

Variability in abundance and fluxes of Dimethyl sulphide in the Indian Ocean

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Abstract

Dimethyl sulphide (DMS) is a biogenic gas of climatic significance on which limited information is available from the Indian Ocean. To fill in this gap, we collected data on DMS and DMSP_t (total dimethylsulphoniopropionate) by participating in a dozen cruises. Here, we discuss on the variability in DMS and DMSP_t in the north and central Indian Ocean regions in terms of their spatial and temporal variation. DMS and DMSP_t exhibited significant spatial and temporal variability. Apart from the concentration gradients in DMS within the Arabian Sea, Bay of Bengal and Central Indian Ocean basins, differences in average abundances were conspicuous between these basins. The Arabian Sea contained more DMS (mixed layer (MLD) averaged was 7.8 nM) followed by the Bay of Bengal (2.8 nM) and the Central Indian Ocean (2.7 nM). The highest concentrations of DMS and DMSP_t (525 nM and 916 nM, respectively) were found in upwelling regimes along the west coast of India during the Southwest monsoon and fall inter-monsoon seasons. Average surface DMS was the highest in the Arabian Sea. On the other hand observed sea-to air fluxes of DMS were higher in the Bay of Bengal due to the prevalence of turbulent conditions. In the Arabian Sea wind speeds were low and hence the sea-to-air fluxes. The total diffusive flux of DMS from the study area to atmosphere is estimated to be about 1.02×10^{12} g S y⁻¹, which contributes to 4.1 - 6.3 % of the global DMS emission.

Keywords

DMS, DMSP_t, Indian Ocean, fluxes, monsoon.

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Introduction

Dimethyl sulphide is the most dominant among the reduced sulphur gas found in surface layers of the ocean (Lovelock et al., 1972). The emission of dimethyl sulphide from seawater is expected to balance the excess sulphur deposition over the remote oceans (Charlson et al., 1992). Charlson et al. (1987) proposed a hypothesis, known as CLAW hypothesis connecting biogenic DMS emissions to changes in albedo, in which increased production of DMS due to global warming is expected to lead to more sulphate aerosols and subsequently to more cloud condensation nuclei (CCN) that in turn enhances back radiation.

In seawater, DMS is produced from dimethylsulphoniopropionate (DMSP) which in turn is produced by phytoplankton. Specific species of phytoplankton are found to be responsible for most of the DMSP production in seawater (Barnard et al., 1984; Holligan et al., 1987; Belviso et al., 1990; Malin et al., 1993). According to Liss et al. (1993) the potential for production of DMS in various taxonomic groups follows the order:

Coccolithophores > Phaeocystis > Dinoflagellates > Diatoms

DMSP in phytoplankton was initially proposed to function as an osmolyte (Dickson et al., 1980; Vairavamurty et al., 1985). Over the years we now have evidence for its role in other processes such as for cryopreservation (Kirst et al., 1991), as a deterrent against grazing (Dacey and Wakeham, 1986; Wolfe and Steinke, 1996) and against bacteria (Wolfe et al., 1997). In the recent past the anti-oxidant role of DMSP is becoming more popular (Sunda et al., 2002). Factors controlling DMSP production, its conversion to DMS, fate of DMSP and DMS sulphur in the marine environment are well documented in the review by Stefels et al., (2000) and in the present issue.

Extensive measurements on DMS and DMSP have been done in different parts of the oceans (Kettle et al., 1999 and references therein) including estuarine and lagoon waters (Iverson et al., 1989; Moret et al., 2000). A few studies have been made in the Indian Ocean, which include Hatton et al. (1999), Shenoy et al. (2000, 2002), Kumar et al. (2002) and Shenoy and Patil (2003) and some unpublished data in eastern Indian Ocean (of T.S. Bates as mentioned by Kettle et al., 1999) and for Amsterdam station (Nguyen et al., 1990, 1992).

Hatton et al. (1999) found elevated concentrations of DMS, DMSP and DMSO in the eutrophic zone (Arabian Sea) immediately after the SW monsoon. In addition DMSO concentrations were found to correlate with near surface DMS and DMSP. Shenoy et al. (2000) reported the first ever measurements of DMS from the Bay of Bengal where salinity appeared to

play an important role in the DMSP production. Shenoy et al. (2002) documented five times higher concentrations of DMS and DMSP in winter season of 1999 than that in 1998, which has been attributed to differences in physical forcing and associated biological processes. In a time series experiment carried out in estuarine waters of Goa, west coast of India, Shenoy and Patil (2003) recorded elevated concentrations of DMS and DMSP during the monsoon season (maximum DMS 15.4 nM and DMSP_t 419.5 nM) due to the possible prevalence of mixed population of diatoms and dinoflagellates. Covariations among oceanic and atmospheric DMS and atmospheric SO_2 , wet deposition of methane sulphonic acid (MSA), NSS SO_4^{-4} , and rain acidity have been shown by Nguyen et al. (1990, 1992) near the Amsterdam island (southern Indian Ocean).

While the biogeochemistry of carbon dioxide and nitrogen species are reasonably well known, particularly in the northern Indian Ocean, our knowledge on sulphur species is limited. DMS and associated sulphur compounds are climatically important, particularly in monsoon dominated tropical Northern Indian Ocean, in regard to air-sea interactions and feedbacks. Significant diversity in physical and associated biological regimes in the Indian Ocean together with dynamic climate offers a potentially interesting region to understand the DMS cycling and its controlling factors. Therefore, this study has been undertaken to study spatial (especially inshore-offshore gradients off the coast of India and surface variation) and temporal variations of DMS and DMSP_t in the Indian Ocean and to evaluate its atmospheric emissions in regional and global context.

Material and Methods

Sampling for this study was attempted keeping in view the large spatial and temporal variability of the biogeochemical processes in the Indian Ocean. Data were collected for temperature, salinity, dissolved oxygen, nitrate, chlorophyll *a*, DMS and DMSP_t in seawater. Besides, data were collected for wind speeds and atmospheric temperature. Figure 1a depicts the study area in the Indian Ocean, where a dozen oceanic expeditions were undertaken on boards ORV Sagar Kanya (SK) and FORV Sagar Sampada (SS) covering a total of 320 stations. Data were mainly collected from the upper 200 m of the water column as the initial study revealed DMS and DMSP_t to be at or below detectable levels at depths greater than 200 m. Surface waters were sampled at many locations besides regular stations. Time series approaches were followed in different ways at various locations: i) diurnal variability experiments lasting 12 to 40 hours at two locations (by maintaining constant positions at 7° and 10°N along 88°E in the Bay of Bengal) during SK 138C, (ii) longer time experiment (nearly a month) at

17.5°N and 89°E during SK147A&B (Bay of Bengal)- this experiment was carried out in two phases wherein weather and oceanographic conditions were remarkably different between the two. (iii) seasonal variability experiment involved in periodic collection of DMS data along a section off Candolim, Goa (Figure 1b), particularly during and after the southwest monsoon, and (iv) two cruises (SK133-first field phase (FFP) and SK141-intensive field phase (IFP)) to find the inter-annual variability of DMS in winter in the Central Indian Ocean. The first two time series experiments were done as part of the Bay of Bengal Monsoon Experiment (BOBMEX), the third as a part of the Land Ocean Interaction in the Coastal Zone (LOICZ) and the last as a part of the Indian Ocean Experiment (INDOEX).

Hydrographic and biological properties

Seawater samples were collected using a rosette attached to the Conductivity-Temperature-Depth (CTD) system, fitted with 12 Niskin bottles. Sub-sampling of water was made in the order: dissolved oxygen (DO), DMS, DMSP_t, nutrients, salinity and chlorophyll *a*. Care was taken while sampling dissolved gases to avoid trapping of bubbles. Temperature and conductivity were measured using the sensors fitted to the Sea-bird CTD system. Temperature sensed by the probe was periodically checked against the values obtained from reversing thermometers. Conductivity measurements of CTD, and hence salinity were calibrated against data obtained using an Autosal salinometer (model 8400). Dissolved oxygen was measured by the classical Winkler titration method. Nutrients (nitrate) were measured using a Skalar autoanalyser (SA 4000).

For chlorophyll *a* analysis, a known volume of seawater sample (0.5 to 1 litre for coastal and estuarine samples or 2 to 3 litres for open ocean waters) was filtered through Whatman GF/F filters under low vacuum. Chlorophyll pigments on the filter paper were then extracted into 10 ml of 90% acetone under dark and refrigerated conditions for 24 hours. Fluorescence was measured using a Hitachi spectrofluorometer (UNESCO, 1994).

DMS and DMSP measurements

Water samples for DMS and DMSP_t analyses were collected in two separate 60 ml dark ground glass bottles. Water samples were not filtered because even with extreme care DMS losses cannot be prevented during the filtrations and this process can alter the contents of DMSP_t. Thus the concentrations reported here refer to total dissolved DMS and DMSP. Following the collection DMS samples were immediately kept in the dark at 4°C. DMS was measured using a Hewlett Packard 5890 Series II Plus Gas Chromatograph fitted with a flame

photometric detector (FPD). The details of the analysis are described in Shenoy et al 2002. The detection limit for DMS measurements, found as twice the standard deviation from regression analysis for values <0.1 nmol, was 0.012 nmol. Precision in DMS measurements was found to be 6% for standards. Reproducibility of DMS and DMSP_t in seawater samples was periodically checked in all the cruises for paucity of time and availability of a dedicated gas chromatograph.

Immediate repeat analysis of a sample showed no detectable DMS indicating its negligible production from DMSP_t, if any, during the stripping process. DMSP_t was measured after hydrolysing the unfiltered water sample for six hours using 10 M NaOH (1 ml), which was added immediately after sample collection (Turner et al., 1990; Shenoy et al., 2002). Alkali hydrolysis resulted in the cleavage of DMSP_t into DMS and acrylic acid. DMSP_t concentrations were read from the DMS calibration curve. Tests revealed about 95% conversion of DMSP_t to DMS during the alkali hydrolysis for 6 hours. Precision of seawater DMS analysis is found to be 8-10%.

Wind Speed

Wind speed data used in DMS flux calculations were collected using automatic weather station (AWS) on board the research vessels or on top of the institute building. Wind speed was measured using Young's wind monitor (model 05103). The wind speeds were normalized for 10 m height and corrected for the direction.

DMS Flux

DMS fluxes were calculated using the formulations proposed by Turner et al. (1996) and the correction factors given by Saltzman et al. (1993). The flux of DMS gas between sea and air is proportional to the concentration gradient across the air sea interface and is calculated according to the following equation.

$$F_{\text{DMS}} = k \cdot \Delta C$$

Where F_{DMS} = net flux of DMS

k = transfer (or piston) velocity and

ΔC = concentration gradient across the air-sea interface.

$$\Delta C = C_w - (C_a \cdot h^{-1})$$

where C_w = concentration of DMS in seawater

C_a = concentration of DMS in air

h = Henry's law constant, expressed as the ratio of air to water concentrations at equilibrium.

As the concentration of DMS in air is very low (nearly three orders of magnitude less than that in water) C_a is generally considered to be zero and thus C_w equals ΔC .

Mixed layer depth (MLD)

Mixed layer depths (MLD) were calculated based on density criteria where MLD was defined as the depth at which density increased by 0.125 kg dm^3 with reference to that at the sea surface. ML-DMS is the DMS averaged in the water column above the MLD.

Results

1. Hydrography in brief

The southwest and northeast monsoons of the Indian Ocean region play a major role in controlling the biogeochemical processes. Between November and February, the Inter-tropical convergence zone (ITCZ) lies in the southern hemisphere ($\sim 10^\circ\text{S}$) and the region experiences the northeast monsoon (NEM). During this period cold dry winds from the north (north easterlies) blow over the northern Indian Ocean. These cool dry winds facilitate enhanced evaporation in the northern Arabian Sea and thus set in winter convection (Figure 2). The occurrence of convection pumps in nutrients into the surface layers. During the present study the average nitrate concentration in surface waters of the Arabian Sea where convection occurred was found to be $\sim 4.9 \mu\text{M}$. Such high nitrate levels promote primary production. During NEM the surface circulation in the Arabian Sea is quite similar to the circulation in the North Pacific and Atlantic Oceans.

The extensive heating of the Indo-Gangetic plain and Himalayan terrain from March to May (Spring Inter-monsoon) leads to the development of low pressure which makes the ITCZ move northward. This transition changes the direction of the winds, which now move in the south westerly direction during the southwest monsoon (June to September, SWM). The surface circulation in the Arabian Sea in SWM is totally opposite to that of the NEM. The circulation along the coasts of Oman and Somali is towards the pole and away from the coast. This causes intense upwelling in these regions. Similarly upwelling also occurs along the south west coast of India, where the flow of water is towards the equator.

One of the major characteristics of the northern Indian Ocean is the differential riverine discharges in to the Arabian Sea and the Bay of Bengal. The Bay of Bengal receives $1.6 \times 10^{12} \text{ m}^3/\text{y}$, which is five times more than the run off received by the Arabian Sea ($0.3 \times 10^{12} \text{ m}^3/\text{y}$). This reduces the surface salinity in the Bay and the lowest salinities are mostly confined to areas of river mouths. The salinity increases from north to south in the Bay. The low salinity lens

results in a strong surface stratification and prevents surfacing of subsurface nutrients through vertical mixing.

The month of October is referred to as the fall inter-monsoon (FIM) period. During the FIM the temperature contours show a stratified structure unlike in the SWM. Low wind speeds during this period lead to shallow MLD's. There are no detectable nutrients in the surface layers. One of the prominent features of the fall-intermonsoon is the occurrence of high nitrate levels near the base of MLD, which occurs within the euphotic zone. This leads to basin wide subsurface chlorophyll maximum (SCM, Bhattathiri et al., 1996) as shown in Figure 3. All these make the Northern Indian Ocean one of the most productive basins in comparison to the rest of the world in general (Behrenfeld and Falkowski, 1997).

Central Indian Ocean characteristics have been discussed in detail in Shenoy et al. (2002). In order to get an overview on the variation of DMS and DMSP in the Indian Ocean, their variability is discussed under the following sections.

2. Vertical distributions

Vertical distributions of DMS and DMSP_t in the coastal and open ocean waters of the Arabian Sea (AS), Bay of Bengal (BoB) and in the Central Indian Ocean (CIO) are shown in Figure 4. Both DMS and DMSP_t showed very distinct trends in their vertical distributions. Concentrations of DMS and DMSP_t were very low (less than or around the detection limits) in waters deeper than 200 m in the Indian Ocean. Therefore, DMS sampling was restricted to the top 200 m of water column in most cruises. In coastal waters of the Arabian Sea (76°E, 10.19°N) maximal concentrations of DMS and DMSP_t were found at shallow depths. In the present study the highest values of 525 nM of DMS and 916 nM of DMSP_t were found, respectively, at a station off Candolim and at a location off Mangalore (10.19°N, 76°E, SK148). In the open Arabian Sea (65°E, 18°N), a DMSP_t maximum of 15.5 nM occurred at 10 m whereas the peak DMS of 1.7 nM was found at the sea surface. Therefore, depths of occurrence of maximal levels of DMS and DMSP_t did not necessarily coincide at all stations but the higher abundances mostly occurred closer to the surface. The DMS and DMSP_t concentrations rapidly decreased to undetectable levels below 120m. Similar behaviors were observed in their vertical distributions in the coastal and open ocean waters of the Bay of Bengal. In the Central Indian Ocean, however, peaks in DMS (9.7 nM of DMS at 41 m) and DMSP_t (DMSP_t maximum of 31.3 nM) were found around a depth of 40 m (Figure 4). Further, DMS and DMSP_t levels were detectable up to 150 – 175 m. This occurrence of DMS at deeper

depths is unique to the central regions of the Indian Ocean (because of deep mixed layers) and contrasts with that in the Arabian Sea and the Bay of Bengal.

Table 1 lists DMS and DMSP_t variations as averages in the surface mixed layer. In the coastal (southwest coast of India) Arabian Sea the highest values occurred during the SWM and the least were during the intermonsoons. In the Bay of Bengal the coastal concentrations of DMS and DMSP_t are almost same as the open ocean value. The DMS and DMSP_t concentrations in the Central Indian Ocean are nearly similar to those in the Bay of Bengal. On the whole, the Arabian Sea (coastal + open ocean) surface mixed layer contained more than double DMS and DMSP_t (7.8 nM and 43.6 nM, respectively) than the Bay of Bengal (2.8 nM and 12.6 nM, respectively) and Central Indian ocean (2.7 nM and 10.1 nM, respectively) and followed the same trend as that of the primary production (Behrenfeld and Falkowski, 1997).

3. Spatial variations

Figure 5 depicts the variation of DMS in the Arabian Sea, along a section near 15°N (off Goa), during January, July, October and December of 1998. Very clear coastal to open ocean trends were observed. In January (winter), the DMS concentrations varied between 6 nM and 12 nM within 50 m of the water column near the coast but between 200 and 400 km away from the shore the concentrations ranged between 2 and 4 nM before increasing to 10 nM further offshore. During the summer (July 1998), DMS values were higher near the bottom of coastal waters (29 nM) that decreased towards the open ocean. Features similar to those in summer were seen in October 1998 when DMS concentration levels near the coast varied between 15 and 40 nM while in offshore waters these were low. In December 1998 coastal DMS concentrations were around 30 nM but decreased offshore. Thus in the eastern Arabian Sea high DMS concentrations were found closer to the coast.

Zonal variations of DMS in the Bay of Bengal are shown in Figure 6. Near Paradip (86.87°E, 19.98°N), DMS concentrations varied between 0.5 and 3.3 nM in coastal waters but showed little change with increasing distance from the coast. Similar features were noticed off Chennai as well. Here, DMS varied from 0.5 to 3.2 nM. Thus the zonal variability trends of DMS were contrasting between the Arabian Sea (with clear inshore-offshore gradients) and the Bay of Bengal (with no significant variation).

In the Central Indian Ocean a different scenario was found. Figure 7 depicts the variation of DMS in the Central Indian Ocean observed during the INDOEX cruises of 1998 and 1999. High DMS concentrations (0.1 to 13.9 nM) were observed between 5°S and 15°S in 1999.

These were observed as pockets between surface and 75 m with detectable concentrations down to 100 to 150 m. While away from these latitudes viz. between 0° and 5°S, and 15°S and 20°S DMS concentrations, in the same depth range, as mentioned above varied between 1 and 5 nM. DMS levels were lower in 1998 with strong latitudinal gradients.

Thus DMS in the Indian Ocean exhibits very high spatial variability in which higher concentrations were observed near to the coast and lower concentrations in the open Ocean. Further, DMS concentrations (MLD average) showed a gradual increase from the Central Indian Ocean (2.7 nM) and the Bay of Bengal (2.8 nM) to the Arabian Sea (7.8 nM).

4. Temporal variations

4.1 Diurnal variation in the Bay of Bengal

At the beginning of the time series (3.30 AM) DMS abundances were lower but with high $DMSP_t$ concentrations at 7°N and 87°E station. Towards late afternoon the trend reversed as higher DMS and lower $DMSP_t$ levels occurred (Figure 8). However, DMS exhibited lower concentrations on the following day while $DMSP_t$ was low during the intervening night (14-26 hrs). Higher $DMSP_t$ concentrations were again observed towards the end (~7.30 PM on the following day) of the experiment. These features reveal that $DMSP_t$ did not exhibit consistent day-night variations. The secondary DMS maximum of ~3.5 nM (at 50 m) coincided with the chlorophyll *a* maximum. The DMS concentrations during the time series observations varied from 0.2 to 5.1 nM with an average of 1.8 nM in the upper 100 m, whereas $DMSP_t$ ranged from 1.4 to 17.6 nM with a mean value of 7.2 nM. DMS showed maximal concentrations around the late afternoon but this trend was not consistent, as these are not observed on the following late afternoon. In addition, DMS and $DMSP_t$ maximal concentrations were decoupled from each other.

4.2 Variability during a cyclone in the Bay of Bengal

Variations in DMS (surface as well as MLD-averaged) and wind are shown in Figure 9. During phase I (SK147A), the sky was largely overcast with rain clouds and wind speed varied between 6.02 m s⁻¹ and 18.2 m s⁻¹ with an average wind speed of 11 m s⁻¹, while in phase II (SK147B) the sky was largely clear with wind speeds varying between 4 m s⁻¹ and 12 m s⁻¹ with an average wind speed of 7.7 m s⁻¹. The DMS variability was significant during phase I in response to intense atmospheric convection. Phase I shows spikes in surface as well as MLD averaged DMS. Phase II does not show such spikes in DMS. In spite of this difference the overall range and average during both the phases remain largely similar. During phase I,

surface DMS varied between 1.5 and 5.5 nM with an average of 3 nM, while in phase II it varied between 1 and 5.3 nM that again averaged to 3 nM. The column DMS (0-200 m) in phase I ranged between undetectable levels and 6.3 nM with an average value of 1.4 nM and in phase II it varied from below detection limits to 5.3 nM with a mean of 1.2 nM.

4.3 Seasonal variation in waters off Goa, west coast of India

Data obtained through periodic sampling along a small transect (~13 km, Fig 1b) off the Candolim coast (Goa) enabled us examine the extent of seasonal variability in DMS and DMSP_t . During the premonsoon (March) of 2001 (Figure 10a) concentrations of DMS and DMSP_t varied, respectively, from 0.2 to 0.9 nM with an average value of 0.6 nM and between 0.5 and 2.9 nM with a mean of 1.4 nM. These concentrations changed in SW monsoon as upwelled water with low oxygen and high nitrate dominate the coastal regions (Naqvi et al., 2000). DMS and DMSP_t are, in general, is higher at surface because of intense biological activities at surface. Very high photosynthetic fixation and the presence of traces of nitrate seem to have aided higher production of DMSP_t (9-113 nM) and DMS (13-132 nM). The DMS levels found in these coastal waters on 29 September 2000 (Figure 10b) are about 100 times more than that occurred in March 2001. On the other hand, the highest value for DMS of 525 nM was found very close to the coast in November 1999, where DMS levels varied from 6 nM to 525 nM and DMSP_t varied between 8 nM and 339 nM (Figure 10c). Coinciding with this trend, extremely high DMSP_t values were found in the same season in the neighbouring Zuari estuary (~419nM) and in western continental marginal waters off Mangalore (~916 nM). Thus, DMS varied from 0.2 nM in premonsoon season to 525 nM in fall-intermonsoon (or post-SW monsoon) season exhibiting a variability by 2500 times in coastal waters of Goa. Such behavior could be occurring in at least a few other places along the west coast of India since the oceanographic conditions are similar.

4.4 Inter-annual variability (Central Indian Ocean)

Subsequent cruises (FFP- 1988 and IFP-1999) undertaken as part of the INDOEX programme throw light on the inter-annual variability of DMS in the Central Indian Ocean (Figure 7). A clear increase (nearly five times) in DMS concentrations was found in 1999 in comparison to that in 1998. During FFP '98 the DMS concentrations in the upper 100 m varied between 0.4 and 2.6 nM with an average value of 0.6 nM whereas during IFP '99 DMS concentrations varied between 0.1 nM and 13.9 nM with an average of 3.4 nM. This high degree of inter-annual variability is due to differential biological production during the two years (Shenoy et al, 2002).

Discussion

1. DMS at sea surface and its emission

1.1 Variations in surface DMS

The envisaged indirect global cooling by DMS products depends on its emission from the ocean.

Variations in surface DMS in the Indian Ocean are shown in Figure 11. During the Northeast monsoon the surface DMS in coastal waters of the Arabian Sea (central and southern regions of the west coast of India) varied between 0.7 nM and 31.7 nM with an average of 6.3 nM while in the open ocean it was between 0.3 nM and 10.9 nM with a mean of 3.3 nM. The average DMS concentrations in the coastal region are nearly twice that of the open ocean. During the SW monsoon the surface DMS in coastal waters varied between 0.5 nM and 525 nM (the highest value is not shown in Figure 11) with an average value of 10.3 nM. The highest value was found in near shore waters of Goa due to very high primary production driven by coastal upwelling and the associated biological processes including hypoxia. In contrast, the intermonsoon, which is a slack period with respect to primary production but rich in bacterial production, experienced low concentrations of DMS. The coastal surface DMS varied between 0.9 nM and 10.7 nM with an average concentration of 2.6 nM while the open ocean surface DMS varied between 0.9 and 1.7 nM (average of 1.3 nM) in this season. The mean DMS concentrations at surface were greater in SW monsoon (10.3 nM) followed by winter (4.3 nM) and intermonsoon (2.1 nM). The annual surface DMS average, in general, for the coastal Arabian Sea was 8.2 nM whereas it was 3 nM for the open Arabian Sea. Thus the surface DMS in the Arabian Sea shows very high spatial and temporal variability. The overall surface DMS average for the Arabian Sea was 6.3 nM.

On the other hand, surface DMS concentration in the coastal waters (off the Paradip in the north and Chennai in the south) in the western Bay of Bengal varied from 1.2 to 3.9 nM with an average of 2.6 nM during the SW monsoon when the central Bay waters contained the surface DMS concentrations of 0.2 – 11.1 nM having a mean of 2.7 nM. The surface DMS concentrations are almost the same in the coastal and open waters of the Bay of Bengal. During intermonsoon, the southern and central Bay of Bengal exhibited surface DMS concentrations between 1.4 and 4.7 nM with a mean of 2.9 nM. The overall average surface DMS concentration in the Bay of Bengal was 2.7 nM. In the Central Indian Ocean surface DMS ranged from 0.2 to 11.9 nM during the northeast monsoon with an average concentration of 2.2

nM. All the observations made in the Central Indian Ocean were in winter as a part of the INDOEX project. The surface DMS in the Arabian Sea was more than double to that in the Bay of Bengal and nearly thrice than that in the Central Indian Ocean. In general the surface DMS concentrations in coastal waters were higher than in the open ocean. The overall average surface DMS for the entire Indian Ocean covered in the present study was 4.2 nM.

Barnard et al. (1982) reported a surface DMS range of 0.55-23.22 nM with a mean of 2.8 nM for the Atlantic Ocean. Wong et al., (2005) in a time series study in the subarctic northeast Pacific Ocean (an HNLC region) found that the mean surface DMS was low in winter (2 nM) in comparison to summer (10 nM). High variability in surface DMS concentrations is found in the southern North Sea with minimal concentrations of 0.13 nM in winter to a maximum of 25 nM in May, which coincided with large blooms of *Phaeocystis pouchetti* (Turner et al., 1996). In the Arctic Ocean surface DMS is found to vary between 0.04 and 12 nM with the maximal values occurring along the ice edge (Leck and Persson, 1996). Surface DMS concentrations in waters around North America have been detected to range between 0.1 and 12.6 nM with an average of 2.2 nM where the highest values occurred in the western Arctic and near the Sargasso Sea on the east coast of the United States (Sharma et al., 1999). Surface DMS concentrations have been found to range between 0.002 to 4.63 nM with a mean of 2.6 nM in the South China Sea, and from 1.8 to 5.7 nM (a mean of 3.4 nM) for the East China Sea, where the highest concentrations occurred in shelf waters and DMS showed strong correlation with chlorophyll (Yang et al., 2000). Uher et al. (2000) found highly variable surface DMS concentrations in the European western continental margin. In September 1994 it ranged between 0.6 and 33.4 nM (mean of 2.8 nM) while in July 1995 it varied from 2.9 to 38.5 nM (an average of 7.2 nM). It is apparent from the above comparisons that the Indian Ocean surface DMS average is nearly double to that of the Atlantic, the Pacific and the other seas in the Northern hemisphere and is also more than the surface DMS concentrations reported for the China Sea. Our values are, however, comparable to the July 1995 average for the European western continental margin. In addition we also compared our data with the data set for the Indian Ocean from the PMEL database (Pacific Marine Environmental Laboratory). Data from <http://saga.pmel.noaa/dms/> were downloaded for the Indian Ocean region which basically covered central and western Arabian Sea and Central Indian Ocean over different seasons. The surface DMS over this coverage varied between 0.76 nM and 12.05 nM with an average of 3.7 nM. Our surface DMS average for the entire Indian Ocean is comparable with that of the PMEL database. A bit higher average surface DMS for the present study might have resulted from the fact that this study covers a lot

of the Indian coastline which is influenced by intense biological activity driven by monsoons (Naqvi et al., 2000).

1.2 Winds

Wind is an important driving force in air-sea interaction processes that largely determines the extents of exchanges of heat and materials. The Indian Ocean covered in the present study experienced seasonally variable winds. During the northeast monsoon wind speeds along the eastern coastal Arabian Sea were between 0.7 and 10.4 m s⁻¹ with an average speed of 4.2 m s⁻¹ while in the open ocean it varied from 1.4 to 9 m s⁻¹ with a mean of 4.9 m s⁻¹. During the SW monsoon the winds along the coastal Arabian Sea of India were weaker and varied between 0.5 and 8 m s⁻¹ with an average speed of 3.1 m s⁻¹. During the intermonsoon periods, it ranged between 1.8 and 12.5 m s⁻¹ and 3.7 and 6.3 m s⁻¹ with average wind speeds of 5.8 and 5.2 m s⁻¹ for the coastal and the open ocean regions, respectively. Thus maximal wind speeds in the Arabian Sea were encountered during the intermonsoons along the west coast of India. Along the east coast of India, winds varied between 2.2 and 8.9 m s⁻¹ with an average speed of 5.5 m s⁻¹ during the SW monsoon while in the open bay winds varied from 4 m s⁻¹ to 18.2 m s⁻¹ (in association with a low pressure system), with an average speed of 9.3 m s⁻¹. During the intermonsoons, however, winds in the open bay varied between 1.6 and 8.6 m s⁻¹ (an average of 3.8 m s⁻¹). In the Central Indian Ocean the winds showed a variation from 1.1 to 12.7 m s⁻¹ (with a mean of 5.7 m s⁻¹) in winter. These wind speeds were higher in 1999, than in 1998, because of the development of stormy conditions associated with the Inter Tropical Convergence Zone. The measured wind speeds were in general agreement with the climatological winds recorded in Hastenrath and Lamb (1979). The observed wind speeds in the eastern Arabian Sea, however, are much weaker than those reported for the western parts (~ 18 m s⁻¹) in the southwest monsoon.

1.3 Sea-to-air fluxes of DMS

In DMS flux calculations it is assumed that DMS in air is negligible compared to that in seawater and therefore the concentration difference used in the flux calculation is essentially equal to DMS concentration in seawater. Figure 12 depicts variations in surface DMS flux in the coastal and open Ocean areas of the Indian Ocean. The DMS fluxes from the coastal waters of India are lower than that from the open Ocean. Higher DMS fluxes in the Central Indian Ocean are associated with higher DMS abundance and high wind speeds in winter of 1999. In the coastal areas the DMS flux varied between 0.04 μmol m⁻² d⁻¹ and 34.4 μmol m⁻² d⁻¹ with an average value of 3.4 μmol m⁻² d⁻¹ while in the open Ocean it varied from 0.03 μmol m⁻² d⁻¹ to

41.1 $\mu\text{mol m}^{-2} \text{d}^{-1}$ having a mean DMS flux of 6.3 $\mu\text{mol m}^{-2} \text{d}^{-1}$. Table 2 lists the DMS fluxes from the Indian Ocean both on seasonal and regional basis. Average DMS fluxes were higher in the northeast monsoon than in other seasons both in coastal and open Arabian Sea regions. During the intermonsoon period, open Arabian Sea fluxes were nearly half of that in winter. Comparatively low winds were found when the measurements were made in coastal waters of the Arabian Sea in summer monsoon. The annual coastal mean DMS flux was 3.4 $\mu\text{mol m}^{-2} \text{d}^{-1}$ while the open ocean mean flux was 3.8 $\mu\text{mol m}^{-2} \text{d}^{-1}$ resulting in an average flux from the Arabian Sea of 3.5 $\mu\text{mol m}^{-2} \text{d}^{-1}$. The fluxes estimated here for the Arabian Sea have to be considered to be low since our coverage is limited to central and eastern parts and excluded regions of strong winds during the southwest monsoon. Fluxes in the Bay of Bengal were higher in the SW monsoon than in intermonsoon. Higher fluxes (average of 11.1 $\mu\text{mol m}^{-2} \text{d}^{-1}$) during the BOBMEX time series experiment resulted from the enhanced production of surface DMS due to increased biological activity triggered by turbulent conditions and strong winds associated with the low pressure system. A large variability in flux at this time-series location is seen in Figure 12. In the Central Indian Ocean the DMS flux varied between 0.06 and 29.1 $\mu\text{mol m}^{-2} \text{d}^{-1}$ with an average value of 3.5 $\mu\text{mol m}^{-2} \text{d}^{-1}$. Thus the Bay of Bengal exhibited the maximal DMS fluxes during the present study (due to the prevailing low pressure systems) with nearly three times to that in the Arabian Sea and in the Central Indian Ocean. The overall DMS flux for the entire Indian Ocean is estimated to be 5.42 $\mu\text{mol m}^{-2} \text{d}^{-1}$. Table 3 shows a comparison between the DMS flux from the Indian Ocean (this study) with those estimated for other regions. It is evident that flux from the Indian Ocean is higher than in many regions of the world Oceans, e.g. the north Pacific, the Atlantic, the North Sea and the Arctic Polar Ocean, but is lower than that reported for the northeast Pacific Ocean. The only DMS estimate for the Indian Ocean, available from the earlier studies, is that of Nguyen et al. (1990) near the Amsterdam Island situated near 38°S, which is nearly one-half of that in the present work. However, our flux estimate agrees well with that made by Yang (2000) for the South China Sea. In the present study an area of around $16.1 \times 10^6 \text{ km}^2$ was covered for DMS and DMSP_t studies which amount to about 21.5% of the total Indian Ocean area or around 4.5% of the global oceanic area. The total flux of DMS from the study area was estimated to be $1.02 \times 10^{12} \text{ g S y}^{-1}$. This works out to contribute 4.1- 6.3 % to the global DMS emission, which has a range of $16\text{-}25 \times 10^{12} \text{ g S y}^{-1}$. One of the limitations of the study is the lack of coverage of certain important areas in the Indian Ocean such as the intense upwelling zones off the coast of Somali and Oman, east coast of Africa, west coast off the Indonesian Islands and the south Indian Ocean. Considering the fact that this study covered only 21.5% of the total Indian Ocean, the Indian Ocean could be an

important source for DMS-sulphur to the atmosphere. It should, however, be cautioned that the flux trends observed among various basins in the Indian Ocean studied here may be different from that expected since we occupied the Bay of Bengal (BOBMEX) and Central Indian Ocean (INDOEX) under turbulent conditions but could not cover high wind regimes in the Arabian Sea during the southwest monsoon.

Conclusions

Significant spatial and temporal variability in DMS was found in the North and central Indian Ocean. Very high DMS and DMSP of 526 nM and 916 nM, respectively, were found in coastal waters of the Arabian Sea. These are among the highest values found in the world oceans. In the Arabian Sea the lowest DMS and DMSP_t concentrations were observed during the intermonsoons. Lower DMS and DMSP_t concentrations were found to occur in the Bay of Bengal compared to that in the Arabian Sea which might have been due to relatively less biological production and trophic diversity in the bay, as also can be seen from generally lower DMSP_t levels in the bay. In addition DMS and DMSP showed high temporal variability in terms of day to day variation and seasonal as well as inter-annual variation. Average surface DMS is the highest in the Arabian Sea whereas the average flux is the highest from the Bay of Bengal. DMS from the study area was found to contribute 4-6% to the total global emission, which is comparable to other regions of the world. Export fluxes of sulphur compounds are important from Indian Ocean since this is one of the most turbulent regions, with regular occurrence of monsoons.

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Figure captions:

Figure 1. a) Station locations occupied for dimethyl sulphide studies in the central and north Indian Ocean. Symbol index indicates cruise number, season and number of stations. b) Station locations off Candolim, Goa.

Figure 2. Vertical profiles of temperature, salinity, density, dissolved oxygen and nitrate in the northern Arabian Sea during the northeast monsoon (SS161).

Figure 3. Profiles of temperature, nitrate and chlorophyll (at 9.94°N, 72.44°E, SK158) showing subsurface chlorophyll maximum.

Figure 4. Typical vertical profiles of DMS and DMSP_t in different regions of the Indian Ocean.

Figure 5. Longitudinal variations in DMS off Goa in different seasons.

Figure 6. Variations in DMS and DMSP_t off Paradip (a) and (b) during SK147A and (c) during SK147B and off Chennai (d) during SK147B.

Figure 7. Variations in DMS (nM) in the Central Indian Ocean in winter season of 1998 (FFP; SK133) and 1999 (IFP; SK141) of the INDOEX project.

Figure 8. Variations in salinity, nitrate, DMSP_t and DMS at a time series station (7°N, 87°E) during the BOBMEX Pilot Experiment (SK138C). Zero hours refer to 03.30 AM on 30 October 1998. Night time is indicated by dark rectangles on the x-axis.

Figure 9. Variations in wind speed and DMS at 17.5°N and 89°E during the southwest monsoon of 1999 (SK147A&B); open symbols indicate surface values and bold symbols indicate MLD averaged values. Here 0 hour refers to 0 Hour of 27/7/99. The weather was cloudy during 31 July – 1 August 1999.

Figure 10. Time series observations of DMS (Left panels) and DMSP_t (Right panels) off Candolim in Goa on (a) 28/3/2001, (b) 29/9/2000 and (c) 11/11/1999.

Figure 11. Variations in surface DMS concentration in the Indian Ocean. Maximum concentration of 525 nM found in the coastal waters of Goa is not shown here.

Figure 12. Variations in DMS flux in the Indian Ocean. Time series station (SK147A & B) in the Bay of Bengal depicts wide variations in the DMS flux.

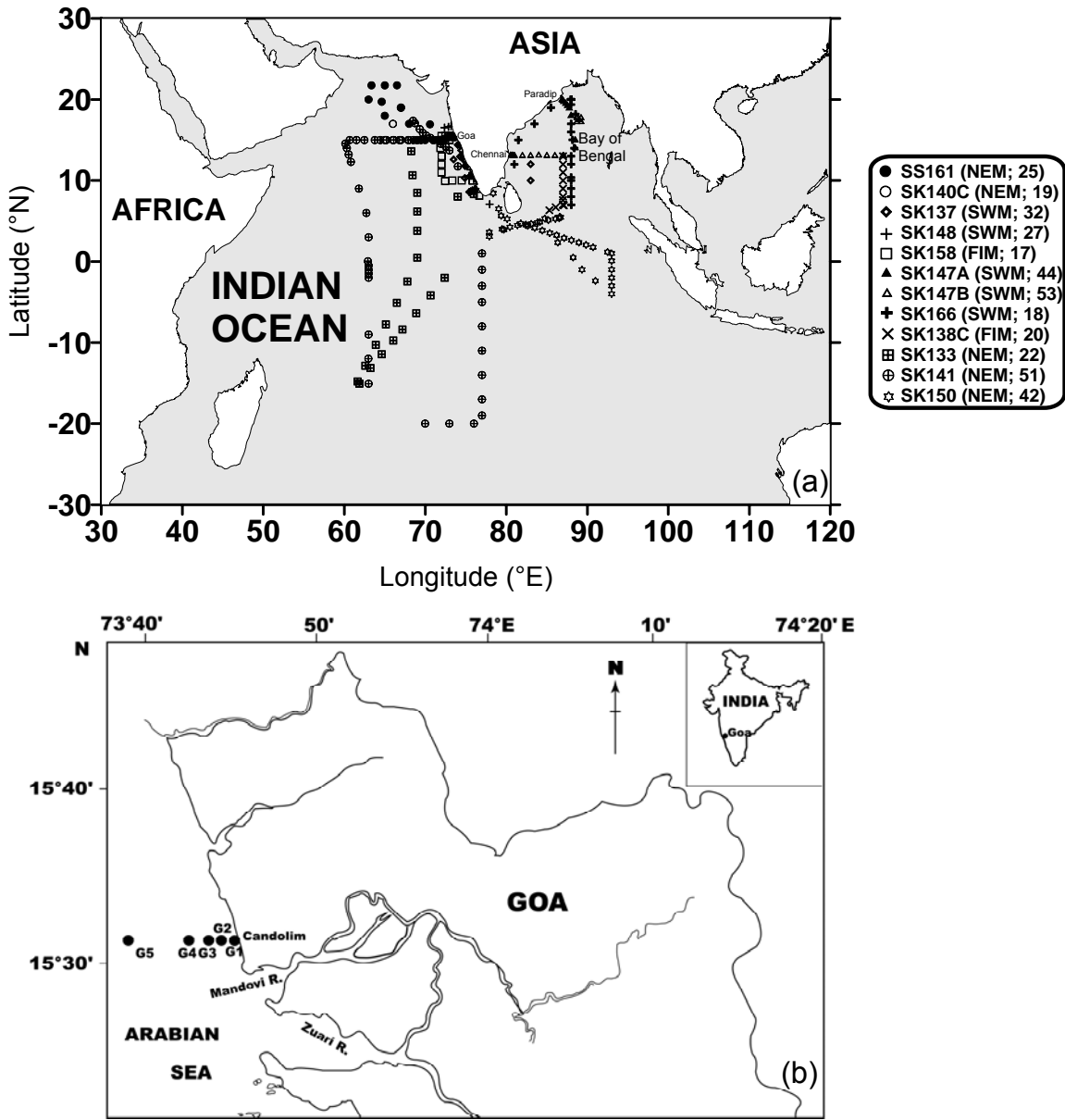


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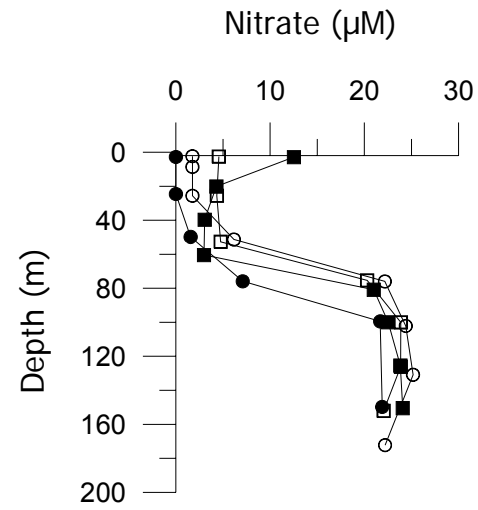
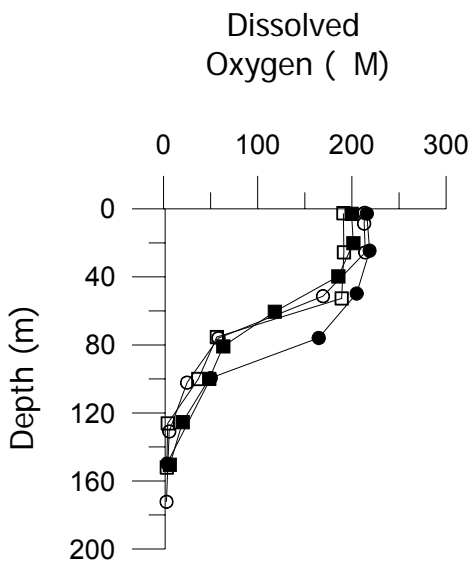
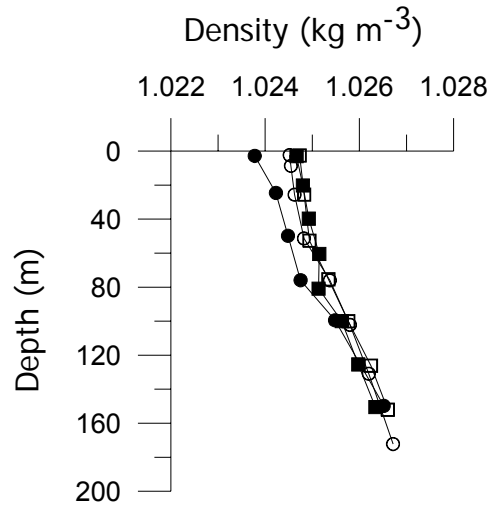
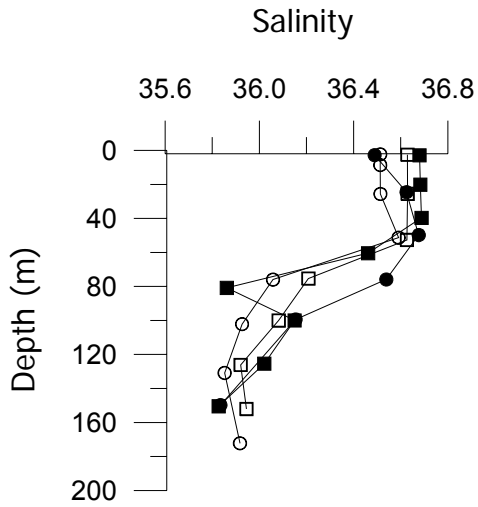
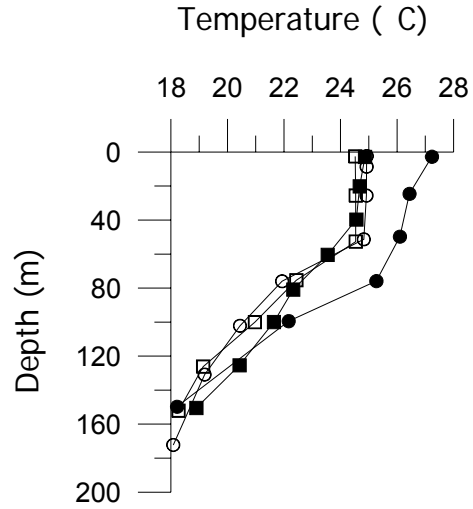
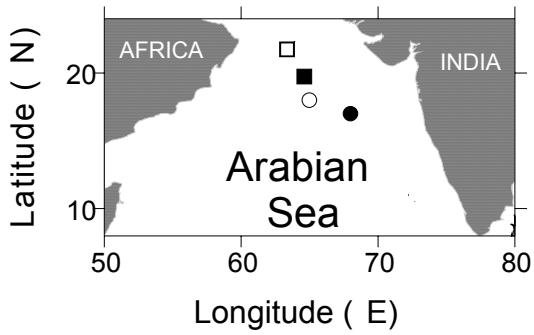


Fig. 2

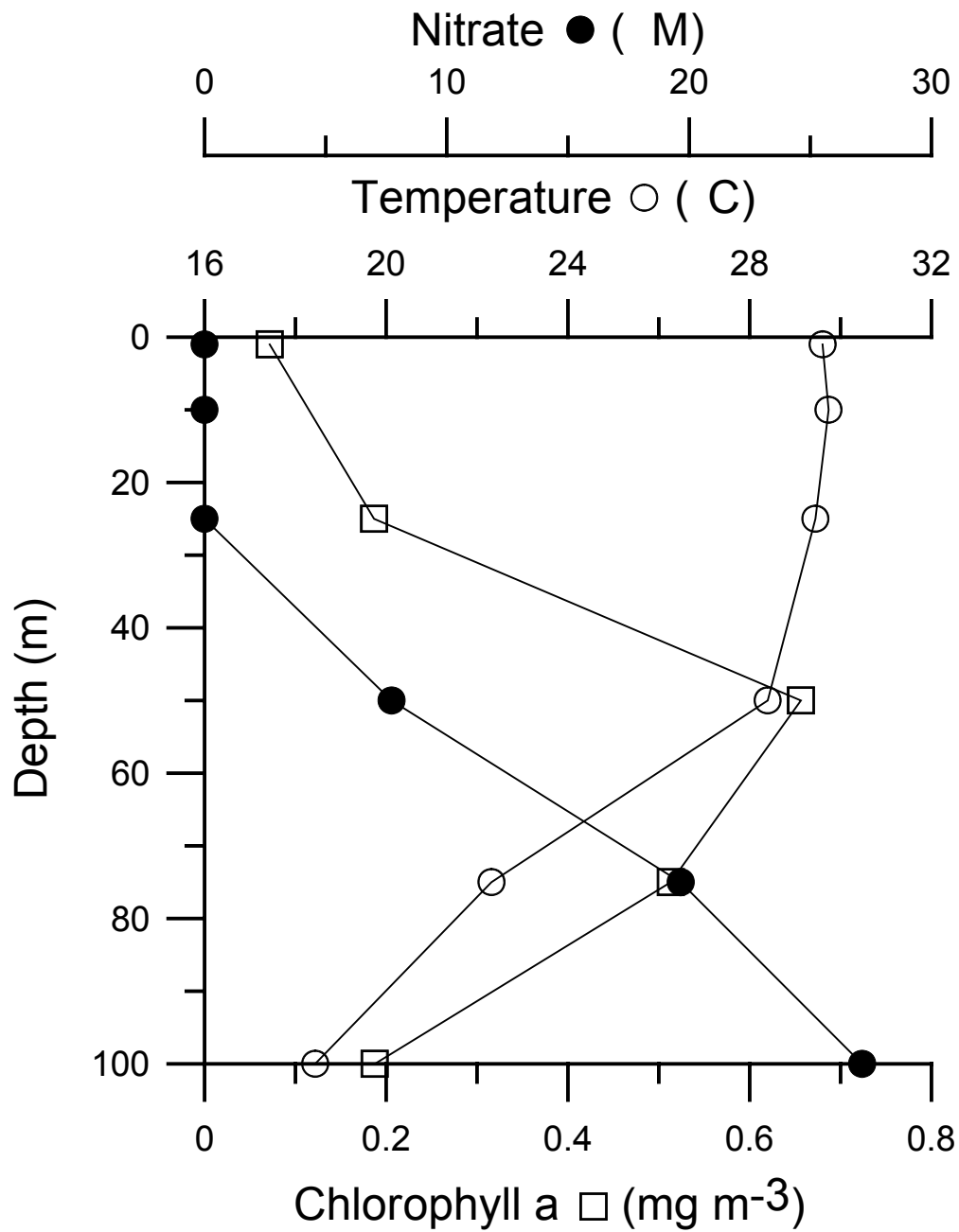


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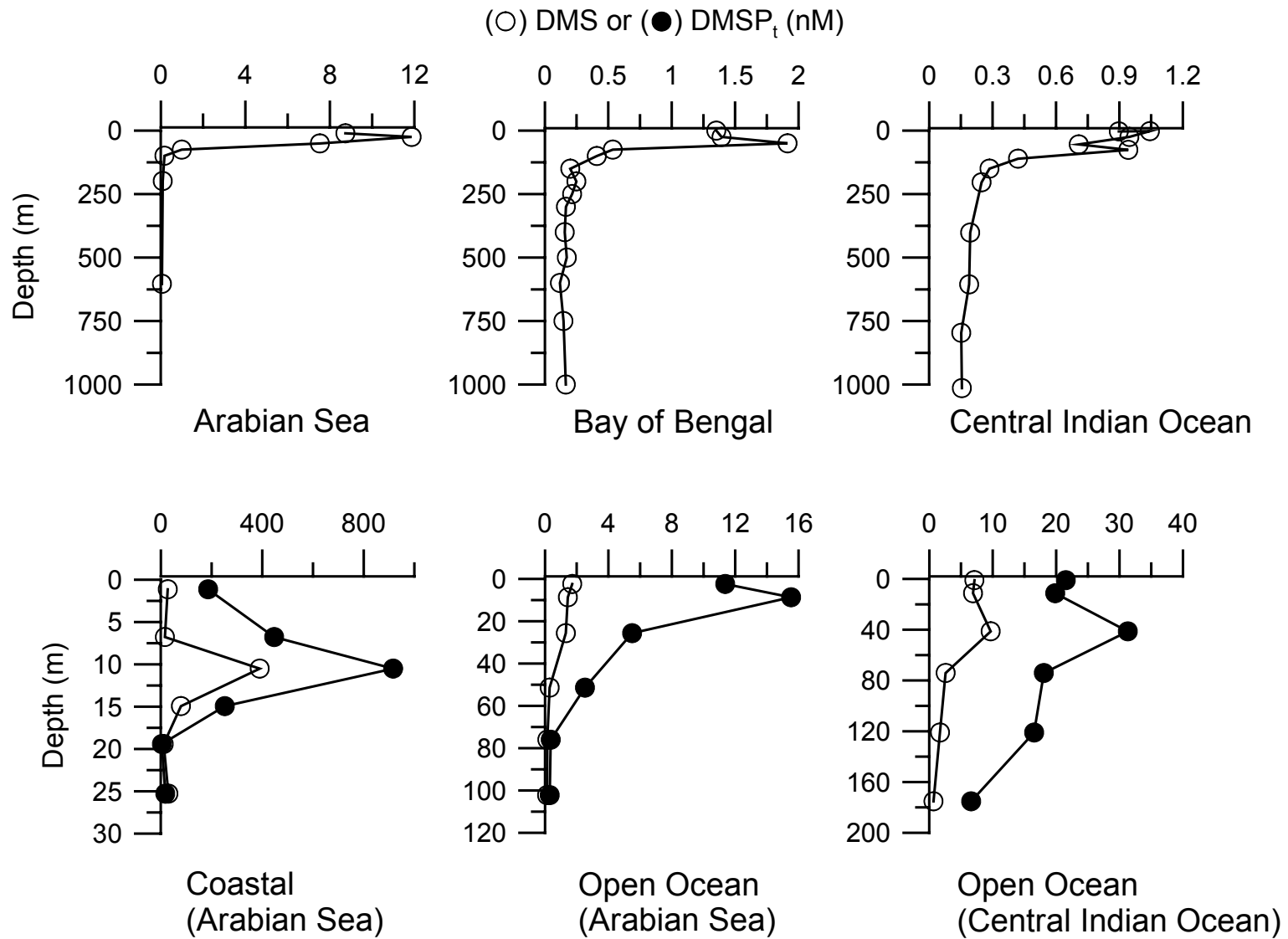


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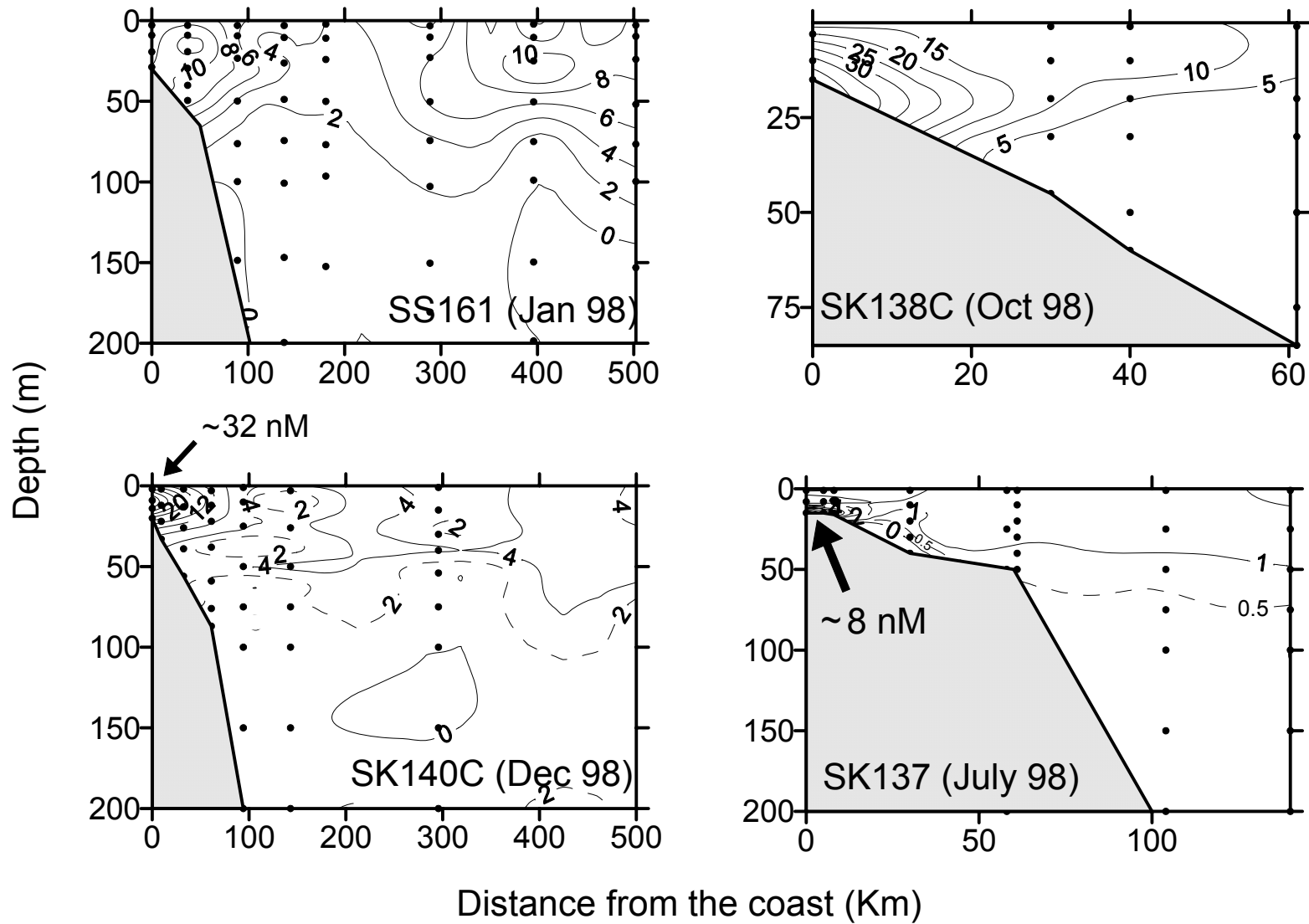


Figure 5. Longitudinal variations in DMS off Goa in different seasons.

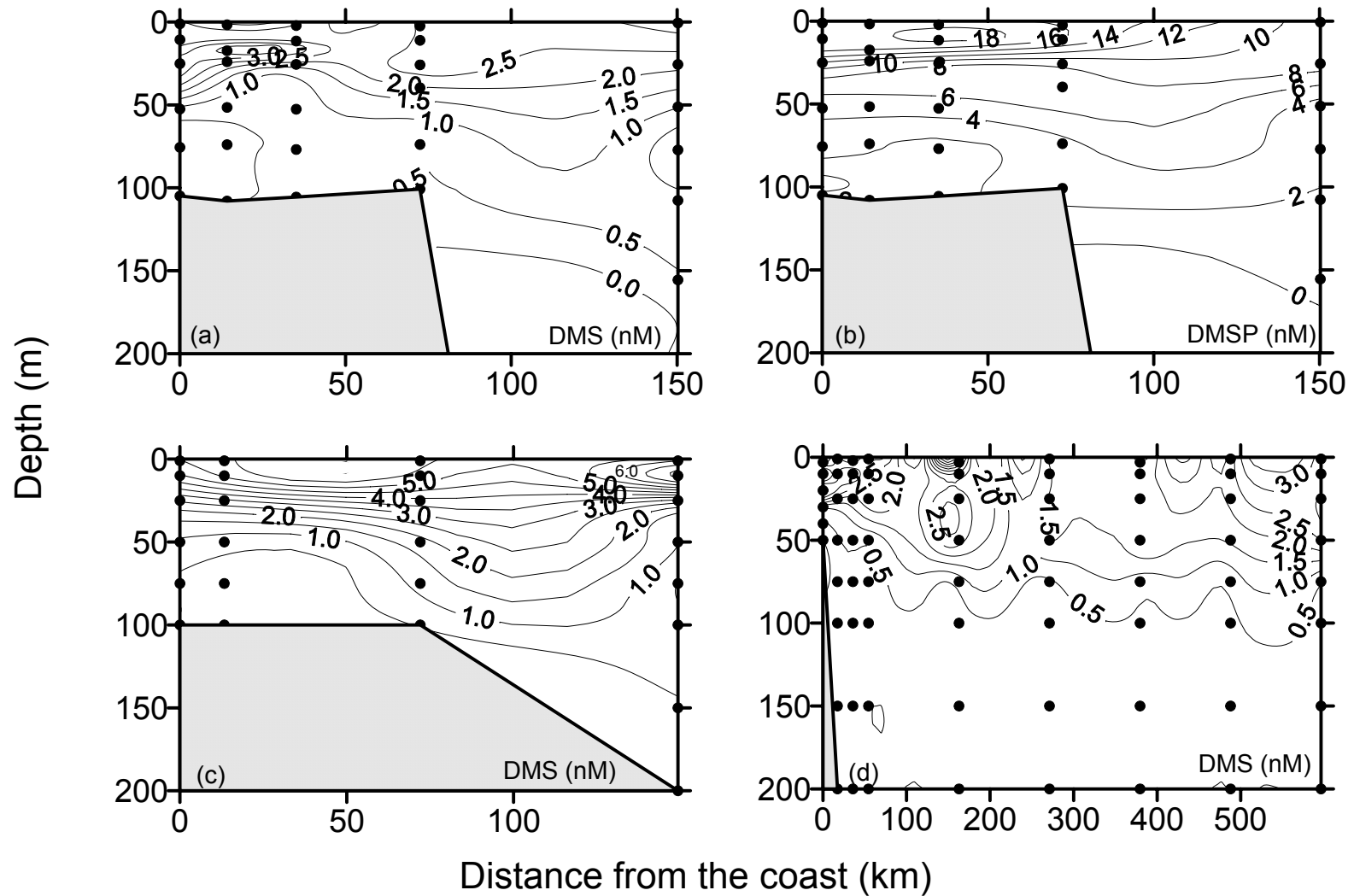


Figure 6. Variations in DMS and $DMSP_t$ off Paradip (a) and (b) during SK147A and (c) during SK147B and off Chennai (d) during SK147B.

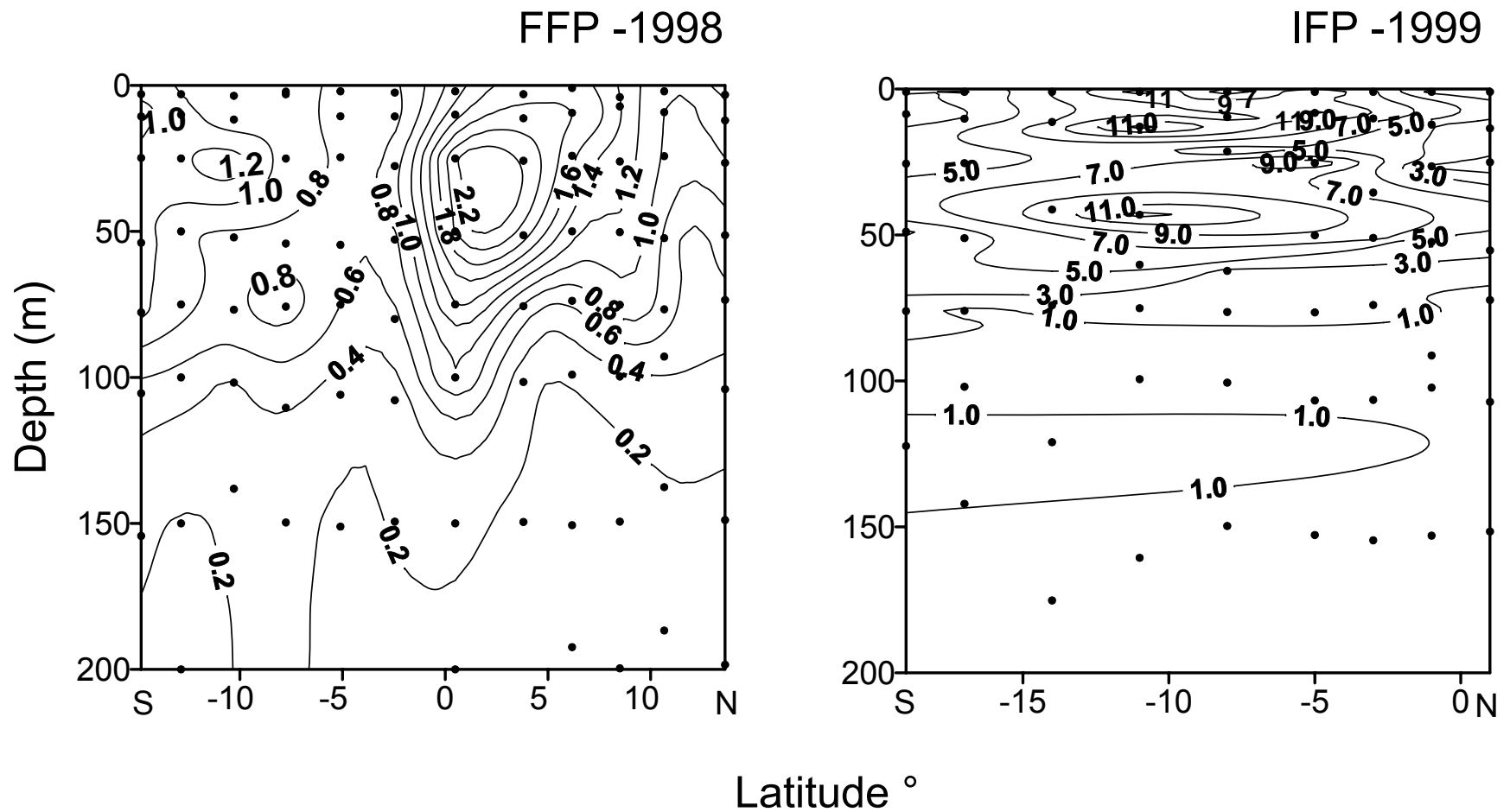


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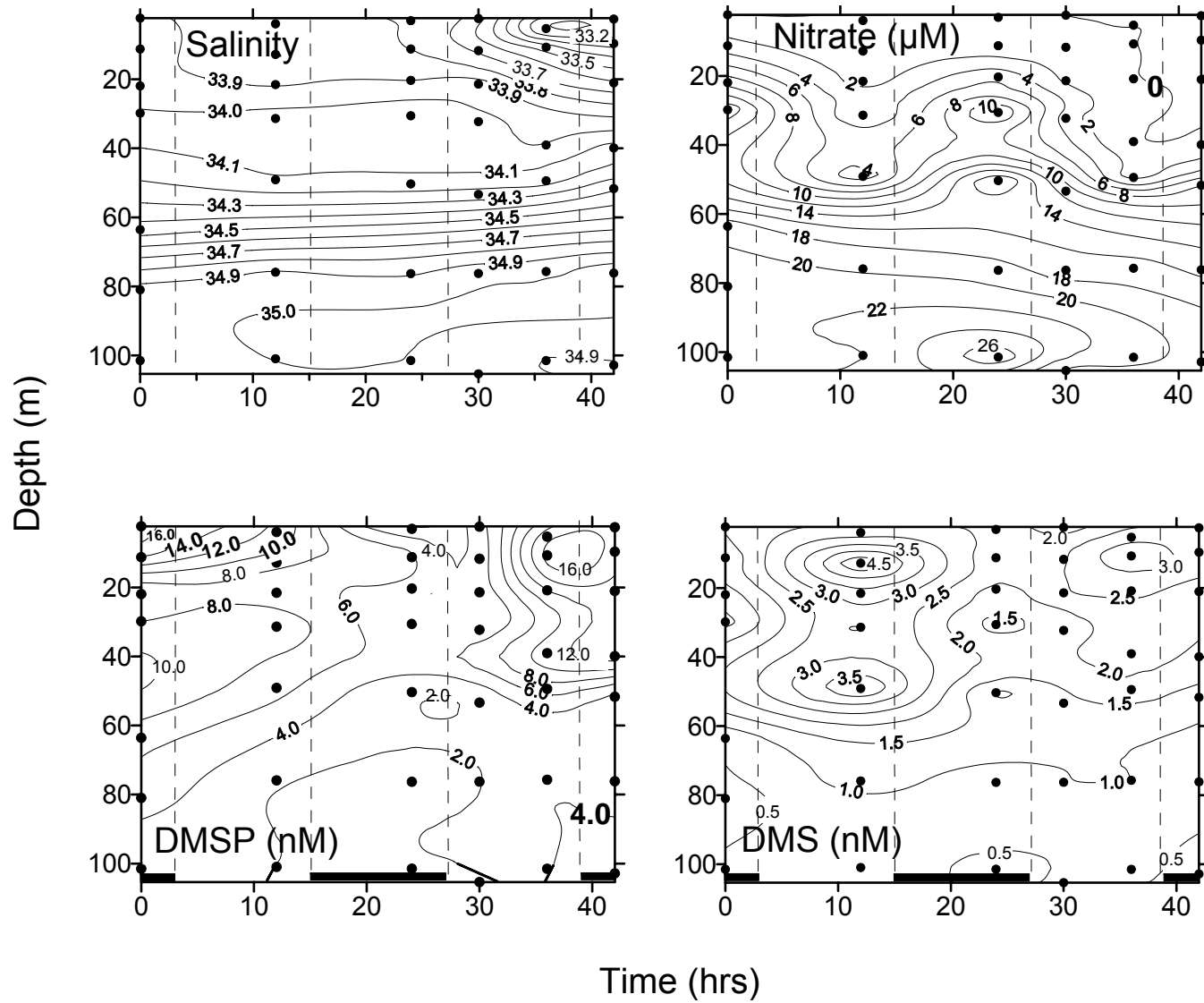


Fig. 8

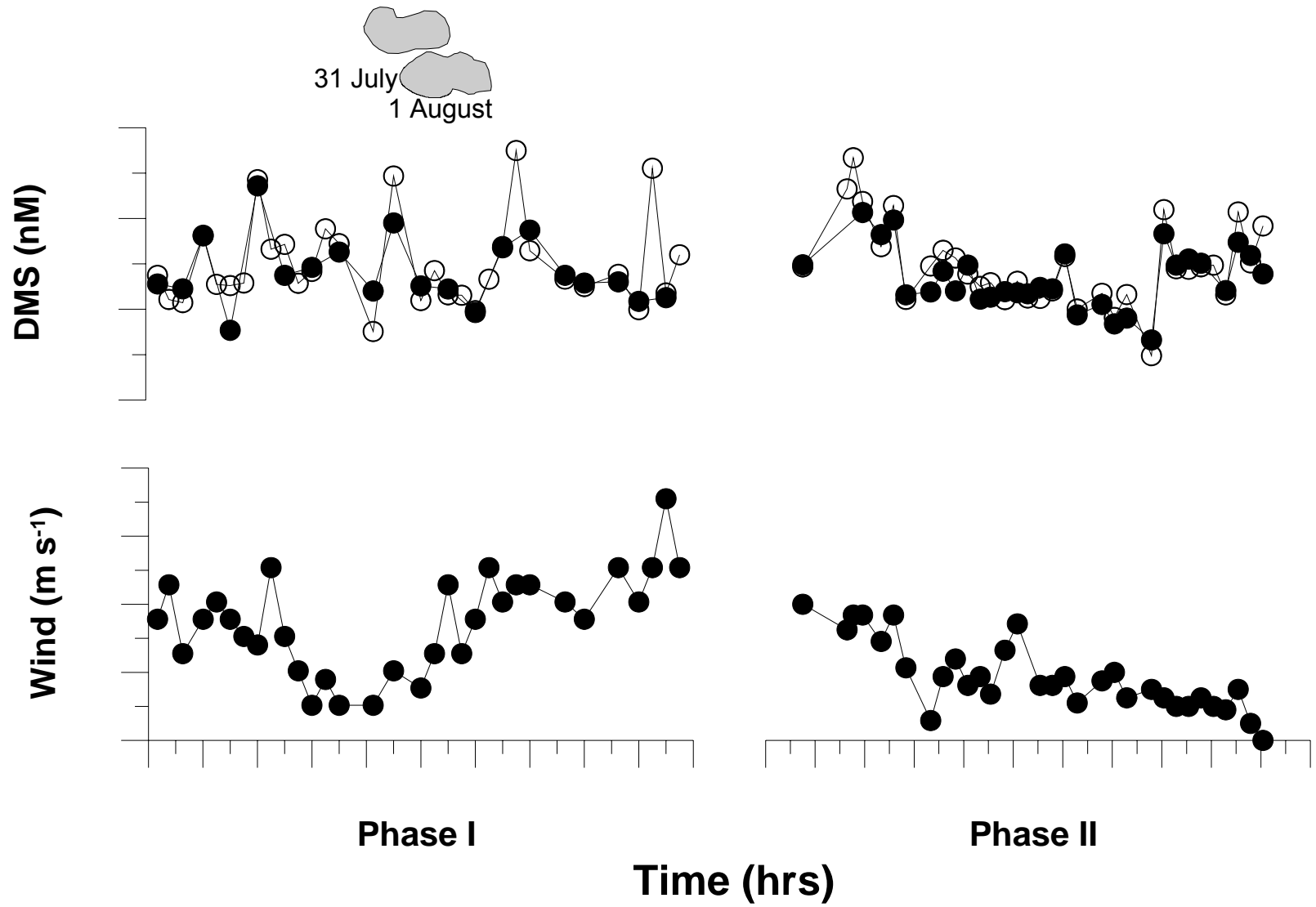


Fig. 9

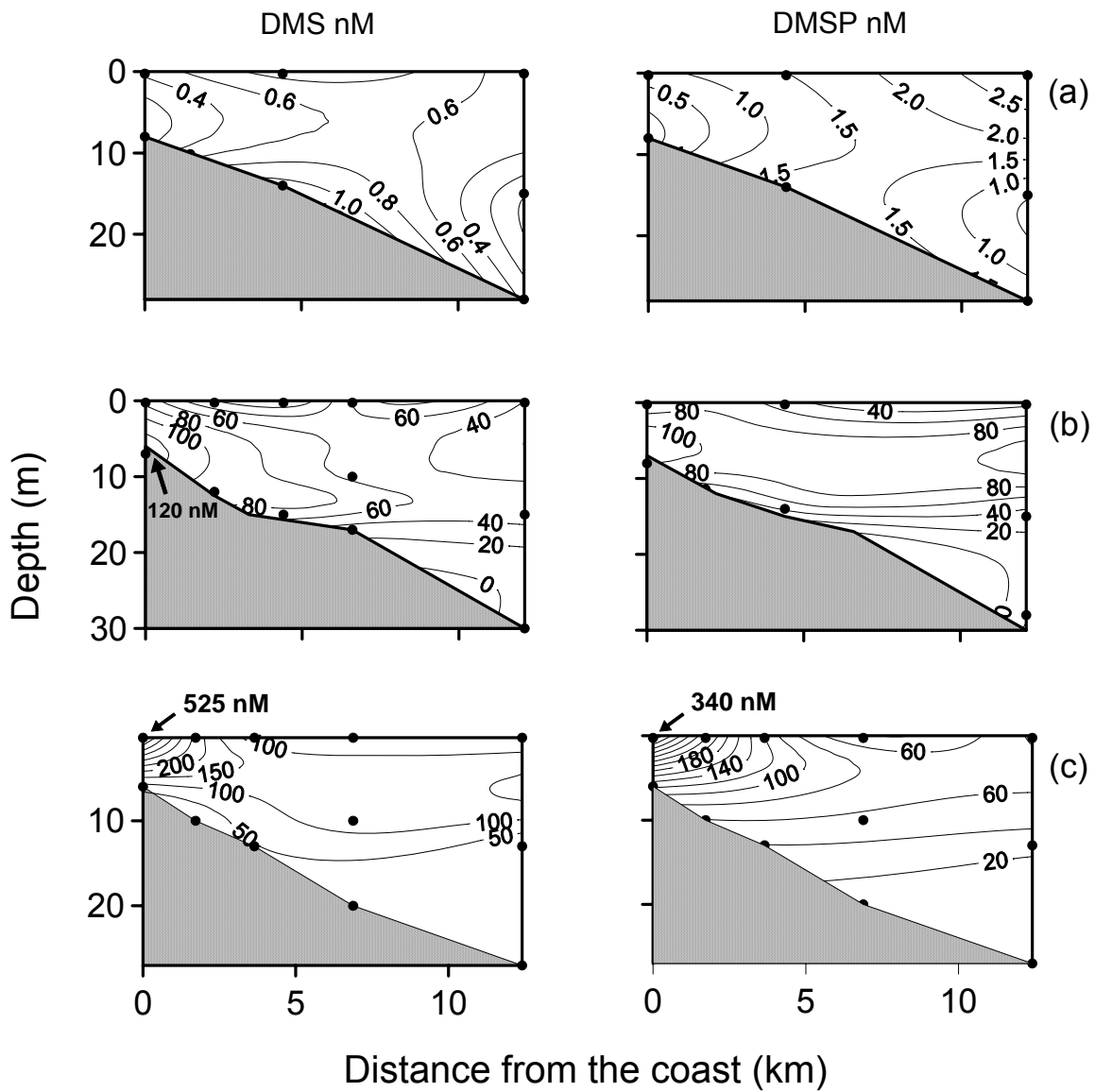


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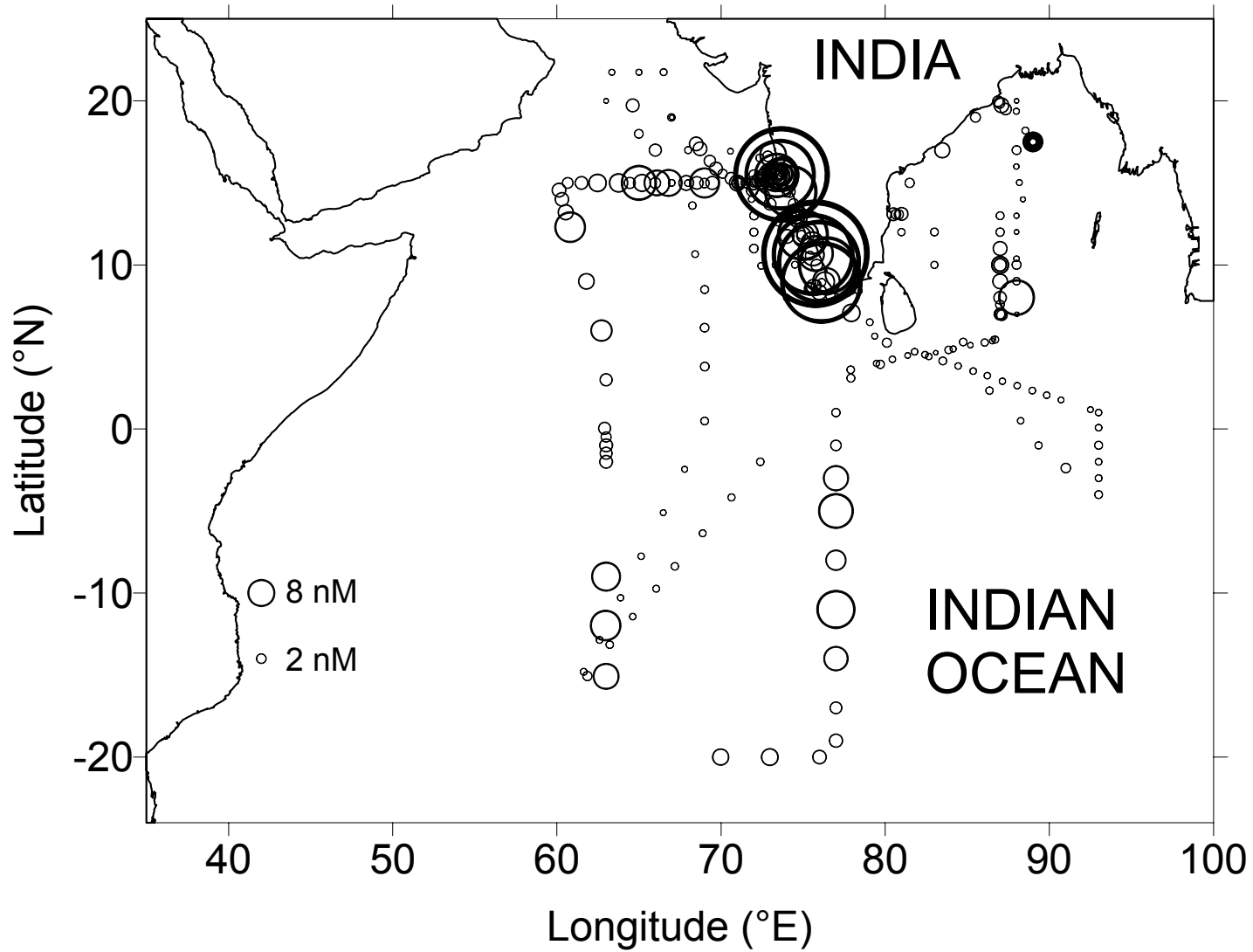


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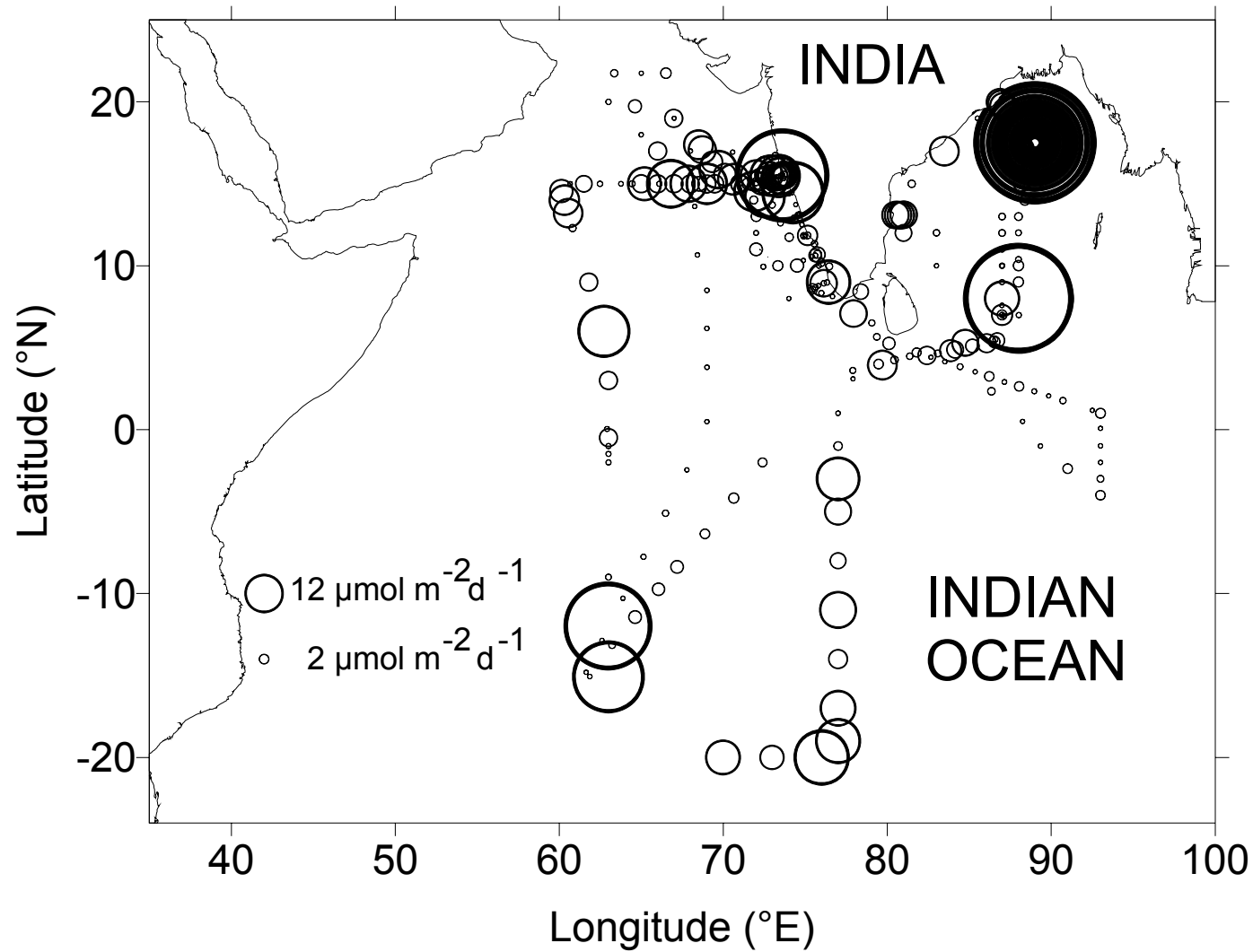


Figure 12. Variations in DMS flux in the Indian Ocean. Time series station (SK147A & B) in the Bay of Bengal depicts wide variations in the DMS flux.

Table 1. Averages and ranges (in parenthesis) of DMS and DMSP_t in the surface mixed layer (defined by 0.125 kg m⁻³ density difference) in different seasons and in different regions of the Indian Ocean.

Area	Season	Coastal		Open Ocean	
		DMS (nm)	DMSP (nM)	DMS (nm)	DMSP (nM)
Arabian Sea	SWM	32.9 (0.3 – 525.6)	81.1 (2.0 – 915.9)	1.6 (0.5 – 3.7)	NM
	NEM	9.0 (1.8 – 51.0)	NM	2.3 (0.2 – 11.9)	9.8 (3.6 – 16.7)
	FIM	5.8 (0.2 – 64.4)	26.1 (0.5 – 160.1)	1.4 (0.9 – 2.2)	9.4 (4.9 – 16.1)
Bay of Bengal	SWM	3.3 (1.2 – 7.2)	12.9 (3.1 – 23.3)	2.8 (0.2 – 11.1)	13.3 (1.5 – 30.9)
	FIM	NM	NM	2.8 (0.8 – 5.5)	10.9 (4.1 – 19.5)
Central Indian Ocean	NEM	-	-	2.7 (0.2 – 13.9)	10.1 (1.3 – 35.9)

NM: Not measured

SWM, NEM and FIM refer to southwest, northwest and fall inter-monsoon seasons, respectively.

Table 2. Sea-to-air fluxes of DMS ($\mu\text{mol S m}^{-2} \text{d}^{-1}$) in the Indian Ocean *.

Area/Season	Coastal Ocean			Open Ocean			Total		
	Min	Max	Mean	Min	Max	Mean	Min	Max	Mean
Arabian Sea									
NE Monsoon	0.05	34.4	4.7	0.03	16.8	4.2	0.03	34.4	4.3
SW Monsoon	0.04	31	2.6	-	-	-	0.04	31	2.6
Intermonsoon	0.1	13.1	3.9	0.4	3.1	1.6	0.1	13.1	3.13
AVERAGE	0.04	34.4	3.4	0.03	16.8	3.8	0.03	34.4	3.5
Bay of Bengal									
SW Monsoon	0.18	9.03	3.7	0.4	41.1	11.5	0.18	41.1	11.1
Intermonsoon	-	-	-	0.16	10.9	1.63	0.16	10.9	1.63
AVERAGE				0.16	41.1	11.02	0.16	41.1	9.63
Central Indian Ocean									
NE Monsoon				0.06	29.1	3.5	0.06	29.1	3.5
AVERAGE				0.06	29.1	3.5	0.06	29.1	3.5
Indian Ocean	0.04	34.4	3.4	0.03	41.1	6.3	0.03	41.1	5.42

*Wherever data are not available for coastal/open Ocean regions or seasons the available values were assumed to represent the same in other regions/seasons in deriving average fluxes.

Table 3. Comparison of DMS fluxes from the Indian Ocean (this study) with that from other oceanic regions.

Source	Area	Average Flux $\mu\text{mol m}^{-2} \text{d}^{-1}$
Wong et al (2005)	Northeast Pacific Ocean	16
Sharma et al (1999)	North Pacific Ocean	2.3
Berresheim et al (1991)	Western North Atlantic Ocean	3.4
Turner et al (1996)	North Sea	3.84
Leck and Persson (1996)	Arctic Polar Ocean (summer)	2.0
Nguyen et al (1990)	Amsterdam Island (Indian Ocean, 38°S)	2.1
Yang G. P. (2000)	South China Sea	5.5
Yang et al. (2000)	East China Sea	3.4
	Arabian Sea	3.5
	Bay of Bengal	9.63
	Central Indian Ocean	3.5
	Indian Ocean (total)	5.42

This Study