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

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Abstract:

Dimethylsulfide (DMS) is a biogenic compound produced in sea-surface water and outgased to the atmosphere. Once in the atmosphere, DMS is a significant source of cloud condensation nuclei (CCN) in the unpolluted marine atmosphere. It has been postulated that climate may be partly modulated by variations in DMS production through a DMS-CCN-cloud albedo feedback. We present here a first modelled estimate of the radiative impact due to changes in DMS air-sea fluxes caused by global warming. A suite of models, including an atmosphere-ocean general circulation model (AOGCM), a marine biogeochemical scheme incorporating a parameterization of DMS emissions to the atmosphere, and an atmospheric model of the global sulfur cycle, are used to simulate the responses of DMS sea-to-air flux and its associated radiative impact to increased greenhouse gase concentration (a 1% increase per year in atmospheric CO_2 until the present-day concentration has doubled). At $2xCO_2$, 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, this radiative impact may partly alleviate the radiative forcing due to anthropogenic CO_2 (2.5/3 Wm^{-2} at $2xCO_2$ compared to $1xCO_2$ conditions).

1. Introduction

Dimethylsulfide (DMS) is the most abundant volatile sulfur-compound at the sea surface and has a strong marine phytoplanktonic origin. Once in the atmosphere, DMS is oxidized and contributes to form sulfate aerosol particles, which may affect the radiative budget as cloud condensation nuclei (CCN). Recently, Jones et al. (2001) and Boucher et al. (2002a) have shown in global model studies that the indirect radiative forcing due to anthropogenic sulfate aerosols was significantly influenced by assumptions made about the marine DMS emission flux. This is because an increase in the DMS flux causes an increase in the concentration of sulfate aerosols, thus increasing background CCN concentrations and reducing the cloud susceptibility to anthropogenic (sulfate) aerosols.

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). 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 circulation model together with a DMS production model to investigate the response of DMS emissions to climate change. Based on a $10°\times20°$ 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 CO_2 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. Penner et al. (2001) did not estimate the impact of such changes on the radiation budget.

In a previous paper (Bopp et al. 2002), a model of the global distribution of sea-surface DMS concentrations (Aumont et al. 2002) was coupled to an atmosphere-ocean general circula-

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tion 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.

2. Method

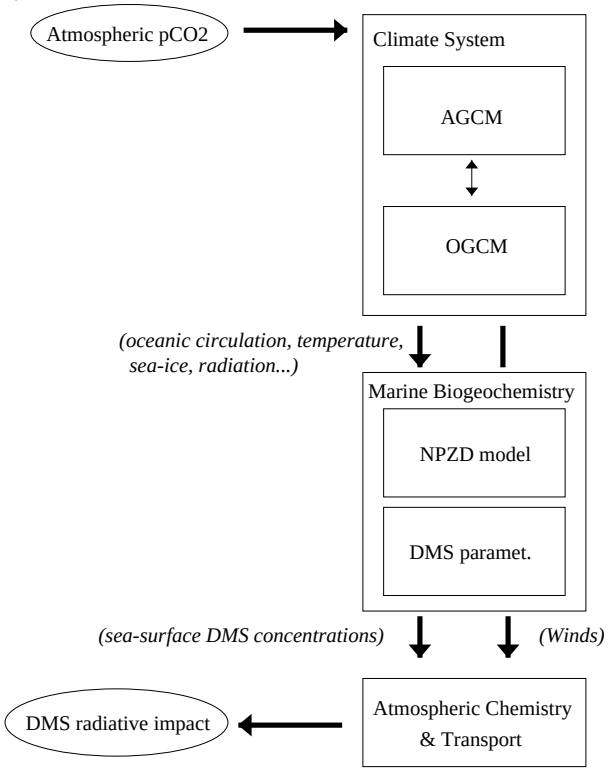
2.1. Marine DMS emissions

Sea-water DMS concentrations for $1xCO_2$ and $2xCO_2$ conditions are obtained from the work of Bopp et al. (2002). (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. 2002). 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 and experiment design

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 oxidised 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

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.



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the IMAGES model (Pham et al. 1995), except H_2O_2 which is calculated interactively in the model.

Three different experiments were carried out with our global model of the sulfur cycle. All three experiments use the same meteorology (i.e., the radiative impact of changing DMS emissions does not feedback on the meteorology). The three 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. Thus the three experiments differ only by their marine DMS emissions which are recomputed interactively in the model using the marine DMS concentrations from Bopp et al. (2002) and the parameterization of Nightingale et al. (2000). In the control experiment (CONTROL), marine DMS concentrations are set to their $1xCO_2$ value. In the "global warming" experiment (GW), 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 a third sensitivity experiment (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 DMS flux induced by the change in sea surface temperature and sea-ice, but these have been estimated to be negligible. 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.

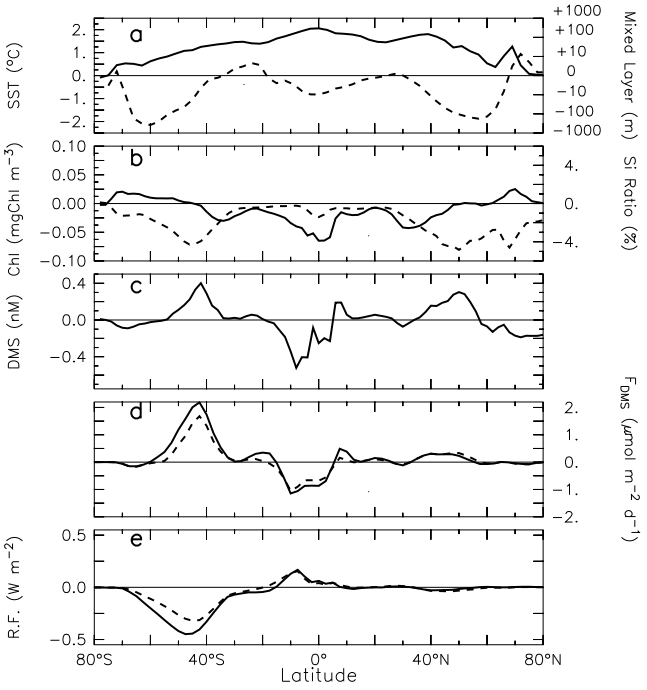
We follow Boucher and Lohmann (1995) and Boucher and Pham (2002) to estimate the cloud properties from the sulfate mass concentration. This calculation is only diagnostic and therefore only includes the first indirect effect (change in cloud optical properties for a fixed liquid water content). Although 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 $2xCO_2$ and $1xCO_2$, 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}). While improvements on the parameterisation of the aerosol indirect effects are desirable (Lohmann et al. 2000), our parameterisation is still useful to perform the sensitivity experiments of the present study.

3. Results and discussion

3.1. Marine DMS production

At $2xCO_2$, the model 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).

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 phytoplakton species (Si-ratio, dashed line, %), (c) DMS sea-surface concentration (solid line, nM), (d) DMS flux to the atmosphere (solid line, μmol d^{-1} m^{-2}), and (e) associated radiative impact (solid line, Wm^{-2}). For the last two panels (d and e), 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).



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 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 $2xCO_2$.

Results, mechanisms, and uncertainties of the response of marine production and DMS concentrations to climate change are described in more details in Bopp et al. (2001) and Bopp et al. (2002). Here, we investigate how these changes propagate up to sea-to-air DMS emissions, and to the atmospheric sulfur cycle.

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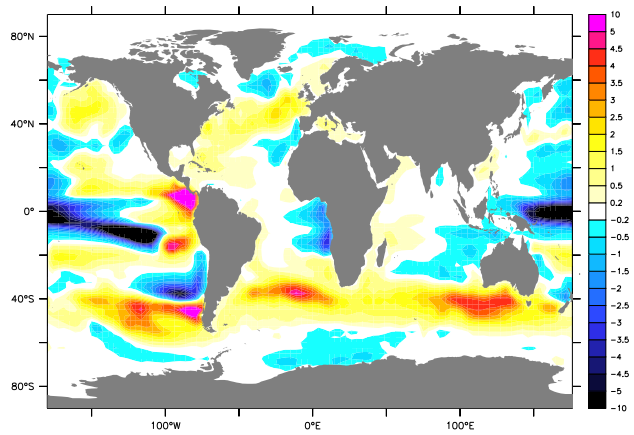
Fig. 3. Change in DMS flux between the CONTROL and GW experiments. Unit is μmol d^{-1} m^{-2}.



3.2. DMS flux to the atmosphere

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 μmol m^{-2} d^{-1} (-50%). Conversely, in the subtropical and subantarctic zones, DMS fluxes are strongly enhanced by up to 6 μmol m^{-2} 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 2xCO_2 amplify the large heterogeneity. On global average, the DMS flux to the atmosphere is increased by about 3%, from 29.1 Tg S yr^{-1} at 1xCO_2 conditions to 29.9 Tg S yr^{-1} at 2xCO_2 conditions. The effects of changes in marine DMS and wind speed account for 0.3 and 0.5 Tg S yr^{-1} of this increase, respectively. 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.

In the 40°S–50°S band, the model predicts a 20% increase in the annual DMS flux. The effect of wind changes 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).

3.3. Atmospheric DMS and sulfate

At 2xCO_2, the mean atmospheric DMS burden is very similar to its 1xCO_2 value (0.076 TgS at 1xCO_2 vs. 0.078 TgS at 2xCO_2). 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 sulphate 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 more or less 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.

3.4. 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 our two experiments (with DMS emissions set to their 2xCO_2 and 1xCO_2 values) is very small at -0.05 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 (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 2xCO_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 2xCO_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).

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 SH than in NH. 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 strenghtening the hemispheric contrast observed in Figure 4 (upper panel).

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Fig. 4. Annually-averaged radiative impact on clouds (first indirect effect only) due to changes in DMS fluxes associated with a CO_2 doubling (upper panel). The corresponding radiative forcing due to CO_2 (from $1xCO_2$ or 350 ppmv to $2xCO_2$ or 700 pmv) is also shown for comparison (lower panel). Unit is Wm^{-2}).

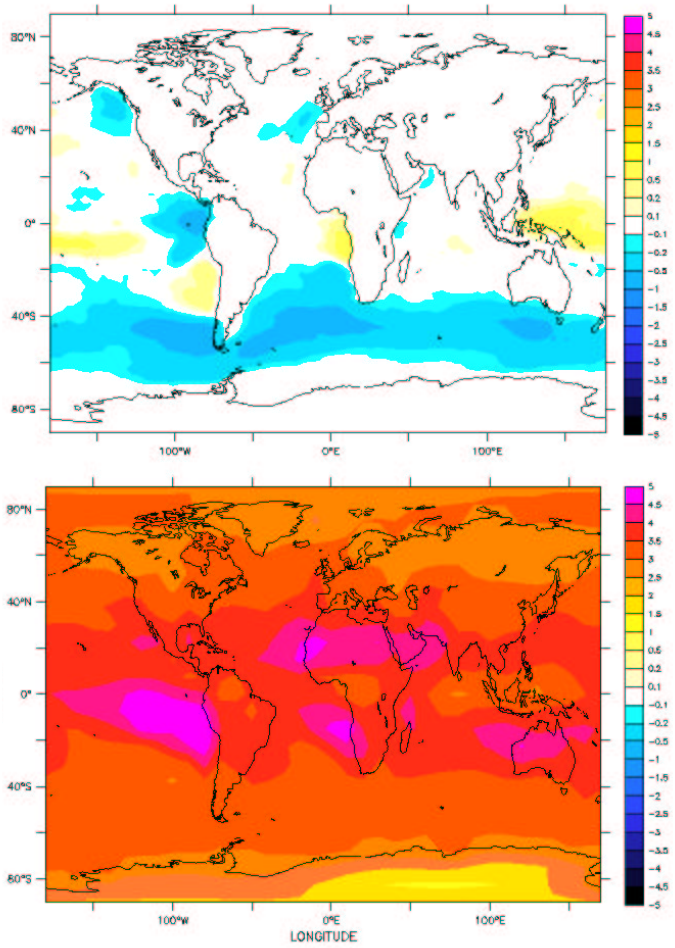

Fig. 5. Latitude-time diagram of the radiative impact (first indirect effect) induced by changes in the DMS flux ($2xCO_2$ minus $1xCO_2$, zonal mean, unit is Wm^{-2}).

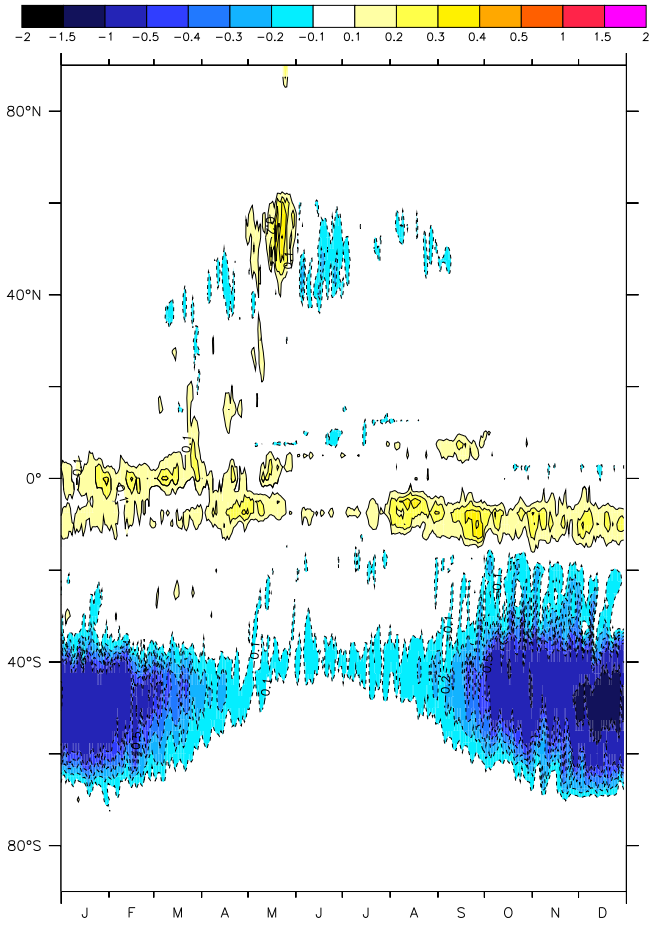

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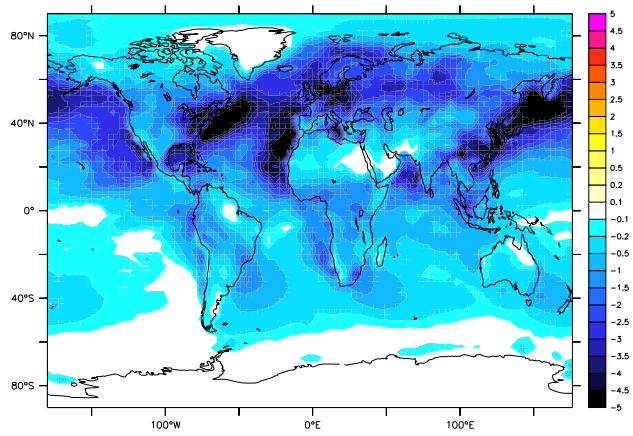
Fig. 6. Indirect radiative forcing due to anthropogenic sulfate from present-day relative to pre-industrial conditions. Unit is Wm^{-2}.



4. Conclusion

Future climate changes are likely to affect the ocean circulation, the marine biological productivity, and in turn the marine and atmospheric sulfur cycle. Here, using (1) a coupled ocean-atmosphere model forced by increasing atmospheric CO_2, (2) a marine biogeochemical scheme, (3) data-based parameterizations of sea-surface DMS, and (4) an atmospheric model of the global sulfur cycle, we could estimate the impact of future climate change on DMS fluxes to the atmosphere and the associated radiative impact through changes in cloud albedo. At $2xCO_2$, the DMS flux is similar to its $1xCO_2$ value (at about 29 Tg S yr^{-1}), however we found large regional contrasts. Main results are a reduction (-15%) of the DMS flux in the tropical band and an increase (30%) in the mid latitudes of the Southern hemisphere. Those regional changes are mainly driven by a chlorophyll decrease at low latitudes and by a shift of phytoplankton species (from diatoms to other species more efficient to produce DMS) at mid latitudes.

Those changes propagate to the DMS atmospheric cycle and significantly impact the regional radiative budget, up to $-1.5\,Wm^{-2}$ 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 Wm^{-2} at $2xCO_2$). The strength of the DMS-CCN-cloud albedo feedback is nevertheless small at the global scale.

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 towards the assessment of the DMS-cloud albedo climate feedback loop.

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