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Biogeochemistry of dimethylsulfide in the South China Sea

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ABSTRACT

The distribution of dimethylsulfide (DMS) was studied in surface seawater and vertical profiles at nineteen stations in the Nansha Islands sea area of the South China Sea. The concentrations of DMS in surface-layer (0–1 m) seawater vary from 64 to 140 ng S/L with high values found in the productive regions, in agreement with the horizontal distribution of chlorophyll *a*. The vertical profiles of DMS show a single peak shape with maximum concentrations occurring at depths between 30–75 m. The DMS concentrations are correlated with chlorophyll *a* levels both in the upper 20 m of seawater as well as in vertical profiles. A clear diel variation in DMS concentration is observed at the 50-m water layer at a fixed station with the highest DMS concentration found in the late afternoon. The DMS concentrations are associated with environmental factors such as seawater temperature, dissolved O₂ and nutrient contents. Although DMS is correlated to chlorophyll *a*, the phytoplankton species is a major factor responsible for the obviously higher DMS concentration than expected from the phytoplankton biomass in this sea area. The sea-to-air flux of DMS from this sea area is calculated to be 7.6 $\mu\text{mol m}^{-2} \text{d}^{-1}$.

1. Introduction

Over the past decade or so a lot of measurements have been made covering coastal, shelf, and open ocean environments, which show that dimethylsulfide (DMS) is ubiquitous in seawater and accounts for about 90% of the reduced volatile sulfur in surface seawaters (Aritsune *et al.*, 1992; Fogelqvist, 1991; Gibson *et al.*, 1994; Kiene, 1992; Leck and Bagander, 1988; Staubes and Georgii, 1992; Turner *et al.*, 1989; Wakeham *et al.*, 1987; Yang *et al.*, 1996). The amount of DMS in surface seawater is often much larger than that expected from equilibrium with the atmosphere (Andreae *et al.*, 1985; Bates *et al.*, 1987a, b; Cline and Bates, 1983; Dacey *et al.*, 1984; Lovelock *et al.*, 1972). The relatively high seawater concentrations of DMS lead to a concentration gradient between atmosphere and ocean that gives a net flux of DMS from the ocean to the atmosphere estimated to be in the range of 19.2–54.4 Tg S a⁻¹ globally (Andreae, 1990). This figure is of the same order of magnitude as the anthropogenic sulfur flux from fossil fuel combustion processes (Cullis and Hirschler, 1980; Möller, 1984). The DMS emitted by seawater into the atmosphere can be rapidly oxidized photochemically to sulfur dioxide, methanesulfonate and non-sea-salt

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sulfate (Andreae *et al.*, 1985; Berresheim *et al.*, 1990; Daykin and Wine, 1990; Grosjean and Lewis, 1982; Hatakeyama *et al.*, 1982, 1985; Putaud *et al.*, 1993; Yin *et al.*, 1986, 1990). These oxidation products of DMS may be a major source of marine aerosol particles, especially those which act as nuclei for the formation of water droplets in clouds, leading to increased albedo over the ocean (Charlson *et al.*, 1987; Malin *et al.*, 1992). Consequently, the production of DMS in the oceans probably has an important effect on the global climate (Bates *et al.*, 1987a). The oxidation products of DMS also contribute to the natural acidity of meteoric precipitation (Nguyen *et al.*, 1992; Nriagu *et al.*, 1987; Savoie and Prospero, 1989; Wagenbach *et al.*, 1988). Therefore, determination of DMS in seawater may serve as a starting point for accurate assessment of the relative roles of anthropogenic and biogenic sulfur sources in contributing to atmospheric sulfate formation, which can affect environmental acidification and the world's climate change.

Phytoplankton is generally believed to be the major source of DMS in the ocean (Andreae, 1986; Barnard *et al.*, 1984; Baumann *et al.*, 1994; Holligan *et al.*, 1987; Keller *et al.*, 1989). Variations in the concentration of DMS in the upper ocean are dependent on the release of β -dimethylsulfoniopropionate (DMSP) from phytoplankton populations, on the rate of enzymatic cleavage of DMSP to form DMS, and the strength of various DMS sinks. The release of DMSP from phytoplankton populations depends on species composition and abundance (Andreae, 1990; Nguyen *et al.*, 1988; Turner *et al.*, 1988), viral infection (Bratbak *et al.*, 1995; Malin *et al.*, 1992), and zooplankton grazing (Belviso *et al.*, 1990; Burkill *et al.*, 1987; Dacey and Wakeham, 1986; Morales *et al.*, 1991; Wakeham and Dacey, 1989). For these reasons, the correlations between parallel measurements of DMS and biological parameters such as chlorophyll *a* are not usually clear and definite (Andreae, 1990; Andreae and Barnard, 1984; Barnard *et al.*, 1984; Bates and Cline, 1985; Holligan *et al.*, 1987; Turner *et al.*, 1988).

The purpose of this work is as follows: to determine both the spatial and temporal distribution of DMS in the South China Sea from which no DMS analysis has been conducted previously, to investigate some of the biogeochemical factors governing this distribution, to obtain more information regarding the relationship between DMS and chlorophyll *a*, and to estimate the sea-to-air flux of DMS from this sea area.

2. Methods

a. Sample collection and treatments

The cruise tracks for sampling in the South China Sea aboard the R/V "Experiment No. 3" during Nov.–Dec. 1993 are shown in Figure 1. The study emphasis of this cruise took place in the Nansha Islands sea area of the South China Sea. Depth profile samples of 19 stations (marked as circles on the cruise track) were collected by standard hydrographic methods with PVC samplers mounted above a CTD apparatus. Once collected, subsamples were drawn into high-density polyethylene bottles and filled to the top to eliminate any head space in an effort to avoid sample degassing. As there were no facilities on board for DMS analysis, these subsamples were then treated according to the methods of Nguyen *et*

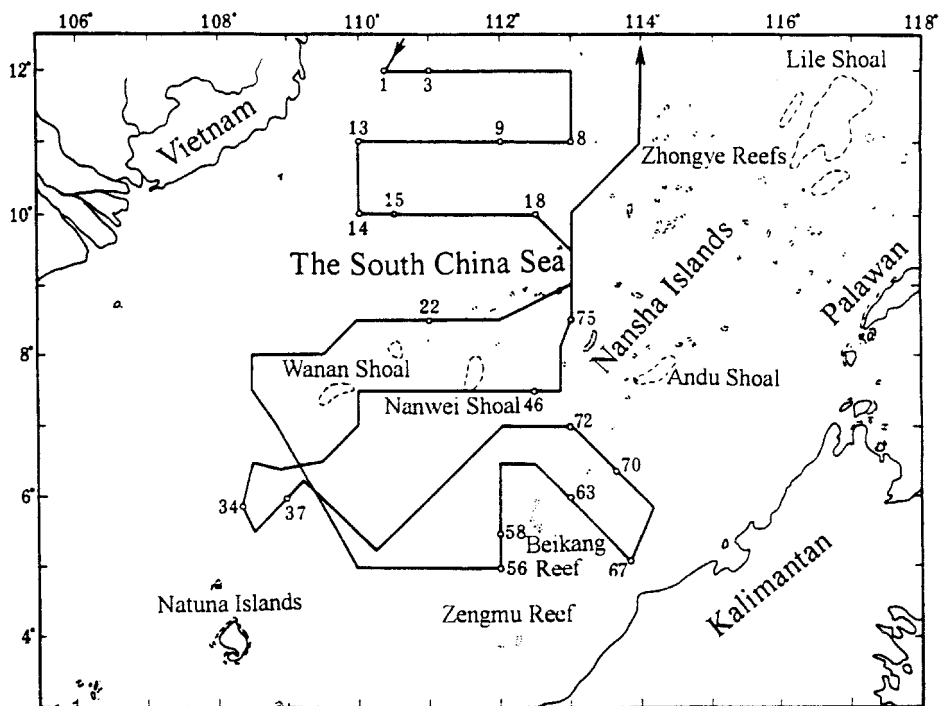


Figure 1. Map of DMS sampling locations and voyage tracks in the South China Sea during Nov.–Dec. 1993.

al. (1978) and Yang *et al.* (1996). In brief, DMS was first extracted from the seawater using carbon tetrachloride (30 ml per liter seawater), then reverse-extracted by 20 ml 5% aqueous solution of mercuric chloride as a DMS-Hg²⁺ complex (2DMS-3HgCl₂) which remained stable for more than six months at 4°C. Since each treatment required approximately 15 minutes, some seawater samples had to be stored for up to 3 h. Andreae and Barnard (1983) have shown that refrigerating samples at 4°C, whether filtered or unfiltered, tends to be the best way to maintain sample integrity at least for periods up to 48 h. We have adopted this sample-storage procedure when immediate treatment is impossible. Indeed, our experiments have shown that DMS concentrations in filtered and nonfiltered seawater are not significantly different in most cases within 12 h. All samples subjected to this treatment were stored in the dark at 4°C and were subsequently analyzed on return to our laboratory.

b. Analysis

DMS analysis was performed using the technique previously described (Yang *et al.*, 1996). Briefly, the DMS-Hg²⁺ complex was decomposed by 6 mol/L HCl, and DMS was redissolved in n-hexane and finally measured by a GC-14 A gas chromatograph equipped with sulfur specific flame photometric detector (Shimadzu Corporation, Japan). The

chromatographic column was a 0.32 mm \times 25 m \times 5 μ m OV-101 fused silica capillary column (SGE of Australia). The injector and detector temperatures were set at 150°C with the column temperature at 80°C. The optimal detection response was observed with gas flow rates for H₂, air, high purity He (carrier gas), high purity N₂ (purge gas) of 45, 45, 0.8, 20 ml/min, respectively. Under these conditions, a detection limit of 0.05 ng S was obtained. Precision for the analysis of DMS was typically within 10% (the relative standard deviation expressed as a percentage of the mean value), based on 5 replicates of DMS samples.

To check the recovery of DMS through the above handling procedures, we added known amounts of DMS to seawater samples from which natural levels of DMS had been removed by ultra-high-purity N₂ stripping and subjected them to the same manipulations. The results showed that the recovery of DMS from seawater was generally greater than 85%. The DMS values reported here have been corrected for recovery. The preparation of the calibration curves was conducted according to the same procedures as above; thus, the analytical blanks were also eliminated.

All glassware had been treated with dimethyldichlorosilane to prevent surface adsorption of DMS (Andreae and Barnard, 1983; Deprez *et al.*, 1986; Farwell and Gluck, 1980). Chlorophyll *a* data (indicating phytoplankton biomass) were collected during the same cruise. The fluorometric method of Strickland and Parsons (1972) was applied to determine the concentrations of chlorophyll *a*. A volume of 200 ml seawater sample was filtrated under vacuum through Whatman GF/F glass fiber filter. The filters were then stored in a freezer. The pigments were extracted with 25 ml 90% acetone, and then determined with a Turner Designs Fluorometer (Model 10-005 R). Nutrient and dissolved O₂ determinations were made according to routine methods (State Oceanic Administration, 1991). Seawater temperature and salinity data were obtained from the CTD.

3. Results and discussion

a. Study area

The Nansha Islands and adjacent sea area occupy a vast area of more than 8.23×10^5 km² in the South China Sea, south of 12N. The southern portion of the area consists of a flat continental shelf. The northwestern portion is a deep-sea basin of rhombic shape. Between the continental shelf and deep-sea basin is a wide continental slope, scattered with numerous coral reefs and shoals. This sea area lies in the low-latitude tropic region, with a monsoon-type marine climate. It is one of the most important passages for equatorial wind current. The water mass in the area originates mainly from the West Pacific. In order to study the effects of a variety of environmental factors on DMS distribution, we chose 19 sampling stations within the area as shown in Figure 1. The environmental and hydrographic considerations for selecting these stations are described in Table 1.

It can be seen from Table 1 that 3 stations (nos. 1, 13, and 67) are not far from the continent, 4 stations (nos. 34, 37, 56, and 58) are located at the sea area of the southern continental shelf with water depth shallower than 150 m, and the other 12 stations are

Table 1. Descriptions of DMS sampling stations.

Station no.	Location	Depth (m)	T °C ^(a)	S (‰) ^(b)	Description
1	12N, 110°23'E	2690	27.02	33.318	not far from Vietnam
3	12N, 111E	2878	27.94	33.284	open sea
8	11N, 113E	4245	28.36	33.121	open sea, not far from Zhongye Reefs
9	11N, 112E	4060	28.36	33.121	open sea
13	11N, 110E	665	27.58	33.327	not far from Vietnam
14	10N, 110E	1580	28.30	33.283	open sea
15	10N, 110.5E	2556	28.25	33.381	open sea
18	10N, 112.5E	2500	28.64	33.242	open sea
22	8.5N, 111E	1857	28.72	33.748	open sea
34	5° 50'N, 108° 25'E	88	27.44	33.434	sea area of southern continental shelf
37	6N, 109E	119	27.42	33.675	sea area of southern continental shelf
46	7.5N, 112.5E	2015	28.92	33.289	open sea, not far from Nanwei Shoal
56	5N, 112E	105	29.23	33.265	sea area of southern continental shelf
58	5.5N, 112E	144	28.69	33.281	sea area of southern continental shelf, near Beikang Reef
63	6N, 113E	1883	28.46	33.351	open sea
67	5°10'N, 113° 51'E	612	29.14	33.247	near Kalimantan
70	6° 20'N, 113° 40'E	2657	29.07	33.292	open sea
72	7N, 113E	1450	27.80	33.585	open sea
75	8.5N, 113E	1987	29.26	32.555	open sea

(a) Surface (0–1 m) seawater temperature, (b) Surface (0–1 m) seawater salinity.

situated in the open sea with water depths from 1450 to 4245 m. Surface seawater temperatures from all the monitoring stations range from 27.02 to 29.26°C, and salinities range from 32.555‰ to 33.748‰.

b. Horizontal distribution of DMS

Figure 2 shows the horizontal distribution of DMS at 0–1 m, 20 m, 50 m, 75 m, and 100 m water layers, respectively and Table 2 lists the concentration ranges and mean values of DMS at different water layers.

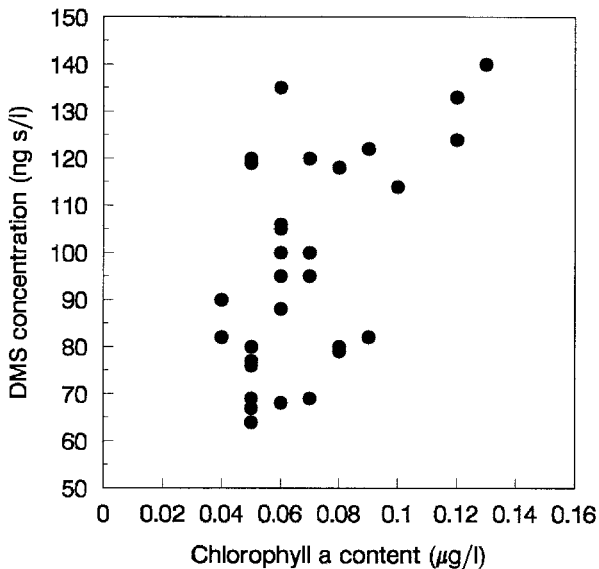
It can be seen that surface layer (0–1 m) seawater concentrations of DMS range from 64 to 140 ng S/L with a mean of 93 ng S/L for the entire investigated area. The surface seawater DMS concentrations available in the literature agree well with our data. Bates *et al.* (1987b) found that surface concentrations of DMS for the tropical North Pacific ranged from 45 to 160 ng S/L. The mean value (91 ng S/L) of DMS from surface seawater of the

Table 2. DMS concentration ranges and its mean values at different water layers.

Water layer (m)	Concentration range (ng S/L)	Mean value (ng S/L)
0-1	64-140	93
20	68-145	87
50	64-168	113
75	35-153	85
100	20-68	43
150	15-37	25
200	<15	<15

Atlantic Ocean (Barnard *et al.*, 1982) also compares well with that (93 ng S/L) from this study area.

Our study showed that the highest concentration of DMS was observed at Station 14 in the northwest part of the investigated area. According to the analytical data of the same cruise, a high concentration of chlorophyll *a* was also present at this station. Many studies have verified that DMS is formed by the microbial decomposition or enzymatic cleavage of DMSP that occurs in many species of marine phytoplankton (Belviso *et al.*, 1990; Gibson *et al.*, 1990; Iverson *et al.*, 1989; Reed *et al.*, 1983; Turner *et al.*, 1988; Wakeham and Dacey, 1989; Wakeham *et al.*, 1987). This suggests that a relationship probably exists between DMS and chlorophyll *a* concentrations. Linear regression analysis shows that a correlation indeed appears between DMS and chlorophyll *a* concentrations in the upper 20 m seawater samples ($r = 0.595$, $n = 33$), as indicated in Figure 3.

Figure 3. Plot of DMS against chlorophyll *a* for the upper 20 m seawater samples.

In the northwestern part of the investigated area, DMS contour lines appeared ringlike and values were higher than 100 ng S/L, which was consistent with the observation that this region was in proximity to Vietnam and exhibited a relatively high primary productivity. In the northeastern part of the investigated area, i.e., from 110E to the east along 10N, 11N and 12N transect, respectively, the DMS levels gradually reduced, and a minimum concentration of DMS (64 ng S/L) was found at Station 8 lying west of Zhongye Reefs. In the southwestern part of the investigated area, DMS contour lines stretched northwestward and southeastward, and values tended to increase from outside to inside (72–97 ng S/L). In the southeastern part of the investigated area, DMS contour lines presented ringlike. Levels progressively decreased outward until relatively low DMS concentration (67 ng S/L) appeared at Station 63. The surface DMS value for Station 67 was 76 ng S/L. This value was somewhat lower than what was expected for the coastal waters of Kalimantan, but it was in good agreement with low chlorophyll *a* concentration and low primary production level observed in this region.

The distribution tendency of DMS at 20 m and 50 m water layers was basically consistent with that at the 0–1 m water layer, except that the maximum concentration of DMS at the 50-m water layer occurred at Station 58 lying west of Beikang Reef. The DMS concentrations at the 75-m water layer varied from 35 to 153 ng S/L, with the maximum level found at Station 13, which was somewhat different from the 0–1-m water layer DMS distribution. Compared with the above layers, the DMS concentrations at 100-m water layer varied in a relatively narrow range of 20–68 ng S/L. This layer appeared to be a transition layer from which DMS distributed homogeneously.

c. Vertical distribution of DMS

The vertical profiles of DMS, chlorophyll *a*, and phosphate concentrations from the stations shown in Figure 1 are detailed in Figure 4. The vertical distribution of DMS in the water column is characterized by a maximum observed for depths typically between 30–75 m. The DMS maximum values at the 30-m and 75-m water layers were found at three stations and one station, respectively; whereas the DMS maximum value at the 50-m water layer was observed at 15 stations, accounting for 79% of the entire investigated stations. Below this maximum, DMS concentrations exhibited a sharp decrease with depth until the level of 1% light penetration was reached. DMS still exists at 1000 m depth (Stations 15 and 63). The presence of DMS in the deep sea probably results from the production of sedimenting particles such as fecal pellets which may contain DMSP (Turner *et al.*, 1989). A close correspondence between DMS and chlorophyll *a* is obvious from the profiles. In some cases DMS concentrations closely follow the vertical distribution of chlorophyll *a* (Stations 14, 15, 22, 58, 72 and 75). This is further evidence for the role phytoplankton play in the production of DMS. However, in most circumstances the DMS maximum is situated above the chlorophyll *a* maximum; i.e., the maximums of chlorophyll *a* and DMS are not usually present at the same depth. This phenomenon suggests that

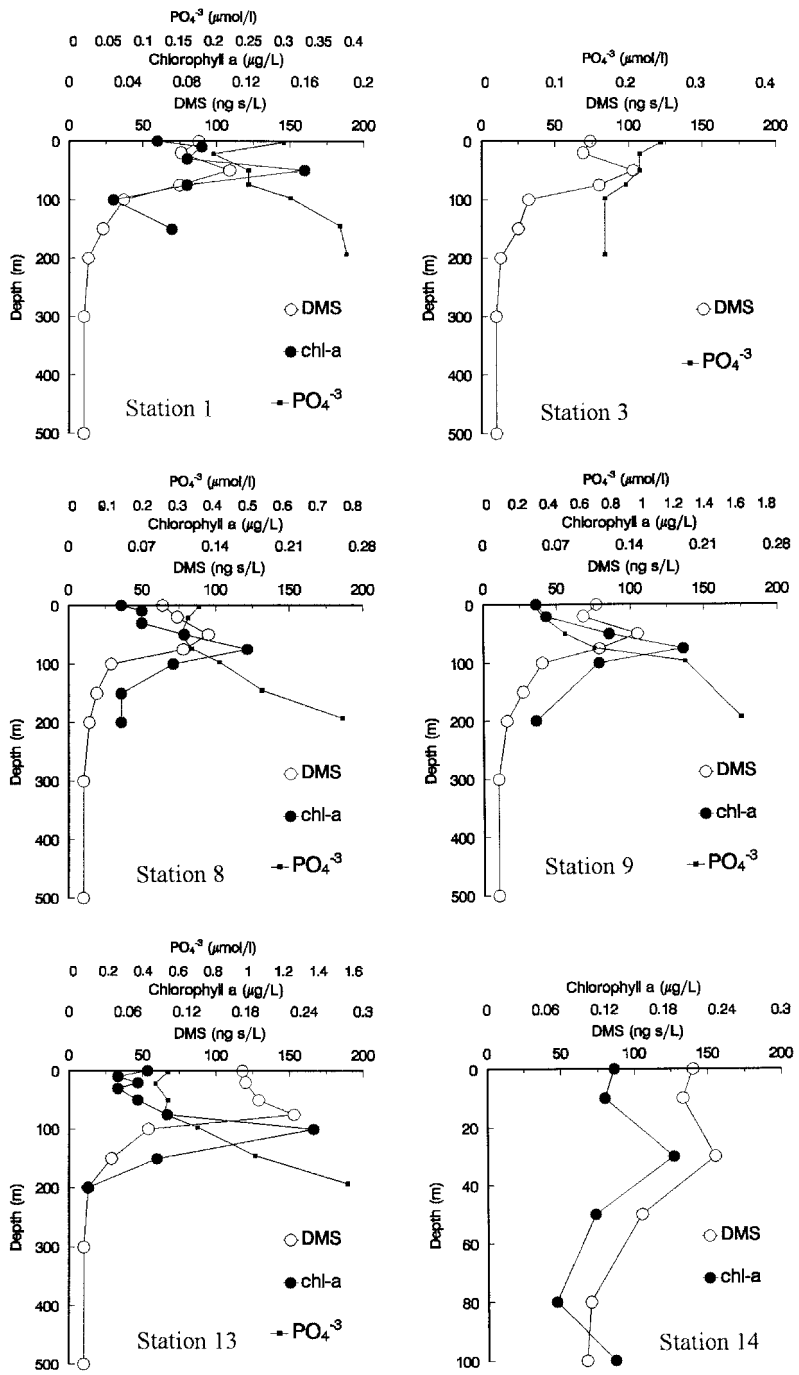


Figure 4. The vertical profiles of DMS, chlorophyll *a* and phosphate concentrations in Nansha Islands waters.

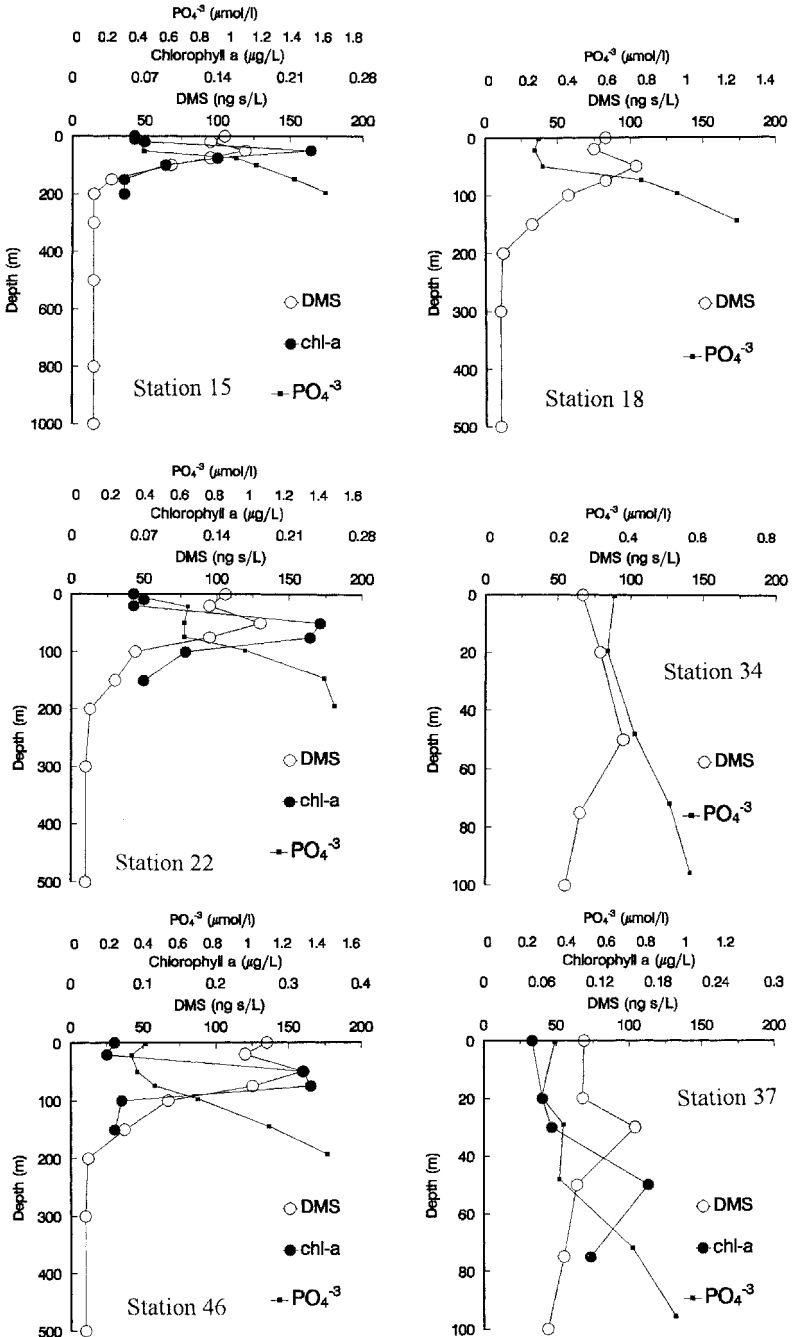


Figure 4. (Continued)

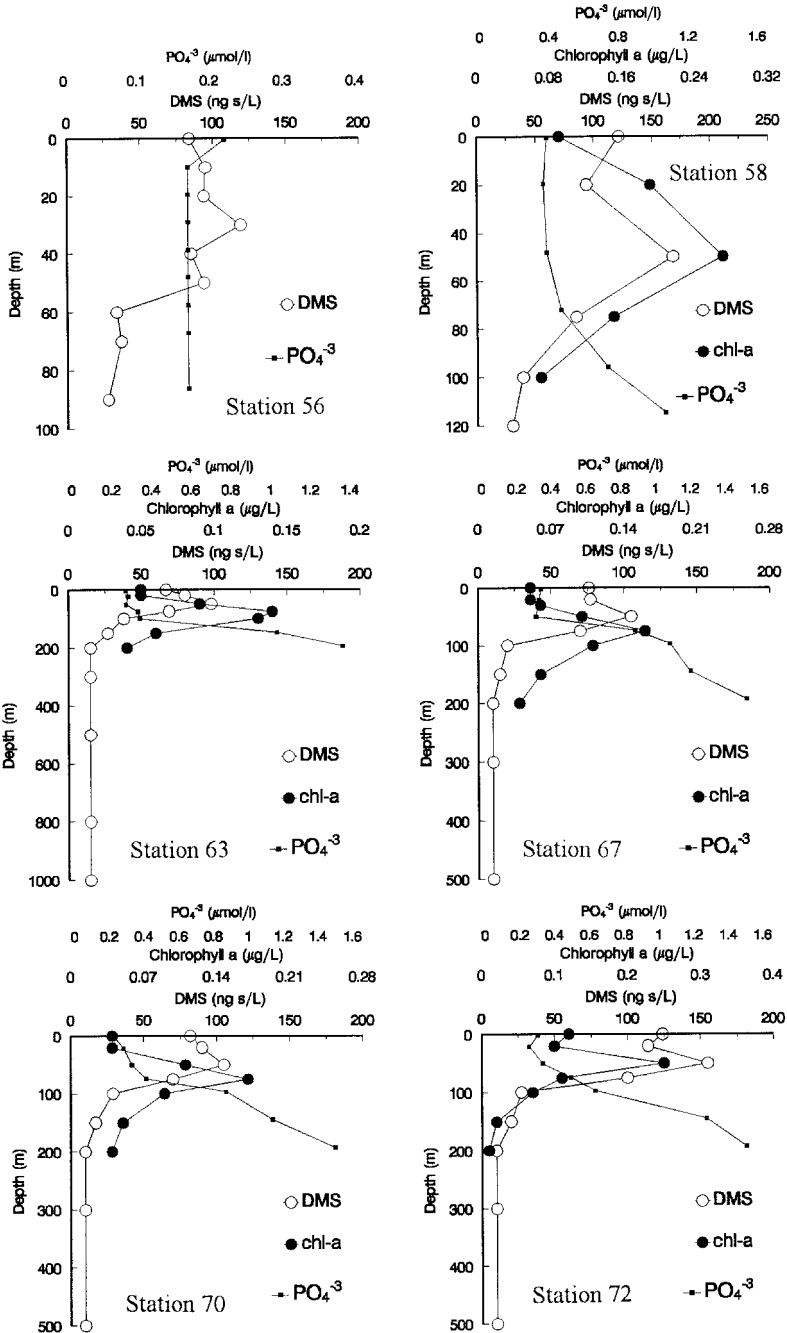


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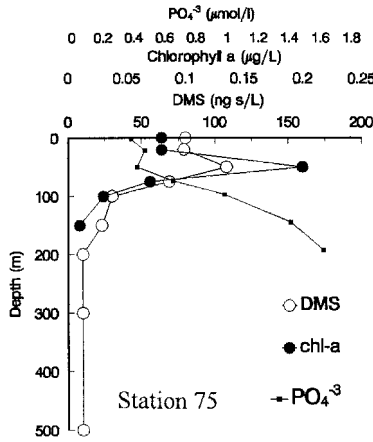


Figure 4. (Continued)

besides phytoplankton density, other factors such as phytoplankton species are also important in governing DMS levels in seawater.

As has been demonstrated, the DMS distribution in the water column is determined by several factors including light-dependent rates of photosynthesis, the vertical distributions of phytoplankton biomass and species, zooplankton grazing on phytoplankton, microbial consumption, photochemical oxidation processes (Brimblecombe and Shooter, 1986; Gabric *et al.*, 1993; Kiene, 1993; Kiene and Bates, 1990; Taylor, 1993; Wakeham *et al.*, 1987; Wolfe and Bates, 1993; Yang *et al.*, 1997) and rate of evasion to the atmosphere (Barnard *et al.*, 1982; Erickson *et al.*, 1990). Also, viruses may infect phytoplankton populations, causing the death of large numbers of phytoplankton cells and the release of DMS (Bratbak *et al.*, 1995; Malin *et al.*, 1992). Moreover, the external factors such as temperature, salinity and nutrients may affect the DMS levels produced by phytoplankton. Although the DMS distributions are complicated by these factors, the single peak shape vertical distribution of DMS in the euphotic zone of this sea area may be explained as follows. Located in the tropical region, the Nansha Islands sea area has little variability in surface seawater temperature all over the year. Therefore, vertical mixing of surface seawater resulting from a decline in temperature in this area is much smaller than that in medium and high latitude areas. So, it is difficult for surface seawater to obtain nutrients from the lower-layer seawater. Inadequacy of nutrients in the surface seawater, together with the undue sunlight radiation, makes primary productivity and chlorophyll *a* concentration here very low. Besides, due to sunlight intensity, DMS appears to be quite readily photo-oxidized in surface seawater (Brimblecombe and Shooter, 1986; Yang *et al.*, 1997). In addition, DMS easily diffuses from seawater into the atmosphere. For these reasons, the highest DMS concentration generally does not exist in the surface seawater. At the lower boundary of the thermocline (there is usually a steep temperature gradient between 25 m and 75 m) in the Nansha Islands sea area, nutrient concentrations begin to rise. Because of

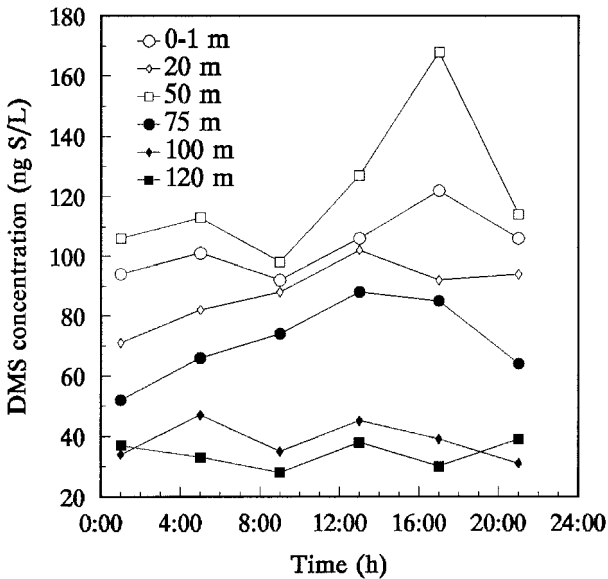


Figure 5. The diel variation of DMS concentration throughout the water column at Station 58.

being simultaneously supplied with light, heat and nutrients, phytoplankton bloom in this layer. Hence, this layer possesses relatively high primary productivity, which is consistent with appearance of the chlorophyll *a* maximum here. According to the vertical distribution of the light-beam attenuation coefficient (α) measured in the Nansha Islands sea area (Multidisciplinary Oceanographic Expedition Team of Academia Sinica to Nansha Islands, 1989), a large light-beam attenuation peak frequently existed at water depths between 40 m and 85 m. The appearance of this peak is mainly the result of plankton. Consequently, a DMS maximum should be expected in this layer. In the range of 70–200 m water depths where the nutrient concentrations increase rapidly with depth, the DMS level gradually decreases since the weak light limits the growth of phytoplankton. At 200 m depth, DMS concentration has approached its detection limit.

d. Diurnal change of DMS

We investigated the diurnal change in DMS concentration throughout the water column with time of day at Station 58 (Fig. 5). Figure 5 shows that no significant DMS diurnal variation is found at 0–1 m and 20 m water layers, in agreement with the observations of Berresheim (1987). However, a DMS diurnal variation is observed at the 50-m water layer where DMS concentration has a maximum in the late afternoon. Because no diurnal variation in biological factors was simultaneously measured, it was difficult for us to explain why this DMS maximum appeared in the late afternoon. Perhaps this DMS maximum was associated with its high production rate or low consumption rate at the same time. Further observations are required.

At the 75-m water layer, DMS levels appear to be higher during the day than during the night. This may be caused by the fact that photosynthesis in the daytime is strong and therefore, DMS release rate from phytoplankton is swift. At the 100-m and 120-m water layers, biological activity does not appear to be an important factor for influencing DMS level, as only a small diel variation of DMS is observed.

e. Environmental factors for influencing DMS distribution

Up to this point, environmental factors that may affect the distribution of DMS, such as seawater temperature, salinity, dissolved O₂ and nutrients, etc. have been briefly studied. Therefore, we examined the relationship between the concentration of DMS and these environmental factors. The results are shown in Figure 6.

Figure 6 shows that the DMS concentrations vary in response to the changing environmental factors. The regression analyses indicate that a significant correlation appears between the DMS concentration and seawater temperature for all water samples during the cruise (Fig. 6a, $r = 0.744$, $n = 122$). However, using data collected from all the water samples (Fig. 6b) we found no correlation of DMS concentration with seawater salinity.

It may be observed from the dissolved O₂ data during the same cruise that an O₂ maximum usually exists at depths between 20–50 m. This depth of the O₂ maximum just lies near the lower boundary of the thermocline, and above the maximums of chlorophyll *a* and of the light-beam attenuation coefficient. The study has shown that the dissolved O₂ maximum in the water column of Nansha Islands sea area is principally caused by the photosynthesis of organisms (Multidisciplinary Oceanographic Expedition Team of Academia Sinica to Nansha Islands, 1989). That means that the water layer with the O₂ maximum corresponds to the photosynthetic zone that is generally called an active layer of the living things in this sea area. This would be consistent with the presence of significant DMS production and, therefore, of the DMS maximum at this layer. It seems to be that DMS content is associated with the dissolved O₂ level in the euphotic zone. In fact, a significant correlation exists between the DMS concentration and the dissolved O₂ level for all samples (Fig. 6c, $r = 0.766$, $n = 122$).

From our investigation of cruise data, it is apparent that the nutrients are almost depleted in the surface mixed layer; i.e., the upper quasi-homogenous layer where the thickness usually ranges from 30–50 m. Owing to the separation of the pycnocline, lower-layer seawater with rich nutrients is difficult to supplement to the upper quasi-homogenous layer. Therefore, DMS shows an inverse relationship to nutrients (Fig. 6d, $r = -0.721$, $n = 115$; Fig. 6e, $r = -0.735$, $n = 115$; Fig. 6f, $r = -0.781$, $n = 113$). This relationship is expected due to the fact algae take up nutrients and release DMS. Andreae and Barnard (1984) also found the inverse distribution trend between DMS and nutrients in the water column.

So far, much effort has been directed toward finding the relationships between DMS and other observable parameters, especially chlorophyll *a* because its level in surface seawater may be estimated by remote sensing and could be used to predict DMS concentrations in

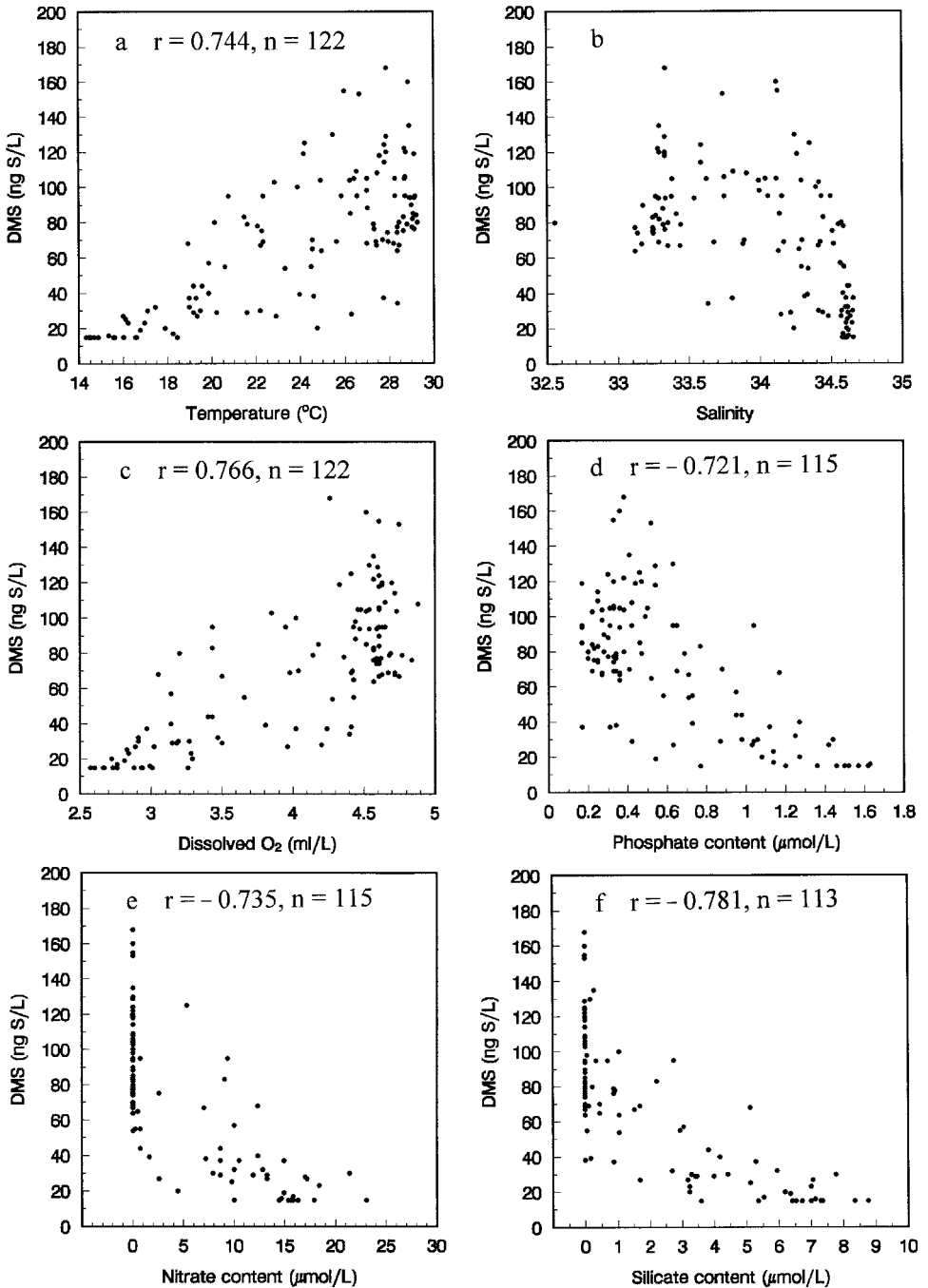


Figure 6. Relationship between the concentration of DMS and temperature (a), salinity (b), dissolved oxygen (c), phosphate (d), nitrate (e) and silicate content (f) from all the seawater samples.

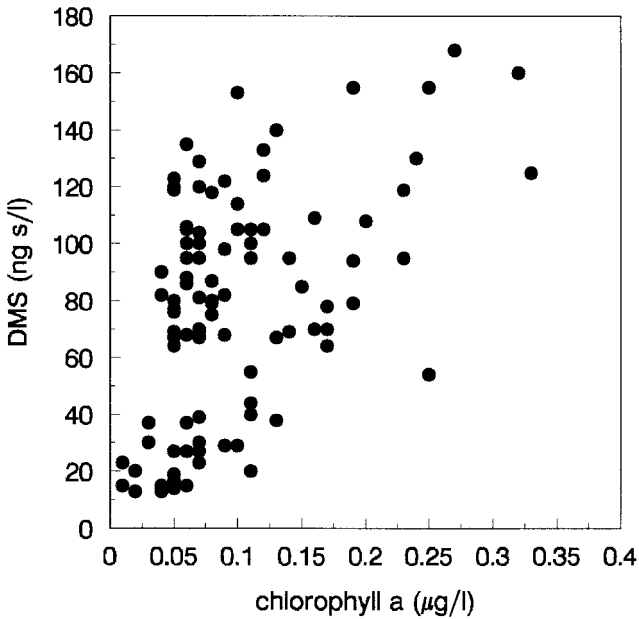


Figure 7. Diagram of the concentration of DMS vs. chlorophyll *a* in all the seawater samples.

some sea areas where no direct measurement of its concentration is available (Thompson *et al.*, 1990). Our attempts at finding consistent relationships between DMS and chlorophyll *a* have resulted in preliminary success. On the one hand, DMS concentration is closely correlated to chlorophyll *a* level in the upper 20 m seawater (Fig. 3). On the other hand, when subjecting our entire DMS and chlorophyll *a* data in vertical profiles to linear regression analysis, we find the existence of a correlation between them ($r = 0.503$, $n = 102$), as shown in Figure 7.

The Nansha Islands sea area is a low-productivity tropical region. However, the DMS concentrations here are obviously higher than expected based on the phytoplankton biomass in the area. This most probably results from species-related effects because only certain groups of algae are known to produce significant amounts of DMS (Keller *et al.*, 1989). The field investigation showed that the Nansha Island waters contain abundant dinoflagellates which include twenty-five individual phytoplankton clones, such as *Dissoadinium lunula* and *Procoentrum* sp. (Multidisciplinary Oceanographic Expedition Team of Academia Sinica to Nansha Islands, 1989). Most of these clones are the major emitters of DMS (Keller *et al.*, 1989). In fact, a single phytoplankton species can be responsible for most of the DMS production in certain cases; e.g., *Phaeocystis pouchetii* in the northwest European continental shelf region (Holligan *et al.*, 1987). A high level of DMS was also found in the oligotrophic waters of the Sargasso Sea, as the Coccolithophorid species common in this sea are prolific producers of DMS (Andreae, 1990). Nevertheless, the vertical distribution of phytoplankton in the Nansha waters indicates that diatoms, which

are, in general, not major producers of DMS (Keller *et al.*, 1989), become dominant algae at the 75-m water layer (Multidisciplinary Oceanographic Expedition Team of Academia Sinica to Nansha Islands, 1989). This is probably the reason why the highest level of DMS is not generally expected here; on the contrary, the chlorophyll *a* maximum usually appears at this depth.

Although the DMS concentrations are well related to the chlorophyll *a* levels, the apparent "scatter" in the DMS and chlorophyll *a* data is still observed in Figure 7. This scatter may be due to the fact that in some cases, high levels of chlorophyll *a* do not correspondingly result in high concentrations of DMS, while oligotrophic low-productivity sea areas probably contain high levels of DMS. As has been observed, silicate is almost completely depleted in the upper quasi-homogenous layer, so that nonsiliceous phytoplankton can effectively compete with diatoms. This would be favorable to the production of DMS. In addition, the lack of nitrate in the upper quasi-homogenous layer may also cause the relatively high abundance of DMS in this sea area. Turner *et al.* (1988) found that planktonic algae grown under nitrate-limited environments exhibited higher intracellular DMSP levels than those grown at high nitrate concentrations. This is attributed to the fact that in the nitrate-depleted sea area, the use of a sulfur osmolyte (DMSP) instead of a nitrogen osmolyte would be necessary for the phytoplankton cells to achieve the required high internal osmotic pressure to balance that of the seawater around them (Andreae, 1990).

Since phytoplankton distributions in this sea area show complex patterns (Multidisciplinary Oceanographic Expedition Team of Academia Sinica to Nansha Islands, 1989) and meanwhile the DMS production rate between different phytoplankton species varies significantly, it is still difficult for us to find a definite relationship between DMS concentration and chlorophyll *a* level. A consideration of phytoplankton species and their respective contribution to the levels of DMS and chlorophyll *a* must be included.

f. The sea-to-air flux of DMS

In order to assess the role of DMS from the oceans as a source of atmospheric sulfur, regional and global scale data on the DMS sea-to-air flux are needed. The flux of DMS across the air-sea interface is usually estimated from the gas exchange model (Liss, 1973), which assumes the sea-to-air flux is proportional to the product of the concentration gradient across the air-sea interface and the transfer velocity (K) across the interface, i.e.

$$F = K(C_l - C_g/H) \quad (1)$$

where C_g is the atmospheric partial pressure, H is the Henry's law constant of the gas, and C_l is the observed DMS concentration in the surface seawater. Since $C_g/H \gg C_l$ is always held, hence the flux of DMS into the air is simplified into the following equation:

$$F = KC_l.$$

Table 3. The calculated Sc and K_{DMS} values and sea-to-air fluxes of DMS from the South China Sea.

Station no.	u_{10} (m/s)	Sc	K_{DMS} (m/d)	C_{DMS} (ng S/L)	F_{DMS} ($\mu\text{mol m}^{-2} \text{d}^{-1}$)
1	10.6	670	4.64	88	12.8
3	10.2	643	4.48	74	10.4
8	10.3	632	4.59	64	9.2
9	10.1	632	4.45	77	10.7
13	10.8	653	4.83	118	17.8
14	10.5	633	4.71	140	20.6
15	8.7	635	3.52	105	11.6
18	3.3	624	0.13	83	0.34
22	7.1	622	2.48	106	8.2
34	6.5	657	2.02	67	4.2
37	4.0	658	0.40	69	0.86
46	6.1	617	1.82	135	7.7
56	8.0	608	3.12	84	8.2
58	6.0	623	1.75	122	6.7
63	2.3	629	0.09	67	0.19
67	3.8	611	0.28	76	0.67
70	4.6	612	0.82	82	2.1
72	7.2	647	2.50	124	9.7
75	4.4	607	0.69	80	1.7
Average	7.1	632	2.49	93	7.6

DMS concentration in surface seawater is relatively easy to measure, whereas the calculation of a reliable K value is usually difficult because this parameter is dependent on a variety of factors, including wind speed (u), Schmidt number (Sc), and surface films (Frew, 1997; Liss and Merlivat, 1986). The DMS Schmidt number at the local temperature is calculated from the equation of Saltzman *et al.* (1993). From the Sc of DMS and the wind speed at the height of 10 m (u_{10}), we calculated K value of DMS (K_{DMS}) from the equation of Liss and Merlivat (1986), by assuming that K_{DMS} is proportional to $Sc^{-2/3}$ for $0 < u < 3.6$ m/s and to $Sc^{-1/2}$ for $3.6 < u < 13$ m/s. Then the sea-to-air flux of DMS (F_{DMS}) is calculated from the measured DMS concentration in the surface seawater and the transfer velocity (K_{DMS}), together with the Sc and K_{DMS} values are listed in Table 3.

From Table 3 it can be seen that the K_{DMS} values range from 0.09 to 4.83 m/d, with an average of 2.49 m/d. This average is comparable with the result ($K_{DMS} = 2.75$ m/d) calculated from the tropical north Pacific Ocean in winter by Bates *et al.* (1987b). On the basis of the DMS concentration in surface seawater given in Table 3, the mean flux value of DMS calculated in this study is equal to $7.6 \mu\text{mol m}^{-2} \text{d}^{-1}$. This flux value is lower than the sea-to-air DMS flux of $10.8 \mu\text{mol m}^{-2} \text{d}^{-1}$ calculated in the coastal region (0° – 20° N) over the North Pacific in winter (Erickson *et al.*, 1990). Also, it is reasonably lower than the DMS flux value of $10.6 \mu\text{mol m}^{-2} \text{d}^{-1}$ obtained from the East China Sea by Yang *et al.* (1996) as that sea area exhibits higher primary productivity and DMS levels than the tropical South China Sea. The South China Sea has an area of $3.5 \times 10^6 \text{ km}^2$. Using the

DMS flux value calculated above, the total annual emission of DMS from this sea is thus 9.7×10^9 mol/a. This annual emission estimate has a factor of 2 uncertainty associated mainly with approximate estimation of the DMS transfer velocity (Andreae, 1990), while the uncertainty from the DMS concentration item is probably small because a very small seasonal or interannual variability in DMS concentration has been observed in the tropical North Pacific Ocean (Bates *et al.*, 1987b).

4. Conclusion

- (1) The DMS concentrations in the surface layer (0–1 m) seawater of this sea area vary from 64 to 140 ng S/L with the highest values found at Station 14 in the northwestern part of the investigated area. This is consistent with the horizontal distribution of chlorophyll *a* and the primary productivity. The DMS concentrations are correlated with chlorophyll *a* levels in the upper 20 m seawater.
- (2) The vertical profiles of DMS show a single peak shape with maximum concentrations occurring at depths between 30–75 m. The vertical distribution of DMS is closely associated with the distribution of chlorophyll *a* in the water column. A correlation is found between the entire DMS and chlorophyll *a* data in vertical profiles.
- (3) The DMS concentrations are related to environmental factors. There appear to be significant linear correlations between the DMS concentrations and seawater temperature and the dissolved O₂ content for all the samples. Whereas an inverse linear correlation is found between the DMS concentration and the content of nutrients (including phosphate, nitrate and silicate) from all the samples. However, we found no correlation of DMS concentration with seawater salinity.
- (4) The sea-to-air flux of DMS is calculated to be $7.6 \mu\text{mol m}^{-2} \text{d}^{-1}$, with an annual emission of 9.7×10^9 mol from the South China Sea.

Acknowledgments. We would like to thank Dr. Wang Hankui who provided us with the nutrients and dissolved oxygen data, and Prof. Huang Liangmin who provided the chlorophyll *a* data. The help of the crew of the R/V “*Experiment No. 3*” is gratefully acknowledged. This research was supported in part by grants from the National “Eighth Five Year Plan” Special Project (85-927-04-03).

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