national programmes PROOF and PNCA of CNRS/INSUE. Computing time was provided by CNRS/IDRIS under projects 020040 and 021167. The Laboratoire d'Optique Atmosphérique is an institute of the "Fédération de Recherche" FR1818 of the CNRS.

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