



Vertical variability of seawater DMS in the South Pacific Ocean and its implication for atmospheric and surface seawater DMS

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ABSTRACT

Shipboard measurements of atmospheric dimethylsulfide (DMS) and sea surface water DMS were performed aboard the R/V Onnuri across the South Pacific from Santiago, Chile to Fiji in February 2000. Hydrographic profiles of DMS, dissolved dimethylsulfoniopropionate (DMSP_d), and particulate DMSP_p in the upper 200 m were obtained at 16 stations along the track. Atmospheric and sea surface water DMS concentrations ranged from 3 to 442 pptv and from 0.1 to 19.9 nM, respectively; the mean values of 61 pptv and 2.1 nM, respectively, were comparable to those from previous studies in the South Pacific. The South Pacific Gyre was distinguished by longitudinal-vertical distributions of DMS, DMSP_d, and DMSP_p, which was thought to be associated with the characteristic modification of biological activities that occurs mainly due to significant change in water temperature. The averaged DMS maximum appeared at 40 m depth, whereas DMSP_p and DMSP_d maxima coincided with that of dissolved oxygen content at 60–80 m. The sea-to-air fluxes of DMS were estimated to be 0.4–11.3 μmol d⁻¹ m⁻² (mean = 2.8 μmol d⁻¹ m⁻²). A fairly good correlation between atmospheric DMS and sea-to-air DMS flux indicated that atmospheric DMS concentration was more sensitive to change in physical parameters than its photochemical removal process or surface seawater DMS concentrations.

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1. Introduction

Dimethylsulfide (DMS), the most dominant sulfur species throughout the ocean, is formed by enzymatic cleavage of dimethylsulfoniopropionate (DMSP), which is produced by a variety of phytoplankton species. Most DMSP is consumed by bacteria and only a fraction is used to produce DMS (Kiene, 1996). Although DMS is removed by bacterial consumption (Kiene and Bates, 1990), sea surface layers are always supersaturated with it, which implies a net flux of DMS to the atmosphere (Huebert et al., 2004). As a result, approximately 1% of the DMSP produced in sea water is transported to the air in the form of DMS through sea-to-air flux (Bates et al., 1994; Simó and Pedrós-Alió, 1999).

After being released into the atmosphere, DMS is readily oxidized to non-sea-salt sulfate (nss-SO₄²⁻) and methane sulfonate (MSA) in the atmospheric boundary layer. Atmospheric DMS is mainly oxidized by OH during the day and nitrate radical (NO₃) at night. The oxidation of atmospheric DMS seems to contribute

largely to the formation of aerosol containing nss-SO₄²⁻ in the marine troposphere. According to the CLAW hypothesis (Charlson et al., 1987), sulfate and MSA produced from oceanic DMS affect the Earth's radiation balance through the formation of cloud condensation nuclei (CCN), thereby altering cloud properties. The overall effect of these couplings on climate is negative feedback, meaning that it tends to stabilize the climate. In recent studies, DMS was positively correlated with atmospheric CCN (Vallina et al., 2006, 2007) and solar radiation (Toole and Siegel, 2004; Vallina and Simó, 2007) over most of the global ocean, which supports the DMS–climate feedback loop for open-ocean environments.

Researchers estimate that the gaseous DMS flux from the ocean to the atmosphere lies between 23 and 35 Tg S yr⁻¹ (Kettle and Andreae, 2000; Simó and Dachs, 2002; Kloster et al., 2006). The oceanic DMS flux comprises ~30% of global sulfur sources (IPCC, 2001) and its contribution to global nss-SO₄²⁻ is 27%, both of which are similar in magnitude (Kloster et al., 2006). The mean annual contribution of DMS to the climate-relevant nss-SO₄²⁻ column burden is the greatest (43%) in the relatively pristine Southern Hemisphere, where a lower oxidative capacity of the atmosphere, a larger sea-to-air transfer of DMS, and a larger surface area of

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emission lead to an elevated atmospheric DMS burden (Gondwe et al., 2003). Therefore, the vast area of the South Pacific is the key region in which to test the validity of the CLAW hypothesis.

DMS flux from the ocean has been estimated through the parameterization of wind fields and the maps of DMS concentrations in the global ocean (Liss and Merlivat, 1986; Wanninkhof, 1992; Kettle et al., 1999). Although Kettle et al. (1999) compiled a seawater DMS and DMSP database of more than 15 000 measurements over the global ocean, the temporal and spatial coverage of DMS is still poor. To obtain a global view of DMS distribution by time, determining oceanic DMS concentration has to be approached using various empirical parameterizations of field observation datasets, such as chlorophyll *a* distribution (Anderson et al., 2001), climatological mixed layer depth (Simó and Dachs, 2002), and SeaWiFS ocean color measurements (Belviso et al., 2006).

Kettle et al. (1999) found no significant correlations between DMS and other oceanographic parameters and no simple algorithm to create temporal fields of sea surface DMS concentrations based on these parameters. Thus, to reduce the great uncertainty inherent in estimates of DMS flux, more measurements with greater temporal and spatial resolution are necessary. This is particularly true for the South Pacific, where measurements of DMS and DMSP are still very sparse. DMS in the South Pacific has been studied most extensively by Bates and his group (Bates and Quinn, 1997; Bates et al., 1998; Bates, 2004). Most of their research, however, has been concentrated on the equatorial Pacific, which is a region that exhibits relatively high DMS emissions throughout the year (Bates and Quinn, 1997). Unlike the equatorial Pacific, the central South Pacific (20–50°S) should have large seasonal and spatial variations of DMS levels due to distinct seasonality and latitudinal variations in sea surface temperature. The central South Pacific has an area of $30 \times 10^6 \text{ km}^2$ and covers about 8% of all oceans and seas worldwide, yet only four sets of latitudinal transit data are available for this region in the Global Surface Seawater DMS Database (Bates, 2004). Convincing evidence also exists for the seasonality of DMSP and DMS concentrations and DMS flux in the Southern Ocean (Simó and Dachs, 2002; Vallina et al., 2007).

Kettle et al. (1999) reported that having DMS measurements is not enough to explain the global DMS distribution, particularly in the South Pacific and Indian Ocean. To evaluate the role of DMS in climate change at regional to global scales also requires measurements of atmospheric and sea water DMS concentrations, quantification of its sea-to-air flux, and identification of the factors that control them. In this experiment, we concurrently measured sea water DMS, dissolved DMSP, and particulate DMSP at various depths, mainly within the thermocline, and the atmospheric DMS along the ship track from Chile to Fiji. We then characterized the behaviors of atmospheric DMS and its sea-to-air flux with regard to various factors such as sea surface DMS and DMSP, momentum flux, and mixed layer depth MLD. The results of this study will be useful to evaluate global ecosystem models for DMS production and to accurately determine the global DMS budget.

2. Methods

As a part of the Southern Pacific Ocean Dynamic Studies, measurements of atmospheric DMS and surface water DMS were made on board the research vessel (R/V) *Onnuri*, which left Punta Arenas, Chile on February 5, 2000 and arrived at Fiji on March 4, 2000. The study area lies between 20°S and 50°S and runs from the equatorial Pacific to the Southern Ocean, in which scarce sets of DMS and DMSP measurements are available. Fig. 1 shows the ship track and the stations at which hydrocast samples were taken. On February 20 and 21, hydrocast sampling was cancelled in order to keep the ship on schedule.

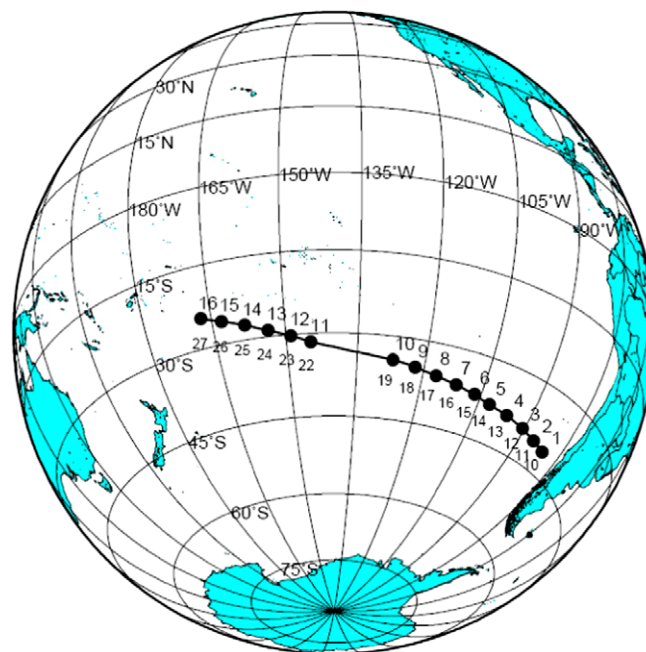


Fig. 1. Ship track and stations (closed circles) for vertical seawater sampling. Numbers above and below each station indicate the station number and date in February 2000, respectively.

For vertical profiles of DMS and DMSP, sea water samples were collected using 11 Niskin bottles from 3 m below the surface down to 200 m at 16 stations. During each hydrocast, conductivity, temperature, and depth were continuously determined with a CTD along with dissolved oxygen content. Water samples were taken from 3 m below surface down to 200 m at each station: seven samples between 3 m and 100 m, and four samples between 100 m and 200 m at a 25 m interval. The hydrocasts were conducted at dawn so that bacterial and phytoplankton populations would be minimally affected upon exposure to ultraviolet radiation (Kiene and Linn, 2000). Upon retrieval of the bottles, sea water was gently drawn from each Niskin bottle into a 130 mL DO bottle through tygon tubing; the new bottle was overflowed with sea water 2–3 times so that no air would be trapped inside. At each hydrocast station, surface water was collected using a bucket for the surface DMS measurement. Along the cruise track, surface seawater also was sampled 4–6 times a day for DMS analysis using the continuous seawater pumping system on the ship.

To measure sea water DMS, an aliquot of 30 mL was withdrawn from the DO bottle with a 50 mL syringe. The water-filled syringe then was connected to an air-tight filter holder equipped with a 47 mm Whatman GF/F glass fiber filter. By applying gentle pressure to syringe, the water sample was filtered into a 100 mL gas stripper bottle. Next, the filtrate was purged with high purity helium at 100 mL min^{-1} for 20 min and the purged air was passed through a Nafion dryer (Perma-Pure, Inc., USA) to remove water vapor. Finally, DMS was captured in a carboxisieve 300 adsorption trap (Supelco, USA). The dissolved DMSP (DMSP_d) in the purged filtrate and the particulate DMSP (DMSP_p) on the filter were converted to DMS using a strong alkaline solution for 1–2 h and then measured as DMS (Kim and Andreae, 1987). A common practice is to leave the particulate DMSP samples in the alkaline solution at least 12 h. As a result, our DMSP_p values were likely underestimated due to incomplete hydrolysis of DMSP_p . DMS and DMSP in sea water were measured right after the water samples were collected from the upper depths. Some samples from lower depths, however, could not be analyzed immediately and had to wait a maximum of 4 h; during this time they remained in the dark at

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