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A study of the carbon dioxide system in the northern Indian Ocean during premonsoon

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Abstract

Extensive data sets on the carbon dioxide system in the northern Indian Ocean have been obtained during the premonsoon season. A marked increase ($\sim 50 \mu\text{M}$) in the total carbon dioxide (TCO_2) content is observed in deep (≥ 1 km) waters of the northern Arabian Sea between approximately 10 and 20°N ; contributions from soft tissue decomposition and skeletal solution to the observed increase are about the same. In the Bay of Bengal, the corresponding increase in TCO_2 observed from 11 to 19°N is relatively small ($17.5 \mu\text{M}$). Significantly, the contribution of the organic decomposition in this region is only $1/7$ of the increase in TCO_2 . This, together with the earlier reports on large sinking fluxes of particulate organic carbon and pronounced near-bottom anomalies, suggests rapid sinking of organic matter with little decomposition in the water column due to the immense fluvial inputs of lithogenic matter into the Bay of Bengal. The greater extent of organic decomposition in subsurface waters of the Arabian Sea is reflected by generally lower pH and higher $p\text{CO}_2$ levels. This also causes the deep waters in the Arabian Sea to be more corrosive to CaCO_3 than those in the Bay of Bengal. However, the TCO_2 concentrations in the deep Bay of Bengal are slightly higher as compared to those in the Arabian Sea, presumably due to the greater CaCO_3 dissolution in deep waters en route the Bay of Bengal. The atmospheric fluxes of CO_2 are found to be larger in the Arabian Sea ($2.9 \text{ mmol/m}^2/\text{d}$) than in the Bay of Bengal ($0.33 \text{ mmol/m}^2/\text{d}$) during the premonsoon season. The overall annual evasion rate of CO_2 from the Arabian Sea and Bay of Bengal has been estimated as $\geq 50 \text{ Tg C}$. These results confirm that the northern Indian Ocean serves as a significant source of atmospheric CO_2 .

1. Introduction

The northern Indian Ocean is different from other oceanic areas in terms of its geographical setting and circulation pattern. The Indian sub-continent splits the region into two zones of vastly different hydrographical regimes. The northeastern Indian Ocean (Bay of Bengal) is an area of positive water balance, where a large excess of precipitation and river runoff over evaporation leads to very low

surface salinities. The northwestern Indian Ocean (Arabian Sea), on the other hand, is a region of negative water balance, where the evaporation far exceeds precipitation and runoff; consequently, the upper layers in this region are much more saline and relatively weakly stratified as compared to those in the Bay of Bengal. Both regions are strongly affected by seasonal changes associated with the monsoons.

The northern Indian Ocean is among the most

productive areas of the world oceans. The productivity is especially high in the northwestern Arabian Sea where intense upwelling occurs during the southwest monsoon season (Qasim, 1977, 1982). The resultant excessive consumption coupled with modest supply of dissolved oxygen through renewal processes leads to the development of an intense oxygen minimum at mid-depths. The acute oxygen-deficiency renders a reducing environment, which causes large-scale nitrate losses with eventual production of molecular nitrogen (denitrification) in the Arabian Sea (Sen Gupta et al., 1976; Naqvi, 1991). The occurrence of denitrification in the Arabian Sea appears to profoundly influence the biogeochemical cycling within the coupled carbon–nitrogen systems. Some recent results suggest that the organic carbon requirement for the microbial metabolism within the denitrifying waters may be severalfold higher than the supply from the surface layer through sedimentation (Ducklow, 1993; Naqvi and Shailaja, 1993). This has led to the suggestions that bacteria could efficiently utilize the dissolved organic carbon (DOC) under reducing conditions and that their proliferation within the denitrifying layer could cause the occurrence of the subsurface turbidity maximum in the Arabian Sea (Naqvi et al., 1993). An investigation on DOC also revealed its non-conservative behaviour and indicated its involvement in denitrification (Kumar et al., 1990). In contrast to the Arabian Sea, the Bay of Bengal does not appear to be an active denitrification site (Rao et al., 1994—this issue).

The northern Indian Ocean appears to serve as a significant source of carbon dioxide (CO_2) to the atmosphere (Takahashi, 1989; Somasundar et al., 1990; Cochran, 1991; Kumar et al., 1992; Naqvi et al., 1993). However, estimates of the CO_2 flux from the northern Indian Ocean suffer from large uncertainties due to inadequate coverage of the region both in space and time. This is especially true for the Bay of Bengal; the only reliable data on the CO_2 system from this region are those generated during the GEOSECS cruises at a few stations located south of latitude 12°N (Weiss et al., 1983). The region receives enormous runoff from three of the world's largest rivers which to a

large extent affects the alkalinity of seawater (Sen Gupta and Pylee, 1968; Takahashi et al., 1980) and the downward transport of organic matter (Ittekkot et al., 1991). Here we provide results of the most extensive measurements of the CO_2 components made so far from the Bay of Bengal and the Arabian Sea.

2. Material and methods

The data sets used for this study were obtained during the premonsoon season. Sampling sites are shown in Fig. 1. Observations in the Bay of Bengal were made during March–April 1991 on board ORV *Sagar Kanya*; the data from the Arabian Sea were obtained using FORV *Sagar Sampada* during February 1992. The data are stored at the Indian National Oceanographic Data Centre, National Institute of Oceanography, Dona Paula, Goa.

Water samples were collected using a Seabird CTD-rosette system fitted with 12 Niskin bottles of 1.8 l capacity. Dissolved oxygen was estimated following the titrimetric Winkler procedure. The pH was measured at 25°C by the photometric technique involving cresol red (Byrne and Breland, 1989) and corrected for the temperature and pressure, in situ, following the procedure of Millero (1979). Total carbon dioxide (TCO_2) content of seawater samples was determined using a coulometer (model 5011 of U.I.C. Inc., USA). Briefly, the analytical procedure (Johnson et al., 1985) involved the acidification of 20 ml aliquot of the sample by addition of phosphoric acid, purging of liberated CO_2 by the CO_2 -free air, removal of interfering nitrogen oxides by silver nitrate scrubber and titration of the liberated carbon dioxide in the coulometric cell. The reliability of the coulometric titration was regularly checked with calcium carbonate standards. Analytical precision was about $\pm 4 \mu\text{M}$ for TCO_2 and ± 0.001 for pH.

Air samples were collected in 1 l glass flasks from the bow of the ship half-an-hour before arrival at the stations. These samples were taken after thorough flushing of the flasks for 10 min using a vacuum pump. Atmospheric CO_2 measurements

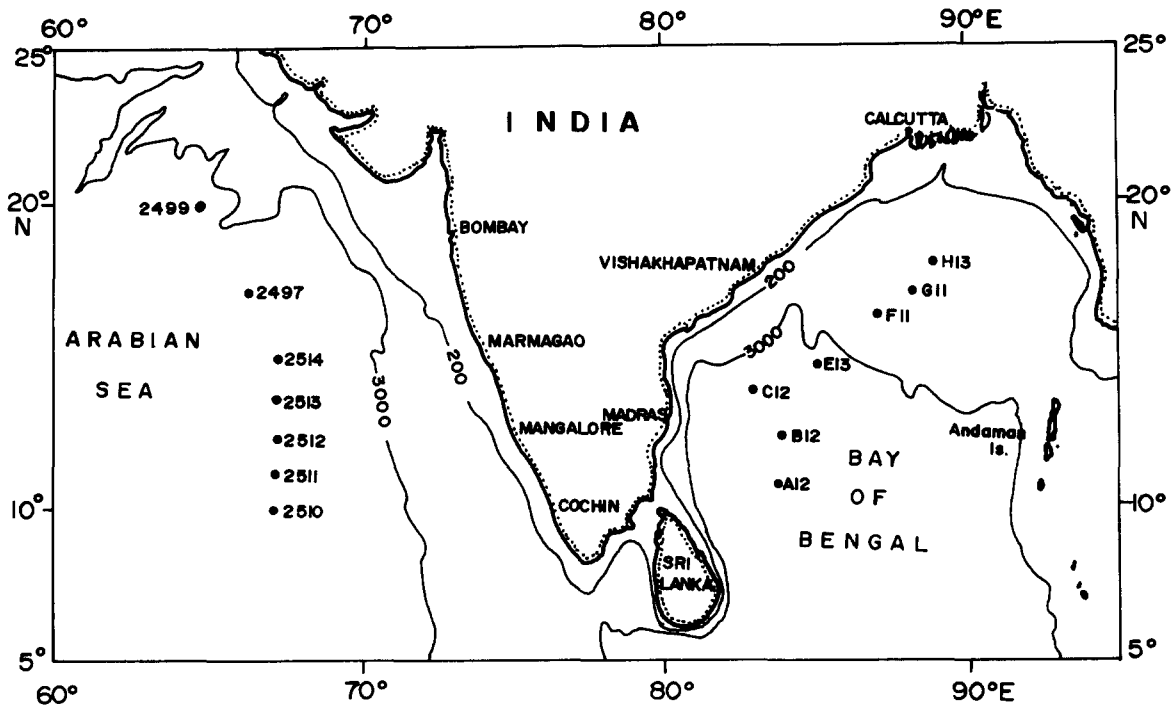


Fig. 1. Station locations.

were made by flushing the sample with the CO_2 -free air into the coulometric cell. As a result of the long analytical times (1 h) for the air samples, it was necessary to make corrections for the instrumental drift.

The colorimetric method followed by us yields pH values on the free hydrogen ion scale (pH_f). Since the most recent and reliable measurements of the carbonic acid dissociation constants are on the total scale (pH_T) it is desirable to convert the pH_f into pH_T . This was achieved by considering the interaction of H^+ with SO_4^{2-} , ignoring the effect of F^- (Dickson, 1993). For this purpose, Johansson and Wedborg's (1982) formulations for the dissociation constant of HSO_4^- and Millero's (1983) data on the partial molal volume and compressibility were utilized. The pCO_2 and CO_3^{2-} abundances in seawater were computed by using the coulometric TCO_2 and pH_T . The carbonic acid ionization constants obtained from the equations that are based on the data sets of Hanson (1973) and Goyet and Poisson (1989), as deduced and recommended by the latter workers, were used in these computations. Use of these con-

stants is further justified from the fact that the offset between TCO_2 calculated from the acid-titration technique and that of the coulometric analysis was found to be the smallest when these constants were considered (Stoll et al., 1993). The partial molal volume and compressibility values were obtained from Millero (1979). Oxygen saturation was calculated according to Benson and Krause (1984). The CO_2 transfer coefficients and fluxes were computed following Wanninkhof (1992) using the observed as well as the average climatological wind speeds.

3. Results and discussion

Variations in TCO_2 with depth and geographical location are shown in Fig. 2 for the Arabian Sea and in Fig. 3 for the Bay of Bengal. The TCO_2 concentrations at all depths exhibited steady increases towards the north in the Arabian Sea. For example, the concentration of TCO_2 at its maximum increased from $2330 \mu\text{M}$ at station 2510 to $2383 \mu\text{M}$ at station 2499. The N–S

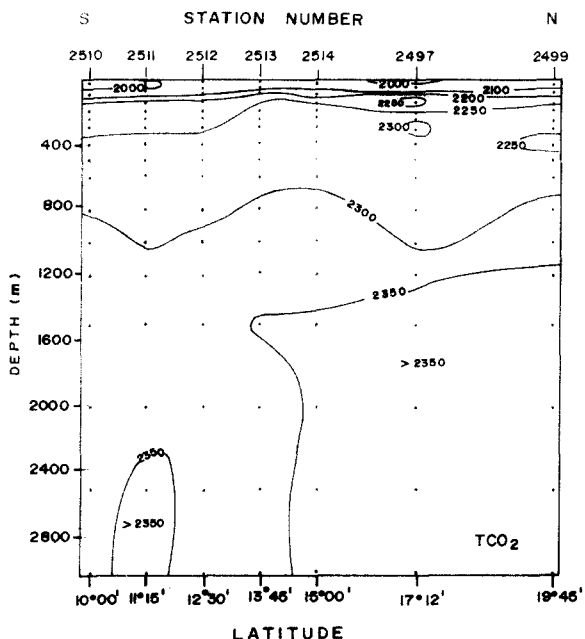


Fig. 2. N-S variations in TCO₂ (µM) in the Arabian Sea.

gradient observed in the Bay of Bengal was much smaller, the maximal difference between stations A-12 and G-11 being 31 µM. Significantly, the N-S gradient (decrease towards north) in dissolved

oxygen was also much more pronounced in the Arabian Sea than in the Bay of Bengal, as could be seen from a comparison of vertical sections presented in Figs. 4 and 5. A northward increase in TCO₂ within the surface layers was observed in the Arabian Sea (from 1985 µM, at station 2510, to 2041 µM, at station 2499) presumably due to the higher salinity as well as the supply from subsurface layers through upwelling and vertical mixing. Surface TCO₂ in the Bay of Bengal showed wide fluctuations, probably related to large salinity changes.

The observed northward increase in TCO₂ in subsurface waters of the Arabian Sea is in agreement with the previous results (Kumar et al., 1990, 1992). It is caused by the regeneration of CO₂ during the decomposition of organic matter as well as the solution of skeletal material. The northern Arabian Sea appears to provide favourable sites for the high rates of CO₂ production through both pathways. As stated above, the high new production not only involves downward exports of large quantities of particulate organic carbon from the euphotic zone, but the occurrence of denitrification may also facilitate the utilization of DOC by bacteria, leading to enhanced regeneration of

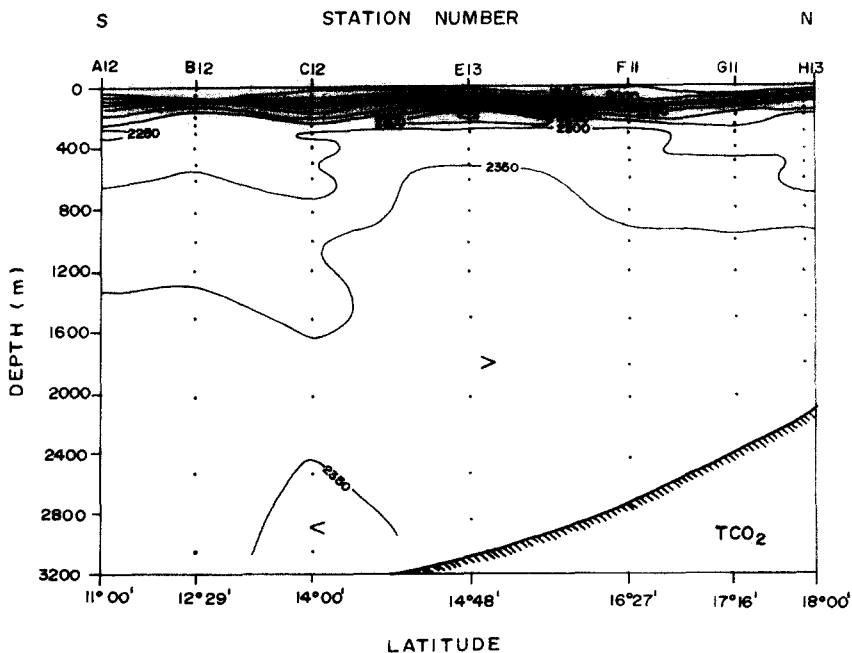


Fig. 3. N-S variations in TCO₂ (µM) in the Bay of Bengal.

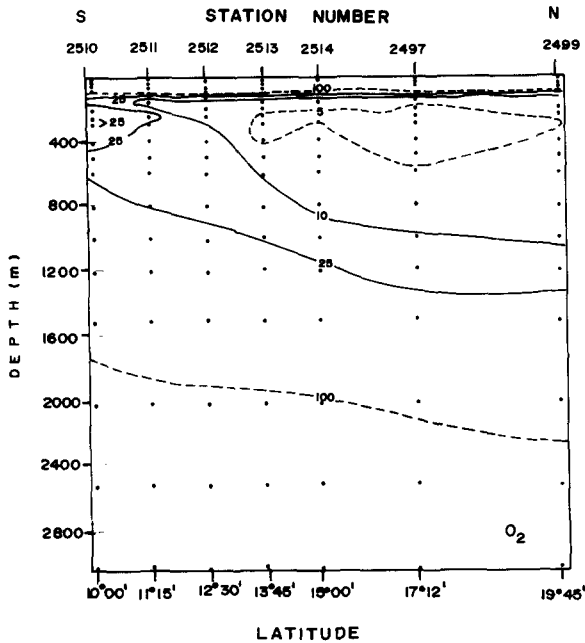


Fig. 4. N-S variations in oxygen (μM) in the Arabian Sea. For brevity only concentrations $< 100 \mu\text{M}$ are shown.

CO_2 . Moreover, as discussed below, a higher degree of CaCO_3 undersaturation in the northern Arabian Sea may facilitate greater dissolution of skeletal material as well.

The contribution from the oxidation of organic matter (OCO_2) to the observed TCO_2 increase from station 2510 to 2499 could be roughly quantified from the profiles of apparent oxygen utilization (AOU). For this purpose we have used a value of 0.75 for $\Delta\text{CO}_2/\Delta\text{AOU}$ obtained by Kroopnick (1985) for the world oceans. It may be noted that the uncertainty associated with this ratio corresponds to an error of 2–3 μM in our estimates of OCO_2 . Also, since nitrate is an additional oxidant in the Arabian Sea, the use of AOU alone leads to an underestimation of OCO_2 within the denitrifying layer ($\sim 200\text{--}500 \text{ m}$ at station 2499). However, these limitations do not negate the conclusions reached below.

As shown in Fig. 6a, OCO_2 is substantially higher at all depths in the subsurface layers at station 2499 than at 2510. The average increase below

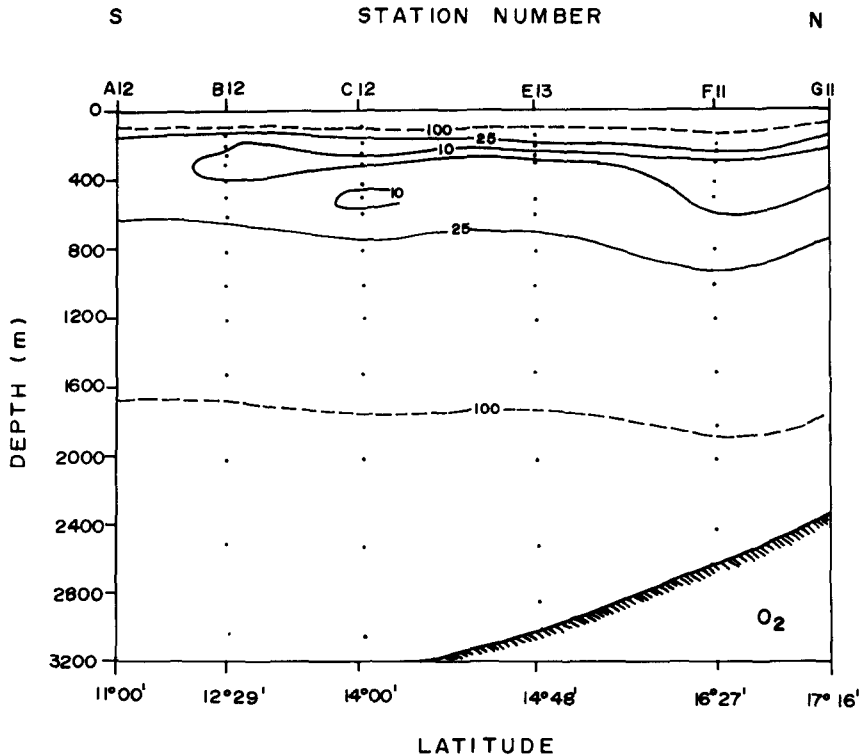


Fig. 5. N-S variations in oxygen (μM) in the Bay of Bengal. Contours as in Fig. 4.

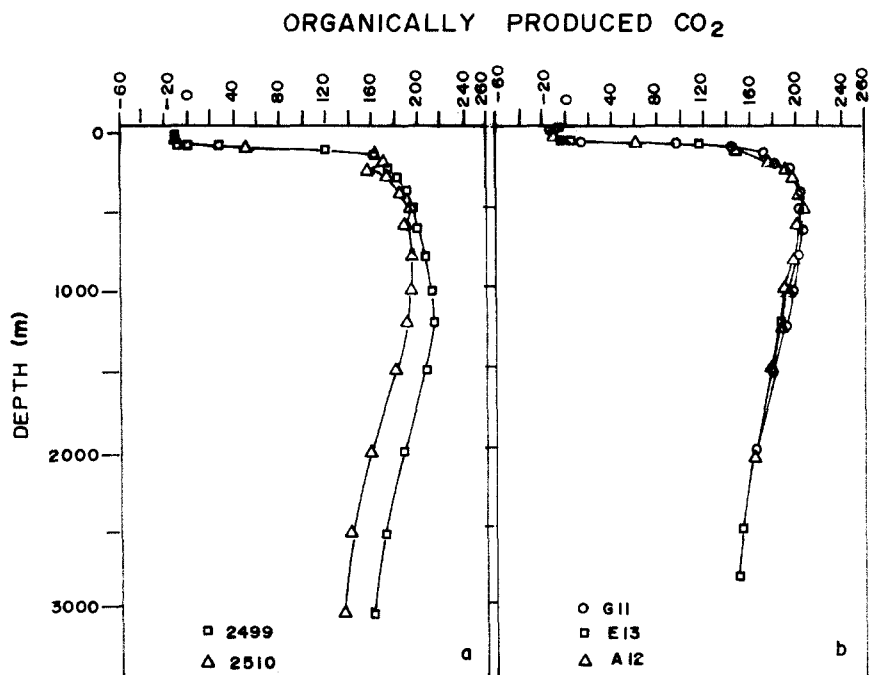


Fig. 6. Vertical profiles of CO₂ (μM) produced due to organic matter decomposition in the northern Indian Ocean.

1 km between the two stations was 24 μM. Thus, it appears that roughly half of the observed TCO₂ increase could be contributed by soft tissue decomposition, with the other half coming from CaCO₃ dissolution. In contrast, the organic carbon decom-

position adds surprisingly little to the TCO₂ pool in the deep northern Bay of Bengal. This is evident from Fig. 6b which compares data from three stations. The remarkably similar OCO₂ profiles, of course, directly reflect the weak N–S gradients

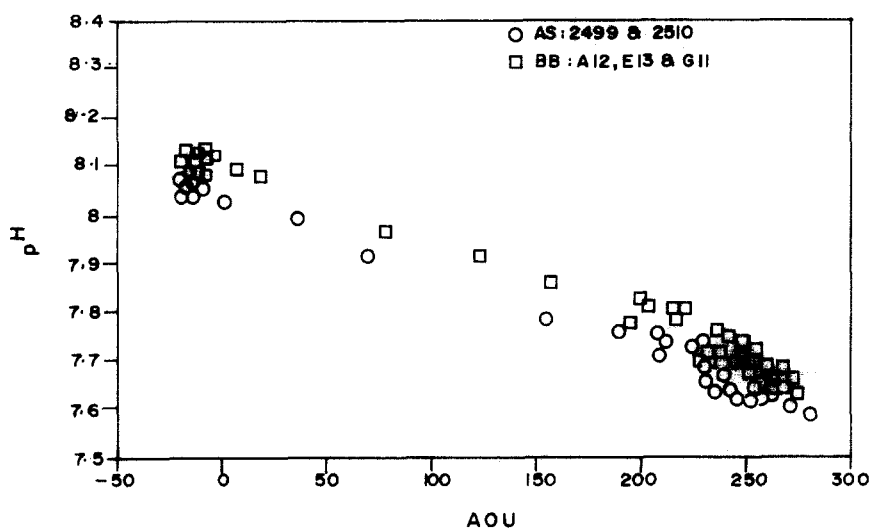


Fig. 7. Relation between apparent oxygen utilization (AOU, μM) and pH for some stations in the northern Indian Ocean.

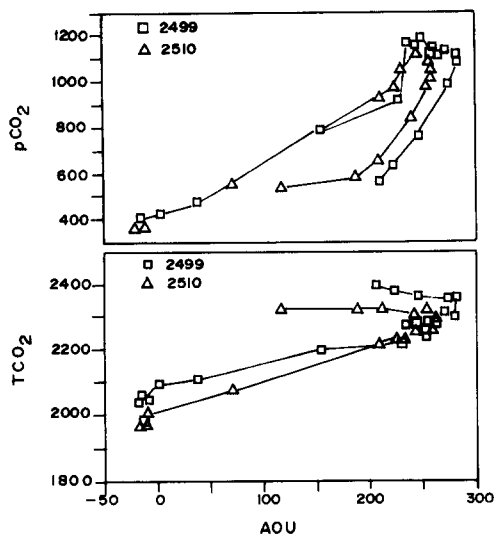


Fig. 8. Variations in TCO_2 (μM) and $p\text{CO}_2$ (μatm) with AOU (μM), respectively, in the Arabian Sea.

in oxygen (Fig. 5). The average increase in OCO_2 between the northernmost and southernmost stations ($2.5 \mu\text{M}$) is only 1/7 of the average TCO_2 increase ($17.5 \mu\text{M}$) at depths exceeding 1 km.

The vast difference in the extent of organic carbon decomposition in the deep waters of the Arabian Sea and the Bay of Bengal might look anomalous at first glance in view of the observations that the fluxes of organic matter associated with sinking particles, as measured by the sediment traps, are considerably higher in the Bay of Bengal ($2.04\text{--}3.59 \text{ g C/m}^2/\text{yr}$; Ittekkot et al., 1991) than in the Arabian Sea ($1.53\text{--}1.80 \text{ g C/m}^2/\text{yr}$; Nair et al., 1989) in spite of the much higher primary production in the latter region (Qasim, 1977). However, it has been proposed that the interaction of biogenic matter with lithogenic particles may play a key role in sedimentation (Ittekkot et al., 1992). In an area such as the Bay of Bengal, which receives enormous amounts of lithogenic material through river outflow, this may result in very rapid sinking of particulate organic carbon through the water column with relatively little decomposition. However, it should be noted that our results do not imply that the organic carbon settling through the water column in the Bay of Bengal is refractory. On the contrary, it appears to be quite "fresh", since it is readily oxidized at the seafloor

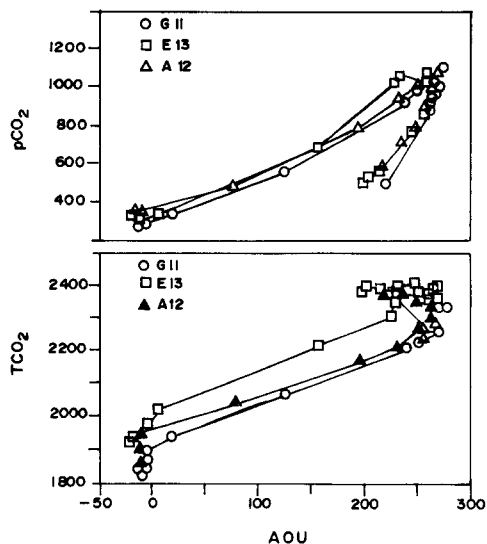


Fig. 9. Variations in TCO_2 (μM) and $p\text{CO}_2$ (μatm) with AOU (μM), respectively, in the Bay of Bengal.

producing near-bottom anomalies that are unique to the Bay of Bengal (Broecker et al., 1980). The extent of material that is decomposed at the sediment–water interface after reaching the seafloor is, however, not known. Nevertheless, given the dependence of the extent of such decomposition on the bulk sediment flux (Sirocko and Ittekkot, 1992), we speculate that the Bay of Bengal sediments may preserve a larger proportion of organic carbon originating in the surface layer than sediments in most other areas.

The shapes of the OCO_2 profiles in the Arabian Sea and the Bay of Bengal are somewhat different: in the Bay of Bengal, the OCO_2 maximum is sharply defined and is located at much shallower depths ($\sim 500 \text{ m}$). This is because the thickness of the intermediate layer, characterized by low oxygen content, is smaller in the Bay of Bengal. As shown by Rao et al. (1994-this issue) the discontinuity defining the transition from the intermediate to deep waters occurs at higher temperatures ($\sim 12^\circ\text{C}$) in the Bay of Bengal than in the Arabian Sea ($\sim 10^\circ\text{C}$).

The large extent of regeneration of CO_2 from soft tissue and the prevalence of reducing conditions are expected to result in a decrease in pH_T in the subsurface waters of the Arabian Sea. As seen in Fig. 7, this is indeed the case since the

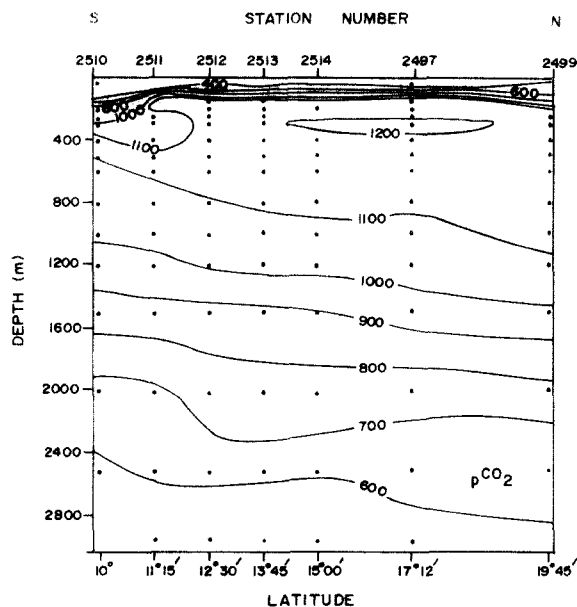


Fig. 10. The $p\text{CO}_2$ (μatm) variations along a N-S section in the Arabian Sea.

pH_T values are consistently lower in the Arabian Sea than those in the Bay of Bengal. Since the production of organic molecules removes protons from the solution (Anderson and Dyrssen, 1994-this issue), a decrease in pH should occur due to the decomposition of organic matter. As a result of this, a linear decrease in pH with an increase in AOU is observed in both areas.

Plots of $p\text{CO}_2$ and TCO_2 vs. AOU for selected stations in the Arabian Sea and Bay of Bengal are shown in Figs. 8 and 9, respectively. In the Arabian Sea, the $p\text{CO}_2$ reached peak values when the AOU was still rising (Fig. 8). This is because of the fact that due to the occurrence of denitrification the AOU does not provide a complete measure of the extent of organic decomposition in the Arabian Sea. Since the denitrifying layer (~ 150 – 500 m) is located substantially above the AOU maximum (~ 1200 m), the $p\text{CO}_2$ maximum (Fig. 10), which is found to coincide with the former feature, does not coincide with the AOU maximum. The $p\text{CO}_2$ gradient observed between stations 2510 and 2499 was quite small at depths deeper than the AOU

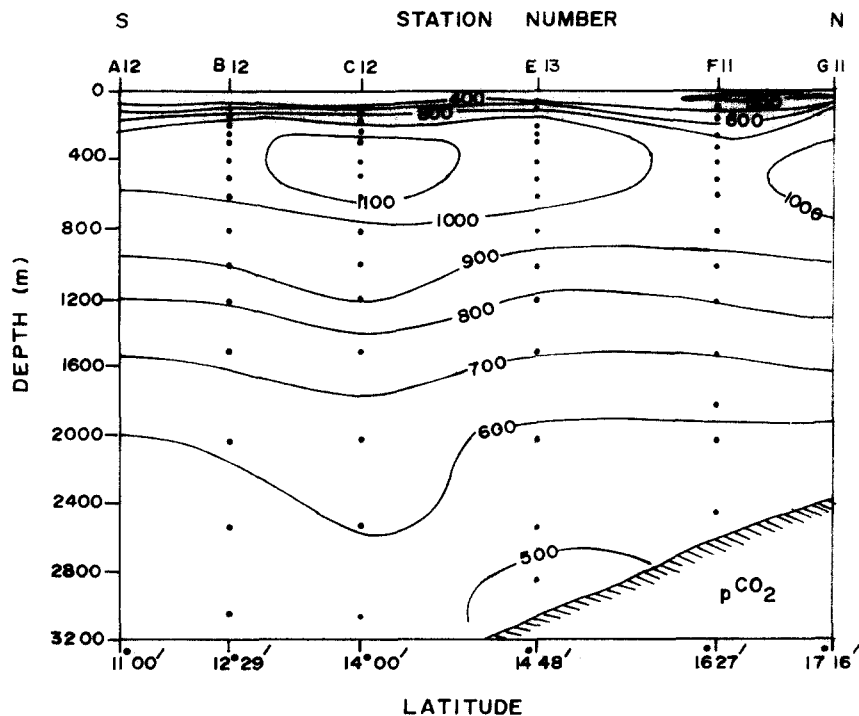


Fig. 11. The $p\text{CO}_2$ (μatm) variations along a N-S section in the Bay of Bengal.

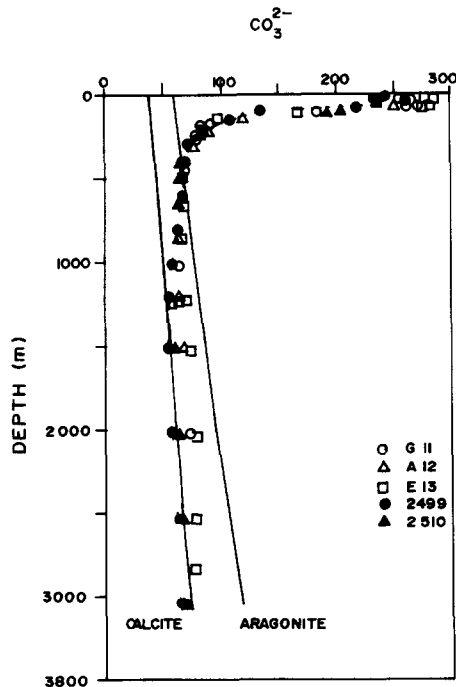


Fig. 12. Vertical variations in the carbonate ion (μM) in the northern Indian Ocean.

maximum, but at shallower depths consistently higher values were observed in the north. The thickening of the oxygen minimum layer towards the north is reflected by the broad $p\text{CO}_2$ maximum at station 2499. While the $p\text{CO}_2$ decreased gradually with depth, TCO_2 remained more or less constant below the AOU maximum (Fig. 8). In the Bay of Bengal, the distribution of $p\text{CO}_2$ was somewhat different in that the peak values were associated with the AOU maxima (Fig. 9) and were observed in regions of the oxygen minima (see Figs. 5 and 11). The N–S gradients in $p\text{CO}_2$ do not seem to be significant except for high values at station C-12. In general, the $p\text{CO}_2$ levels were substantially higher in the Arabian Sea than those in the Bay of Bengal, as a result of the more intense oxidation of organic matter and the consequently lower pH_T .

A very significant observation made by us is that the TCO_2 concentrations in the deep Bay of Bengal are somewhat higher than those prevailing in the Arabian Sea. We are satisfied that this difference does not result from an offset between the two sets of data as the data were generated following the

same technique. Moreover, our results agree quite well with those obtained during the GEOSECS cruises (e.g. at station 2499, which had the same location as GEOSECS station 416). In view of the minor decomposition of organic matter as well as consistently higher oxygen concentrations in the Bay of Bengal, the excess CO_2 should arise from skeletal dissolution.

In order to evaluate the regional variability in the extent of CaCO_3 dissolution, vertical profiles of carbonate ion concentrations at several stations are compared in Fig. 12. Saturation carbonate ion concentrations, estimated following the equations of Mucci (1983), for the effects of temperature and salinity, and of Millero (1979), for the influence of pressure on the solubility products of aragonite and calcite, are plotted as a function of depth. As the regional changes in temperature and salinity at a given depth are not very large in deep water, within the area of our study, only average profiles are presented. As noted previously (Naqvi and Reddy, 1975; Naqvi and Naik, 1983; Kumar et al., 1992), the surface waters of the northern Indian Ocean are supersaturated with respect to both calcite and aragonite. The change-over from supersaturation to undersaturation occurs at ~ 500 m for aragonite. However, the deep water seems to be generally in near-equilibrium with calcite.

Considerable regional variability of the degree of undersaturation of deep waters is evident in Fig. 12. In general, the deep northern Arabian Sea is the most corrosive to CaCO_3 while the deep Bay of Bengal is the least. The northern Arabian Sea, where the calcite saturation depth lies at ~ 1500 m, is the only region in the northern Indian Ocean that is undersaturated with respect to this mineral. Based on the observed changes in alkalinity, Anderson and Dyrssen (1994-this issue) found the dissolution of CaCO_3 to be significant below 600 m in the Arabian Sea.

The higher saturation levels coupled with very similar carbonate fluxes [$10.72\text{--}16.08$ $\text{g}/\text{m}^2/\text{yr}$ in the Bay of Bengal (Ittekkot et al., 1991) as compared to $11.88\text{--}19.00$ $\text{g}/\text{m}^2/\text{yr}$ in the Arabian Sea (Nair et al., 1989)] are inconsistent with an enhanced dissolution of CaCO_3 within the Bay of Bengal as compared to the Arabian Sea. These are, however, compatible with our previous

Table 1
Flux ($\text{mmol/m}^2/\text{d}$) and amounts (Tg/yr) of CO_2 evasion to atmosphere from the northern Indian Ocean

Region	Based on observed winds Observed air CO_2		Based on climatological winds ^d Assumed average air CO_2	
	Flux	Amount	Flux	Amount
Arabian Sea	2.90	79	2.52	69
Bay of Bengal	0.33	6	0.66	12
North Indian Ocean ^b	1.381 ^c	62	1.09 ^c	50

^aAn annually-averaged value of 5 m/s is used for the northern Indian Ocean (Naqvi et al., 1993) together with an average atmospheric CO_2 concentration of 342 ppmv.

^bFluxes from both the Arabian Sea and the Bay of Bengal (total area of $10.312 \times 10^6 \text{ km}^2$).

^cAverage of all observations in the Arabian Sea and the Bay of Bengal.

calculations showing that skeletal dissolution in the Bay of Bengal is somewhat smaller than in the Arabian Sea. In view of the above discussion, it is difficult to escape the conclusion that, in order to account for the higher TCO_2 levels, enhanced dissolution of CaCO_3 might have occurred in deep waters en route the Bay of Bengal rather than within the region. Considering the fact that the deep waters of the Arabian Sea and Bay of Bengal are fed by different western-boundary currents which flow through different terrains (Warren, 1981), differential CaCO_3 dissolution in these waters may be plausible.

3.1 Atmospheric fluxes

As stated earlier, the northwestern Indian Ocean seems to eject large amounts of CO_2 into the atmosphere. However, previous studies have relied on sparse geographical coverage, particularly in the Bay of Bengal. Our results provide additional data which further reinforce this view and help refine estimates of atmospheric fluxes from this region.

The differences in mean tropospheric CO_2 concentrations observed over the Arabian Sea and Bay of Bengal were virtually indistinguishable (330 ppm). On average, the $p\text{CO}_2$ in water was substantially lower in the Bay of Bengal ($352 \mu\text{atm}$) than in the Arabian Sea ($383 \mu\text{atm}$) although the highest value of $476 \mu\text{atm}$ was observed in the Bay of Bengal at station B-2 ($12^\circ 42.84' \text{N}$, $80^\circ 28.58' \text{E}$, not shown in Fig. 1). Moreover, during the periods

when our observations were made, the winds were stronger in the Arabian Sea (9.3–31.5 km/h) than in the Bay of Bengal (0.4–13 km/h). This resulted in a higher rate of CO_2 evasion to the atmosphere from the Arabian Sea (Table 1). The atmospheric fluxes varied from -0.007 to $8.67 \text{ mmol/m}^2/\text{d}$ in the Arabian Sea and from 0.0001 to $0.98 \text{ mmol/m}^2/\text{d}$ in the Bay of Bengal. Using our average flux values, the overall fluxes were computed for the Arabian Sea (area $6.225 \times 10^6 \text{ km}^2$) and Bay of Bengal (area $4.087 \times 10^6 \text{ km}^2$) as 79 and 6 Tg C/yr ($1 \text{ Tg} = 10^{12} \text{ g}$), respectively. The large difference in CO_2 emissions from the two regions is due to a combination of lower $p\text{CO}_2$ in surface waters of the Bay of Bengal, and stronger winds and a larger area in the Arabian Sea. However, if we assume an annual average wind speed of 5 m/s over the northern Indian Ocean (close to the climatological mean; Naqvi et al., 1993) and an average $\Delta p\text{CO}_2$ of 18 ppmv the average flux amounts to $1.09 \text{ mmol/m}^2/\text{d}$ that corresponds to an overall evasion rate of 50 Tg/yr from the northern Indian Ocean. The presently estimated fluxes are comparable with those given by Takahashi (1989) and Somasundar et al. (1990). Thus, our results confirm the previous suggestions that the northern Indian Ocean serves as a moderately strong source of atmospheric CO_2 . It must, however, be stated that these computations are based on observations made during the premonsoon season. Hence, these estimates may be conservative since the exchange coefficients of carbon dioxide are seven times higher during the southwest monsoon as compared to the other seasons (Etcheto et al., 1991).

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