

Environmental constraints on the production and removal of the climatically active gas dimethylsulphide (DMS) and implications for ecosystem modelling

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Abstract Seawater concentrations of the climate-cooling, volatile sulphur compound dimethylsulphide (DMS) are the result of numerous production and consumption processes within the marine ecosystem. Due to this complex nature, it is difficult to predict temporal and geographical distribution patterns of DMS concentrations and the inclusion of DMS into global ocean climate models has only been attempted recently. Comparisons between individual model predictions, and ground-truthing exercises revealed that information on the functional

relationships between physical and chemical ecosystem parameters, biological productivity and the production and consumption of DMS and its precursor dimethylsulphoniopropionate (DMSP) is necessary to further refine future climate models. In this review an attempt is made to quantify these functional relationships. The description of processes includes: (1) parameters controlling DMSP production such as species composition and abiotic factors; (2) the conversion of DMSP to DMS by algal and bacterial enzymes; (3) the fate of DMSP-sulphur due to, e.g., grazing, microbial consumption and sedimentation and (4) factors controlling DMS removal from the water column such as microbial consumption, photo-oxidation and emission to the atmosphere. We recommend the differentiation of six phytoplankton groups for inclusion in future models: eukaryotic and prokaryotic picoplankton, diatoms, dinoflagellates, and other phytoflagellates with and without DMSP-lyase activity. These functional groups are characterised by their cell size, DMSP content, DMSP-lyase activity and interactions with herbivorous grazers. In this review, emphasis is given to ecosystems dominated by the globally relevant haptophytes *Emiliania huxleyi* and *Phaeocystis* sp., which are important DMS and DMSP producers.

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Introduction

Dimethylsulphide (DMS) is a semivolatile organic sulphur compound that accounts for 50–60% of the total natural reduced sulphur flux to the atmosphere, including emissions from volcanoes and from vegetation (Andreae 1990; Bates et al. 1992; Spiro et al. 1992). By providing 95% of the flux to the atmosphere, the oceans are the main source for DMS, with estimates of its emission ranging between 15 and 33 Tg S y⁻¹ (Kettle and Andreae 2000). In the late 1980s, the hypothesis that DMS is involved in the biological regulation of global climate was put forward (Bates et al. 1987; Charlson et al. 1987). It is, however, only recently that DMS has been incorporated in global climate models (e.g., Aumont et al. 2002; Bopp et al. 2003, 2004; Gabric et al. 2004; Kettle and Andreae 2000; Simo and Dachs 2002). After emission to the atmosphere, this volatile sulphur compound is oxidised to sulphur dioxide (SO₂) and other products. From SO₂, non-sea-salt (nss) sulphate is produced, which can form sulphate (SO₄²⁻) particles that act as condensation nuclei for water vapour. These nuclei affect the radiative properties of the atmosphere and clouds, with implications for climate. Higher numbers of condensation nuclei will deflect more incoming solar radiation back into space and thereby reduce the temperature on earth. The hypothesis that this process may modulate the greenhouse effect of increased anthropogenic CO₂ input to the atmosphere, was indirectly supported by the modelling results of the effect of anthropogenic SO₂ input to the atmosphere (Andreae et al. 2005; Mitchell et al. 1995). Although this study gave a rough indication of the counteracting effects of atmospheric SO₂ and CO₂, a quantitative understanding of all sources and sinks of atmospheric aerosols is still lacking (Andreae and Crutzen 1997).

Currently, anthropogenic SO₂ production exceeds natural SO₂ production by a factor of 3 (Bates et al. 1992), but the impact of the former on aerosol production is largely confined to industrialised areas of the Northern Hemisphere. The oceans, on the other hand, cover approximately 70% of the Earth's surface and much of this area is remote from man-made atmospheric contaminants. Consequently, the exchange of marine DMS is of high regional importance and may affect climate globally. Since the publication of a global inventory of DMS data by

Kettle and co-authors (Kettle et al. 1999), it has become possible to include DMS in global climate models. Indeed, recent model calculations have shown that in the Southern Hemisphere, where anthropogenic sulphate emission is low, DMS plays a major role in the production of atmospheric nss-sulphate (Gondwe et al. 2003). Gondwe et al. calculated that the contribution of DMS to the total (global) atmospheric nss-sulphate burden is 18% and that it shows significant regional and temporal differences; e.g., in the Southern Hemisphere its annual contribution is 43% and over the Southern Ocean it is in excess of 80% during summer.

In one of the early attempts to add DMS in a global ocean climate model, Bopp et al. (2003) showed that a doubling of the atmospheric CO₂ concentration resulted in a reduction of the DMS flux at low latitudes and in enrichment at mid-latitudes. Thus, depending on the sign of the change in DMS flux, the subsequent climate forcing by sulphur products could either alleviate or amplify the greenhouse effect (Bopp et al. 2004). The inclusion of DMS in the model was achieved by coupling the production of dimethylsulphoniopropionate (DMSP) and its conversion to DMS with the trophic status of the ecosystem, which in turn was based on the silica ratio, defined as the local simulated production of biogenic silica relative to the maximum production and is related to the proliferation of diatoms. It is computed from local silica concentrations and a variable Si:C ratio, which depends on the silica concentration. Although simplistic in its ecological approach, this study showed that an increased CO₂ concentration does not necessarily result in increased DMS production that may counterbalance the greenhouse effect, as suggested by the Charlson-Lovelock-Andreae-Warren (CLAW) hypothesis (Charlson et al. 1987). Clearly, an improved understanding of the biological processes is necessary to address the role of DMS in climate feedback mechanisms.

The production of DMS is almost exclusively through biogenic processes and shows strong seasonal and latitudinal variation (Kettle et al. 1999). DMS mainly results from the enzymatic cleavage of DMSP, a compound that is produced in several groups of marine phytoplankton. A complex network of production and consumption pathways of both DMSP and DMS involves most of the microbial food web (Fig. 1) and determines the concentration of