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Denitrification processes in the Arabian Sea

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Abstract. Recent information on some consequences of the acute mid-water oxygen deficiency in the Arabian Sea, especially on carbon-nitrogen cycling, is reviewed. An evaluation of published estimates of water column denitrification rate suggests an overall rate in the vicinity of 30 Tg Ny^{-1}, but the extent of benthic contribution remains unknown. A decoupling of denitrification from primary production, unique to the Arabian Sea, is revealed by nitrite, electron transport system (ETS) activity and bacterial production data. Results of both enzymatic and microbiological investigations strongly point to a major role of organic carbon other than that sinking from surface layers in supporting denitrification. Although denitrification is associated with an intermediate nepheloid layer, it seems unlikely that the excess carbon comes with particles re-suspended along the continental margins and transported quasi-horizontally into the ocean interior; instead, the particle maximum may directly reflect a higher bacterial abundance. It is proposed that denitrification may be predominantly fuelled by the dissolved organic matter.

Keywords. Nitrogen system; denitrification; organic matter; Arabian Sea.

1. Introduction

While in most parts of the open ocean the seawater is well oxygenated at all depths, severe depletion of oxygen occurs at mid-depths in some areas. Typically, such waters underlie tropical zones of high biological productivity associated with upwelling, where the demand of oxygen in subsurface waters is high and a strong upper thermocline limits its supply from the surface layer. Thus, oxygen concentrations are often below detection limit of the Winkler procedure within large bodies of intermediate waters on both sides of the equator in the eastern tropical Pacific (Reid 1965) and in the northern Indian Ocean (Wyrtki 1971).

The transition from the 'oxic' to 'anoxic' conditions involves several important biogeochemical changes (Richards 1965). The first is that the facultative bacteria switch over to nitrate ions which are the next most abundant source of free energy for oxidation of organic matter. Thus, nitrate is reduced to molecular nitrogen with nitrite, which accumulates in the water column, as one of several intermediates; this process, known as denitrification is a major component of the nitrogen cycle (figure 1). Denitrification has vital geochemical significance since free nitrogen refluxed to the atmosphere makes up for the inputs of combined nitrogen to the sea from the atmosphere and by the rivers (Emery *et al* 1955; Codispoti and Christensen 1985). After a complete, or almost complete, removal of nitrate and nitrite from water, sulphate ions serve as the next preferred reducible substrate leading to the production of hydrogen sulphide. However, this stage, representing true anoxia, is rarely reached in the open oceanic waters, since the supply of nitrate is generally sufficient to meet the bacterial demand. The environments affected by denitrification but not experiencing

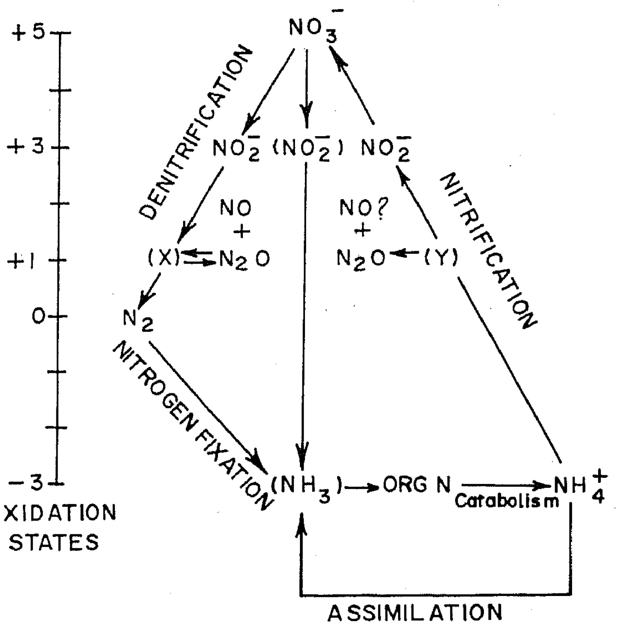

Figure 1. The marine nitrogen cycle. 'X' and 'Y' are intra-cellular intermediates that do not accumulate in water column (from Codispoti and Christensen 1985).

sulphate reduction are generally termed as 'suboxic'. The disappearance of oxygen in seawater also leads to a lowering of the redox potential. This affects the oxidation states of several metals. For example, iron is reduced from +3 to +2 under suboxic and anoxic conditions.

The Arabian Sea has long been known to experience an intense depletion of dissolved oxygen at intermediate depths. However, oxygen data from this region have been acquired almost entirely following the Winkler procedure. At extremely low oxygen concentrations, blank levels become a problem with Winkler titrations lowering precision and accuracy of the oxygen data. The limited measurements made with the more reliable colorimetric procedure of Broenkow and Cline (1969) have shown that oxygen concentrations (figure 2) are often below the threshold values (1·2–3·85 μM) required for the onset of denitrification (Devol 1978). The widespread 'secondary' nitrite maximum within the core of the oxygen minimum (in addition to the global 'primary' nitrite maximum at the base of the euphotic zone) is believed to arise from dissimilatory nitrate reduction. However, it was only in the mid 1970s that studies were initiated to quantify the extent of denitrification in the Arabian Sea (Sen Gupta *et al* 1976a, b; Deuser *et al* 1978). These authors demonstrated that the combined nitrogen (nitrate + nitrite) concentrations within the secondary nitrite maximum (SNM) were significantly lower than those expected from the nitrate-oxygen relationships in nitrite-free waters. A number of reports have since appeared in the literature dealing with various aspects of water-column denitrification in the Arabian Sea (Sen Gupta *et al* 1980; Naqvi *et al* 1982, 1990, 1993; Naqvi and Qasim 1983; Naqvi and Sen Gupta 1985; Naqvi 1986, 1987, 1991; Naqvi and Shailaja 1993; Noronha 1992; Mantoura *et al* 1993). Earlier work has been reviewed by Sen Gupta and Naqvi (1984) and Naqvi *et al* (1992); a synthesis of the more recent results is attempted here.

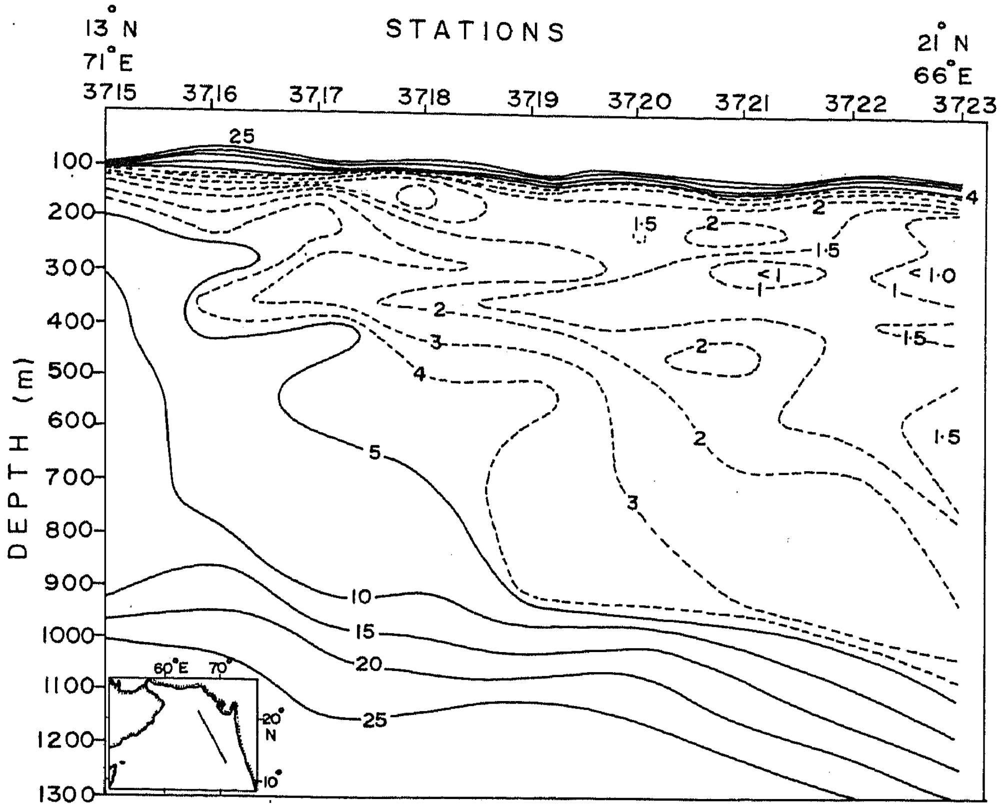

Figure 2. Colorimetrically-determined oxygen levels (μM) within the oxygen minimum along a transect parallel to the Indian coast (inset) (from Naqvi 1987).

2. Geographical boundaries of water-column denitrification

Geographical limits of the Arabian Sea denitrification zone have recently been delineated by Naqvi (1991) from an analysis of nitrite distribution using historical data (figure 3). Operationally bounding the denitrification zone by the 0·2 μM nitrite contour, the total area of the Arabian Sea affected by water-column denitrification was computed as $1{\cdot}37 \times 10^6 \mathrm{km}^2$, comparable to each of the two Pacific denitrification sites. The most intriguing observation is that, unlike the eastern tropical Pacific, zones of the most intense denitrification (maximum nitrite) and the highest primary productivity are geographically separated in the Arabian Sea. As expected, the most fertile waters are those which are upwelled seasonally (during the southwest monsoon) off Arabia and Somalia in the western Arabian Sea. This is evident from the maps of primary productivity (Qasim 1977; Naqvi 1991) and of sea-surface pigment concentrations observed from the CZCS satellite (Codispoti 1991; Brock and McClain 1992). These two centres of intense upwelling are clearly located outside the denitrification zone, as is the centre of relatively weaker upwelling off the southwest Indian coast (figure 3). In contrast, the high-nitrite ridge, which represents the locus of the most intense denitrification, extends from the continental margin off Gujarat well into the relatively oligotrophic central Arabian Sea. This unique decoupling of subsurface denitrification from primary productivity of the overlying surface waters could arise from a higher supply of oxygen to intermediate waters beneath the

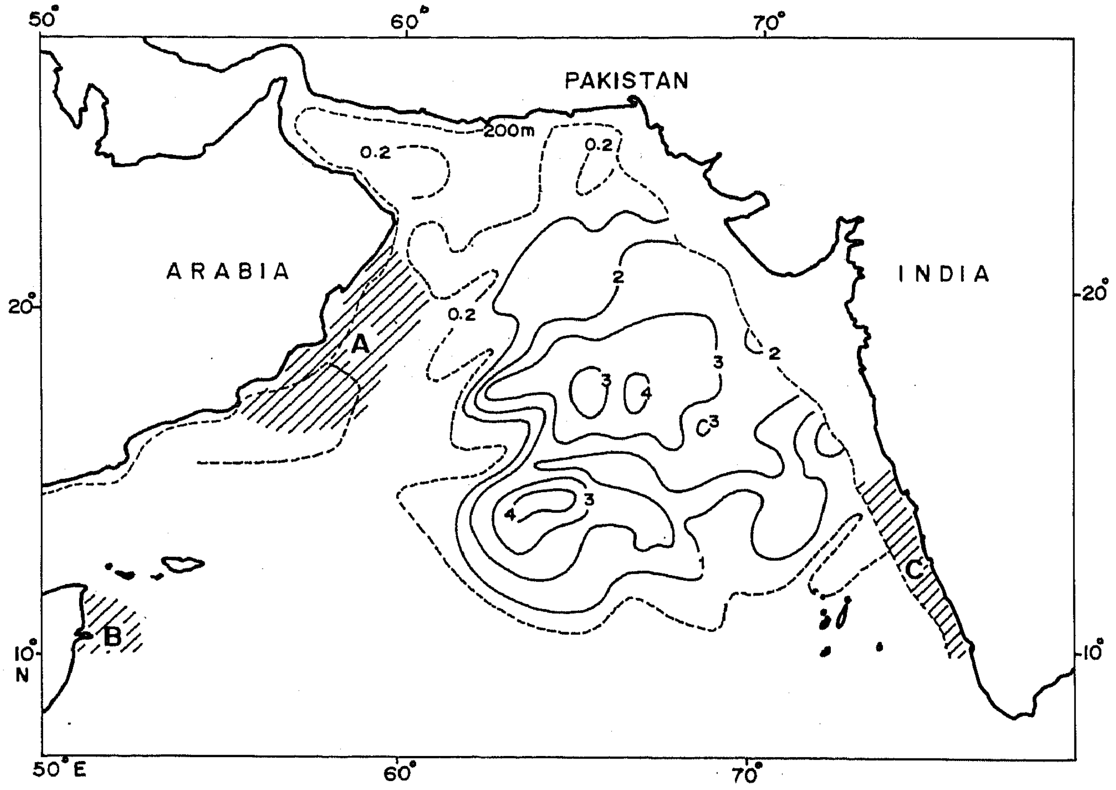

Figure 3. Distribution of nitrite (μM) within the oxygen minimum averaged for each 1-degree square using all the available data (maximum values at each station). The hatched areas represent zones of upwelling off Arabia (A), Somalia (B) and southwest India (C). Extensions of zones B and C south of Lat. 10°N are not shown due to absence of the secondary nitrite maximum (from Naqvi 1991).

productive zones, and/or a different, possibly additional, mode of input of biodegradable organic matter to subsurface layers in the central Arabian Sea.

Naqvi (1991) pointed out that the western and northern parts of the Arabian Sea are much more weakly stratified than the central and eastern regions. Proximity of land mass causes convective overturning of surface waters at rather low latitudes off the Pakistani coast during the winter (Banse 1984). The prevailing anti-clockwise circulation at the sea surface helps in maintaining lower sea surface temperatures in the western Arabian Sea. In the eastern Arabian Sea, on the other hand, the advection of fresher and warmer equatorial surface waters during the northeast monsoon leads to greater stability and presumably a smaller oxygen flux from the surface to subsurface waters. The outflows from the Persian Gulf and the Red Sea also appear to provide some oxygen input to the oxygen-minimum zone in the western Arabian Sea (Olson *et al* 1993)*. In addition, a more vigorous advection is expected to suppress

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*Due to their low preformed oxygen contents (reduced solubility at elevated temperatures and salinities), the outflows from the two marginal seas may favour the development of the oxygen-deficient conditions in the Arabian Sea *as a whole* (Swallow 1984; Olson *et al* 1993). However, *within the Arabian Sea*, even small inputs of oxygen can dramatically affect the delicately poised redox transformations within the nitrogen system.

denitrification along the continental margins, although its importance cannot be evaluated from the oxygen distribution due to scarcity of the colorimetric data.

3. Extent of denitrification

Numerous attempts have been made to quantitatively evaluate the extent of denitrification in the Arabian Sea (Sen Gupta *et al* 1976a, b, 1980; Deuser *et al* 1978; Naqvi *et al* 1982, 1990, 1993; Naqvi and Qasim 1983; Naqvi 1986, 1987, 1991; Naqvi and Shailaja 1993; Mantoura *et al* 1993). All but two studies (Naqvi and Shailaja 1993; Naqvi *et al* 1993) involved the estimation of nitrate deficits (i.e., the difference between the nitrate concentrations expected if there were no denitrification and the sum of the observed nitrate and nitrite concentrations). Deuser *et al* (1978) estimated the expected nitrate levels from a regression of nitrate and salinity. This approach was based on the premise that denitrification was confined to the Persian Gulf water (PGW). The PGW, having a shallow origin, is nutrient depleted, and as this watermass mixes with the Arabian Sea intermediate water, nitrate is added to it while salt is lost in an apparently linear manner. However, this procedure was found to yield low deficits by the subsequent workers (Naqvi *et al* 1982; Naqvi and Qasim 1983), who also demonstrated that denitrification was not confined to the PGW alone. All other authors have exploited the well-known statistical constancy of the relative changes in oxygen, carbon, nitrogen and phosphorus during the biological uptake and regeneration processes (Redfield *et al* 1963) for computing the expected nitrate. These ratios were first estimated for the Arabian Sea by Sen Gupta *et al* (1976a, b) from the regressions of the apparent oxygen utilization (AOU), nitrate and phosphate. Following a procedure previously employed by Cline and Richards (1972) in the eastern tropical North Pacific, Sen Gupta *et al* (1976a, b) estimated the expected nitrate as a sum of regenerated and preformed fractions. While the former was computed from the AOU and nitrate oxidative ratio, the latter was obtained from the regression between preformed nitrate and preformed phosphate. The same approach was also adopted by Naqvi *et al* (1982) and Naqvi and Qasim (1983) with minor modifications. However, the use of phosphate data introduces some uncertainty in this method. In order to eliminate this, Naqvi and Sen Gupta (1985) proposed the use of the nitrate tracer 'NO', first introduced by Broecker (1974) to delineate water masses with different preformed nitrate concentrations. NO was found to show excellent linear relationship with potential temperature (θ) in waters which were not affected by denitrification, and the NO-θ regressions were utilized to computed expected NO and hence nitrate within the denitrifying layer. This procedure was used by Naqvi (1986, 1987) for computing nitrate deficits using a very large data base. The NO-θ relations have since been refined by incorporating recent high quality data (Naqvi *et al* 1990), leading to deficits that are on an average about $1{\cdot}5\,\mu$M higher than those obtained by the procedure of Naqvi and Sen Gupta (1985).

A typical plot of NO versus θ in the northeastern Arabian Sea is shown in figure 4. Nitrate deficiency within intermediate waters is reflected by a pronounced minimum in NO between approximately 14° and 18°C (coinciding with the SNM). Linear correlations between NO and θ are obvious outside the denitrifying waters, albeit within two ranges with an apparent discontinuity at 10·4°C. Similar discontinuities are also seen in several other property–property relationships in the Arabian Sea (at

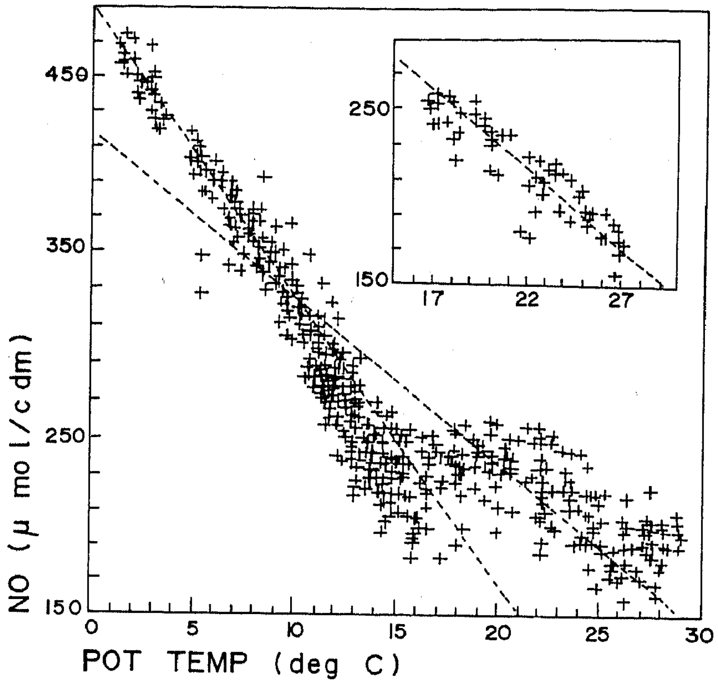

Figure 4. Plot of 'NO' vs. potential temperature for the eastern Arabian Sea (between 12° and 17°N latitude, east of 67°E longitude). The straight lines give the least squares fits to the data corresponding to $\theta \leqslant 9°C$ and $17 \leqslant \theta \leqslant 27°C$; for the latter range (inset), samples from only outside the denitrification zone were considered (from Naqvi *et al* 1990).

~ 10°C – Somasundar *et al* 1987) as well as in the Bay of Bengal (at ~ 12°C–Rao *et al* 1994); these probably represent the transition from the intermediate to deep waters. The NO–θ regressions for the two segments are given by the equations (Naqvi *et al* 1990):

$$\mathrm{NO} = 415{\cdot}934 - 8{\cdot}974\,\theta \qquad 10{\cdot}39 \leqslant \theta \leqslant 27°\mathrm{C} \tag{1}$$

$$\mathrm{NO} = 490{\cdot}254 - 16{\cdot}126\,\theta \qquad \theta < 10{\cdot}39°\mathrm{C} \tag{2}$$

from which the nitrate deficit (ΔN) could be computed using:

$$\Delta \mathrm{N} = (\mathrm{NO} - \mathrm{O}_2)/9{\cdot}1 - \mathrm{NO}_3 - \mathrm{NO}_2 \tag{3}$$

The above simple vertically-integrated treatment involves numerous water masses with potentially different preformed nitrate concentrations. For example, the outflow of low-nitrate water from the Persian Gulf produces a minimum in the depth profiles of nitrate in the Gulf of Oman (Mantoura *et al* 1993). In contrast, the waters advecting northward at this density level are nutrient-rich. In order to assess potential errors resulting from lateral mixing of these waters and to verify the applicability of the vertical approach, two attempts have been made involving binary mixing along 26·6 σ_θ surface (i.e. the density level of PGW). In both the studies (Naqvi *et al* 1990; Mantoura *et al* 1993), the northern end-member was located within or close to the Gulf of Oman, while the southern end-member was located in the southern Arabian Sea.

As shown graphically in figure 5, the conservative mixing line of Mantoura *et al* (1993) yields expected NO values that are statistically indistinguishable from those

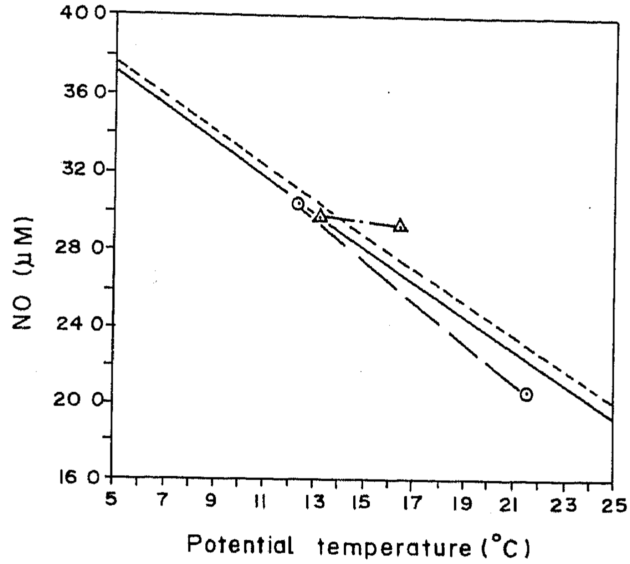

Figure 5. A comparison of NO–θ relationships observed in the northern Indian Ocean. Solid and short-dashed lines represent the vertically-integrated relationships in the Bay of Bengal (Rao *et al* 1994) and in the Arabian Sea (Naqvi *et al* 1990), respectively. Lines joining the triangles and circles are based on two-end member isopycnal treatments along $\sigma_\theta = 26{\cdot}6$ by Naqvi *et al* (1990) and Mantoura *et al* (1993), respectively.

obtained with equation (1) within the depth range of denitrification. For the near surface samples ($\theta > 20°$C), however, the former gives anomalously low NO and hence highly negative nitrate deficits. Of the two end-members used by Naqvi *et al* (1990) for their isopycnal mixing model, the southern end-member falls in line with the other results. The northern end-member, however, differs significantly presumably due to an overestimation of preformed nitrate. Significantly, the NO–θ relationship observed in the Bay of Bengal at $\theta > 12°$C (Rao *et al* 1994) is virtually the same as equation (1). And since the Bay of Bengal is not a water column denitrification site (Rao *et al* 1994), the agreement validates the relationships between NO and θ observed in the Arabian Sea giving more confidence to the estimates of nitrate losses.

Depthwise distribution of nitrate deficits along a meridional transect at 67°E, which samples both the oxic and suboxic waters, is reproduced in figure 6. A comparison with nitrite distribution along the same transect (figure 7) reveals that while the maxima in nitrite and nitrate deficit occur at about the same depth, the two do not coincide laterally: the highest nitrite concentrations occur towards the south far from the maximum nitrate deficits observed near the northern end of the section. This is because of the fact that while nitrite concentration provides a measure of current denitrification, nitrate deficit is also affected by the history of denitrification in the watermass. The waters in the northern Arabian Sea appear to accumulate larger nitrate deficits due to longer residence times eventhough the denitrification rate may be lower as evident by the observed low nitrite concentrations in the northernmost parts (figure 3).

In addition to the main denitrifying layer seen in figures 6 and 7, a deeper (700–1200 m) denitrifying layer may also develop occasionally, probably due to the advection of bottom nepheloid layers from the continental margin (Naqvi 1987; Naqvi *et al* 1993).

Inventories of nitrate losses resulting from denitrification have been made by several

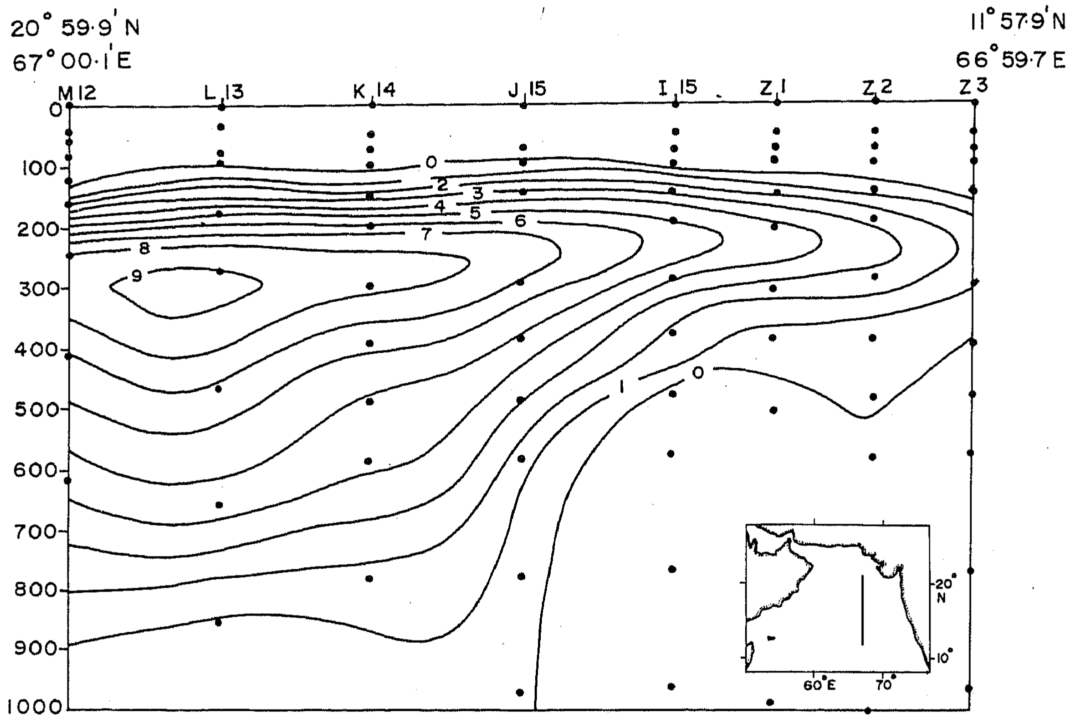

Figure 6. A meridional section (along 67°E, inset) showing the distribution of nitrate deficits (μM) (from Naqvi and Noronha 1991).

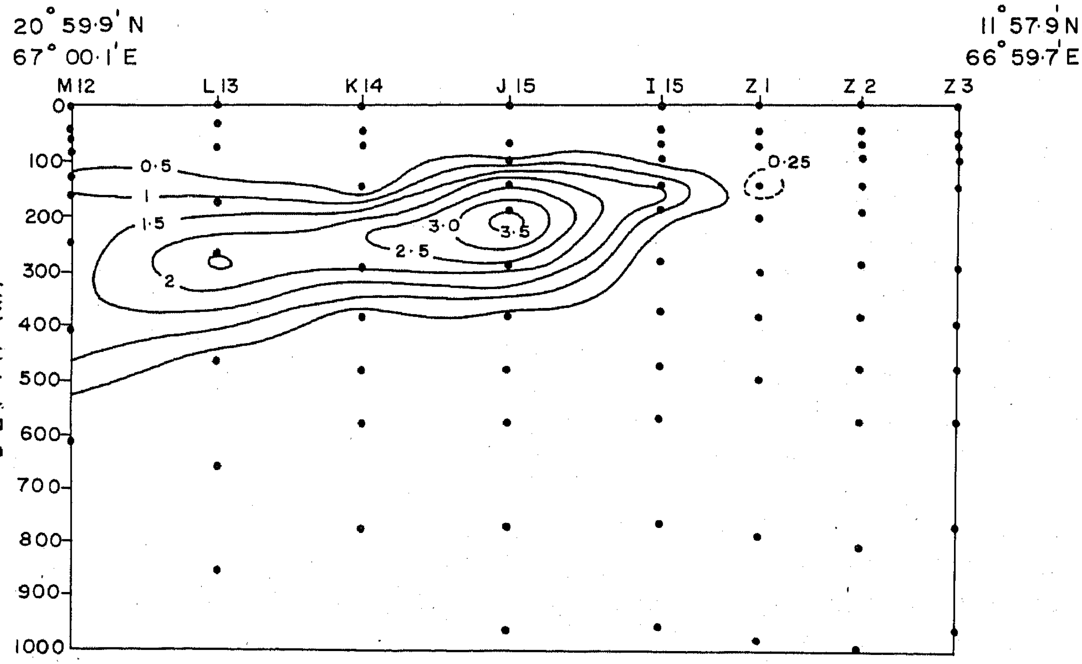

Figure 7. Distribution of nitrite (μM) along the same transect as in figure 6 (from Naqvi and Noronha 1991).

workers from the vertical integration of profiles of nitrate deficits (Deuser *et al* 1978; Naqvi *et al* 1982; Naqvi and Sen Gupta 1985; Naqvi 1987; Mantoura *et al* 1993). All these estimates are in close agreement with each other (95–118 Tg N y^{-1}) except that of Deuser *et al* (1978); the latter is anomalously low (4 Tg N y^{-1}) reflecting rather

Table 1. Denitrification rate in the Arabian Sea based on the advective and diffusive losses of nitrate deficits from the denitrifying zone (from Naqvi 1987).

Nature of losses	Rate (10^{12} gN y^{-1})
Horizontal advection	9·5
Vertical advection	1·0
Diffusion through the southern boundary	16·0
Diffusion through the upper boundary	1·5
Diffusion through the lower boundary	1·5
Total (rate of denitrification)	29·5

low deficits obtained by their procedure. However, the denitrification rates derived from these inventories differ widely (from 0·1 TgN y^{-1} by Deuser *et al* to 11·9 TgN y^{-1} by Mantoura *et al*) as a consequence of the various choices of the renewal times (3–30 years) of denitrifying waters.

Using all the data then available, Naqvi (1986, 1987) used a box model involving the advective and diffusive exports of nitrate deficits from the denitrification zone (table 1) to arrive at a rate of denitrification of ~ 30 Tg N y^{-1}.

Naqvi and Shailaja (1993) recently estimated rate of denitrification from activity of the respiratory electron transport system (ETS). In this method, activity of the enzyme systems that control respiration is measured by extracting the enzymes from microplankton retained on GF/F filters and incubating with an electron acceptor and with the natural substrates of the enzyme system. The ETS activities, however, give potential rates that must be converted into *in situ* rates with the aid of suitable constants. This approach is based on the assumption that ETS activity in the oxic and denitrifying waters is proportional to oxygen consumption and denitrification rates respectively. Results of batch culture experiments utilizing bacteria under both oxic and anoxic conditions have, in fact, revealed linear relationships between the ETS activity and respiration rates (Christensen *et al* 1980; Packard *et al* 1983). Although, the conversion of *in vitro* activity of the enzymes to *in vivo* respiration rate suffers from considerable uncertainty, the ETS-based denitrification rates appear to agreé well with those estimated by other methods (Codispoti and Richards 1976). Naqvi and Shailaja (1993) used a coefficient of 2·7–3·68 to convert the ETS activity (μeq l^{-1} h^{-1}) to denitrification rate (gN m^{-3} y^{-1}) on the basis of the published results of calibration of ETS assays (Devol 1975; Garfield *et al* 1983; Packard *et al* 1983).

Close to the boundaries of the oxic-suboxic zone, much of the nitrite produced through dissimilatory nitrate reduction might be re-oxidized to nitrate (Anderson *et al* 1982). Thus, the coupling of nitrification and denitrification could result in a recycling of combined nitrogen compounds without the terminal production of molecular nitrogen. In order to account for this effect, the ETS-derived denitrification rates were integrated only over the depth range with nitrite concentrations exceeding 0·5 μM (Naqvi and Shailaja 1993). As argued by Codispoti and Christensen (1985) the omission of data with $NO_2 < 0·5\,\mu$M most likely eliminates the possible errors due to nitrite recycling in the ETS-based denitrification rate estimates. The mean rate so obtained (20·8–28·35 gN m^{-2} y^{-1}) applied over the area with $NO_2 > 0·5\,\mu$M ($1·15 \times 10^6$ km^2), led to an overall rate of denitrification as 23·9 – 32·6 Tg N y^{-1}. This is quite close to the estimate of Naqvi (1987), but higher by a factor of 2–2·7 than the estimate of Mantoura *et al* (1993) utilizing freon-11 (F-11)-based ventilation time of the oxygen-deficient layer (Olson *et al* 1993).

A crude check on the above computations could be made as follows. A modest annual net northward geostrophic flux (surface to bottom) of $4 \times 10^6\,\mathrm{m^3\,s^{-1}}$ required to compensate the southward flux in the Ekman layer (Olson *et al* 1993) should cause a net annual input of $\sim 50\,\mathrm{Tg\,NO_3\text{-}N}$ to the Arabian Sea. This roughly should be the strength of the net local sink of combined nitrogen since the nitrate concentrations in the surface layer are close to zero. This, of course, includes sedimentary denitrification as well. The benthic denitrification rate is unknown, but is expected to be much smaller than the water-column denitrification rate (Naqvi *et al* 1992). Thus, it seems that a rate of water-column denitrification of $\sim 30\,\mathrm{Tg\,N\,y^{-1}}$ is probably of the right magnitude. If so, then the Arabian Sea may make about the same contribution to the global nitrogen flux to the atmosphere as each of the Pacific denitrification sites (Codispoti and Richards 1976; Codispoti and Packard 1980).

A denitrification rate of $30\,\mathrm{Tg\,N\,y^{-1}}$ corresponds to a carbon mineralization of $0{\cdot}87 \times 10^5\,\mathrm{mol\,s^{-1}}$. For comparison, the oxygen influx estimated by Olson *et al* (1993) can sustain an oxidation rate of $\sim 2 \times 10^5\,\mathrm{mol\,s^{-1}}$. These figures highlight the importance of denitrification as a major respiratory process within the subsurface layers of the northwest Indian Ocean.

4. Temporal variability and renewal time of intermediate waters

The unique monsoon circulation in the North Indian Ocean introduces large seasonality in nutrient inputs to the euphotic zone and hence in biological production (Qasim 1977, 1982; Banse 1987). Consequently, particle fluxes to the deep Arabian Sea exhibit a large seasonal signal (Nair *et al* 1989). Moreover, the intermediate-depth water exchange between the Arabian Sea and the rest of the Indian Ocean, which is mostly confined to the western Arabian Sea, also seems to be monsoon-dependent (Swallow 1984). In view of monsoon-related changes in the supply of organic carbon to subsurface waters and in water circulation, one may expect the intermediate waters to undergo appreciable temporal changes.

Observational support for significant short-term changes in the composition of oxygen-poor waters was first provided by Naqvi (1987). Subsequently, Naqvi *et al* (1990) and Noronha (1992) presented data, based on observations in the northeastern Arabian Sea, during successive southwest and northeast monsoon seasons. These clearly indicated variations in denitrification intensity on a seasonal scale. Mapping the distribution of nitrate deficit and nitrite at four isopycnal surfaces within the suboxic zone, Naqvi *et al* (1990) observed changes which seem to result from a seasonal reversal of subsurface water circulation off the Indian coast. During the southwest monsoon, when the eastern Arabian Sea behaves like a typical eastern boundary area (Shetye *et al* 1990), a subsurface poleward undercurrent, common to all eastern boundaries, supplies oxygen and nitrate to this region suppressing denitrification. However, this feature is absent during the northeast monsoon. Seasonal changes may also occur in the northern Arabian Sea due to the aeration associated with winter convection (Banse 1984) [some evidence for this also exists in the work of Noronha (1992)], and in the western Arabian Sea which receives seasonally the outflows from the Red Sea and Persian Gulf (Olson *et al* 1993). Unfortunately, the data available from the northern and western parts are inadequate to assess the extent of seasonal changes in these areas.

The observed changes in chemical composition of the oxygen-deficient waters imply that these waters should be renewed quickly. Swallow (1984) was the first to postulate a vigorous circulation at intermediate depths in the northwestern Indian Ocean. Several efforts have since been made to estimate the ventilation time using chemical data. An understanding of the time scales of renewal of the oxygen-minimum layer is required to assess the roles of various processes (poor circulation versus excessive oxygen consumption) responsible for the development of the intense oxygen minimum. It is also needed to quantify the denitrification rate, since most of the current uncertainty in the estimates of denitrification rate directly reflects that in the ventilation time, as stated earlier.

Naqvi (1987) combined his estimates of the total inventory of denitrified nitrogen and denitrification rate to compute an average renewal time of 4 y. Somasundar and Naqvi (1988) used the model of Liu and Kaplan (1984), which relies on the global relationships between denitrification rate and availability of sinking particles, to arrive at an estimate of 1·6–3·4 y. The ETS-derived denitrification rate yielded a value of ~ 1 y (Naqvi and Shailaja 1993). Olson *et al* (1993) employed a box model involving F-11 saturations observed north of Lat. 12°N. Assuming that F-11 inputs to the oxygen-deficient zone were solely through outflows from the Red Sea and Persian Gulf [which may not be strictly valid since significant winter convection occurs in the northern Arabian Sea (Banse 1984)] and that the waters advecting from the south had negligible F-11 concentrations in 1987, these authors computed the average renewal time of the oxygen-deficient layer ($O_2 < 4\,\mu M$) as ~ 11 y. These results provide quantitative support for Swallow's (1984) hypothesis. Although the range of the above estimates (1–11 y) is rather large, it may be noted that these apply to different volumes. For example, while the estimate of Naqvi and Shailaja (1993) is for the denitrifying layer (ca. 150–500 m), that of Olson *et al* (1993) is applicable to the entire oxygen-deficient layer (150–1000 m). The shallower denitrifying layer, occupying a much smaller (1/4 to 1/5) volume, is expected to have a much shorter renewal time.

5. Relative importance of sinking and dissolved organic matter in supporting denitrification

As described below, recent studies have revealed some intriguing aspects of carbon-nitrogen cycling in the Arabian Sea which may have important implications on the global biogeochemical cycles.

Microbiological investigations using modern techniques had not been taken up in the oxygen-deficient waters of the Arabian Sea until very recently. First measurements of the whole water column bacterial abundance and ^3H-thymidine incorporation (TdR) were made by Ducklow (1993) along a transect extending from the equator to the Gulf of Oman. His results for the *Charles Darwin* Sta. 5, located within the denitrifying zone in the central Arabian Sea, are reproduced in figure 8. The bacterial abundance profile shows a distinct maximum coinciding with the SNM. [^3H]thymidine incorporation is also substantially higher (by a factor of 5) within the upper parts of the SNM than in the overlying and underlying waters. Thus, the high bacterial biomass within the denitrifying waters is probably sustained by higher *in situ* growth

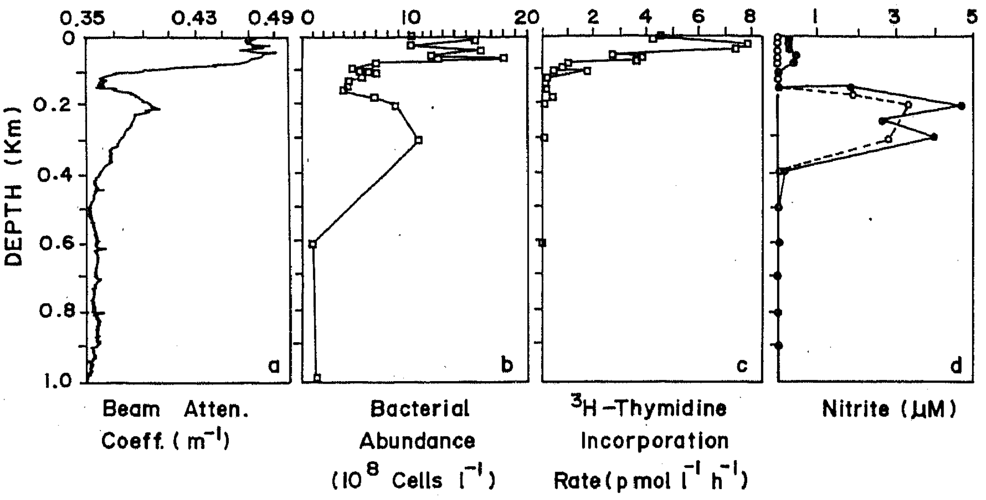

Figure 8. Vertical profiles of (a) beam attenuation coefficient, (b) bacterial abundance, (c) thymidine incorporation, and (d) nitrite. Bacterial abundance and thymidine incorporation data at *Charles Darwin* station 5 (14° 23′N, 66° 53′E) are from Ducklow (1993); nitrite data at the same station (open circles) are from Saager *et al* (1992); beam attenuation and nitrite (solid circles) data at *Sagar Sampada* station 2514 (15° 01′N, 67°E) are from Naqvi *et al* (1993).

rates. A similar bacterial abundance maximum associated with denitrification has been reported from the denitrifying zone off Peru (Spinrad *et al* 1989). What is unusual in the Arabian Sea is that the BP estimated from the observed thymidine incorporation appears to be much higher than the rate of dissolved organic carbon (DOC) release from sinking particles computed from the sediment trap data collected during the same period (figure 9). A comparison of BP integrated within the depth range 100–1000 m with sinking fluxes at 100 m depth is made in table 2. It is evident that the vertical fluxes alone are inadequate to meet the carbon demand of bacteria in subsurface waters of the Arabian Sea. It may be pointed out here that the factors used by Ducklow (1993) to convert thymidine incorporation into bacterial production were very conservative (2×10^{-14} gC per cell, $1{\cdot}18 \times 10^{18}$ cells per mole of thymidine incorporated, and 65·7% TdR incorporation into DNA). Thus, the inferred inadequacy of the vertical fluxes to sustain subsurface respiration cannot result from an overestimation of BP.

Naqvi and Shailaja (1993) and Naqvi *et al* (1993) have provided estimates of subsurface respiration rates from measurements of ETS activity. A typical ETS profile from the reducing zone is reproduced in figure 10. The ETS activity exhibits a maximum coinciding with the SNM. This, of course, is as expected since the ETS activity is a measure of the viable bacterial cells and, as stated above, there occurs a maximum in bacterial biomass at about the same depth. The observed ETS activities within the oxygen minimum are among the highest observed in the ocean at comparable depths (Naqvi and Shailaja 1993). This lends support to the view that the acute oxygen depletion in the Arabian Sea may be to a large extent due to an excessive oxygen demand.

The enzymatic work has led to two unique and significant observations. First, the ETS activities and consequently denitrification rates exhibit geographical changes which are against the trend expected from primary productivity of the overlying

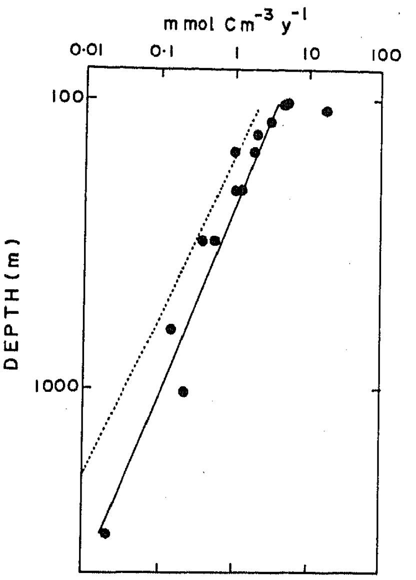

Figure 9. Log–log plots of bacterial production (BP) vs. depth at *Charles Darwin* station 5. BP estimated using conversion factors 2×10^{-14}gC per cell, $1{\cdot}18 \times 10^{18}$ cells per mole of thymidine incorporated, and 65·7% TdR incorporation into DNA. Solid line represents linear regression of BP data. Dotted line is Martin *et al*'s (1987) equation for carbon regeneration from sinking particles based on the data collected using drifting sediment traps (from Ducklow 1993).

Table 2. Estimates of bacterial production below 100 m in relation to vertical POC flux (from Ducklow 1993).

Station locations	Bacterial production 100–1000 m ($\mathrm{mg\,C\,m^{-2}\,d^{-1}}$)	Vertical flux at 100 m ($\mathrm{mg\,C\,m^{-2}\,d^{-1}}$)
00° 02′N, 65°E	758	16·5
08° 01′N, 67°E	185	8·3
14° 23′N, 66° 53′E	180	9·1
21° 15′N, 63° 22′E	248	21·8
23° 38′N, 59° 01′E	350	25·6

surface waters. That is, the highest activities were measured beneath the relatively oligotrophic surface waters offshore. But the ETS activities (figure 11) show a similar geographical distribution as nitrite (figure 3) providing additional evidence for a decoupling between primary production and denitrification. Secondly, the ETS-based denitrification rates appear to be too high to be supported solely by the sinking fluxes. This was first demonstrated by Naqvi and Shailaja (1993) from a comparison of the ETS-derived respiration rates with the particulate carbon supply to the denitrifying layer as obtained from the model of Betzer *et al* (1984). Using a generous estimate of primary production of $278\,\mathrm{gC\,m^{-2}\,y^{-1}}$, the weighted mean of column productivity north of 15°N obtained from the data base of Naqvi (1991), it was computed that at Sta. L13 (figure 11) the carbon requirement for denitrification may exceed its availability through sinking particles by a factor of 4. This inequality has been further accentuated by the findings of Naqvi *et al* (1993) who compared their

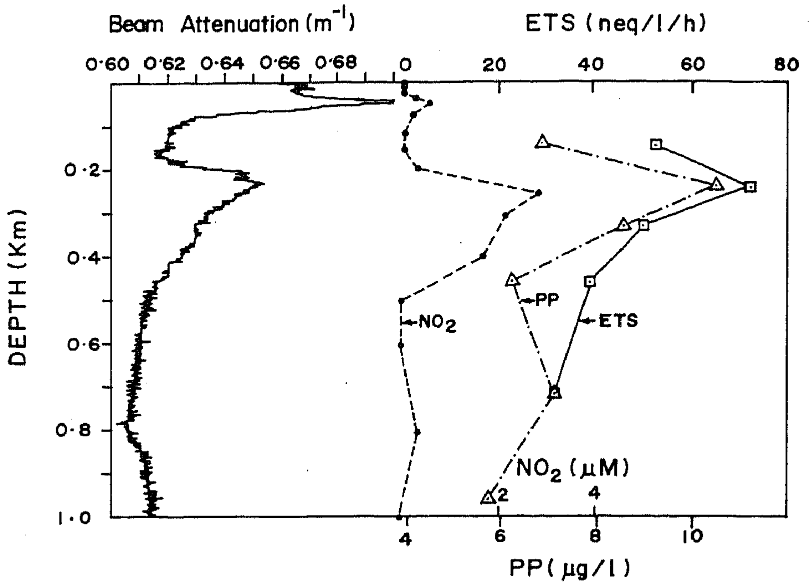

Figure 10. Depthwise distributions of nitrite, electron transport system (ETS) activity, and particulate protein (PP) in relation to beam attenuation at *Sagar Sampada* station 2496 (17° 30′N, 67° 30′E) (from Naqvi *et al* 1993).

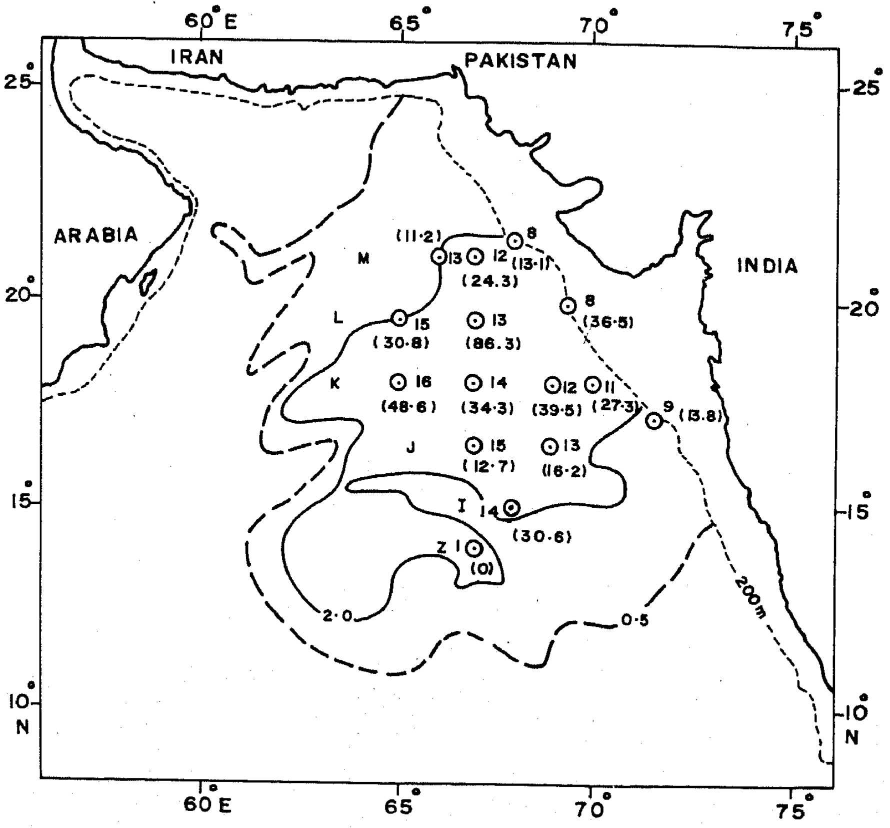

Figure 11. The ETS-based denitrification rates (figures within parentheses, gN m^{-2} y^{-1}) integrated over the water column with $NO_2 \geqslant 0{\cdot}5\,\mu M$. The sampling stations are identified by the legs M, L, etc. and the numbers outside parentheses. The contours represent the distribution of nitrite (μM) at the secondary maximum (see figure 3). Note the denitrification rate = 0 at Sta. Z1 reflects $< 0{\cdot}5\,\mu M$ nitrite at the time of observation (from Naqvi and Shailaja 1993).

estimates of ETS-derived respiration rates (corresponding to a mean carbon requirement of 155 $\mathrm{mgC\,m^{-2}\,d^{-1}}$) with the carbon fluxes at 100 m (8·3–25·6 $\mathrm{mgC\,m^{-2}\,d^{-1}}$) measured by B. Zeitzschel (table 2). It may be pointed out that since the ETS method yields denitrification rates that are consistent with those estimated by other methods, as demonstrated earlier, it is unlikely that the above anomaly is caused by the uncertainty in the conversion factors. The apparent inadequacy of the sinking fluxes as well as a lack of correlation between denitrification and primary productivity indicate that the processes usually assumed to supply carbon to support microbial metabolism in subsurface waters may be very different in the Arabian Sea. This is corroborated by the data on BP as discussed earlier.

It has been suggested that the excess carbon required at intermediate depths could be in the form of dissolved organic carbon (DOC) (Ducklow 1993; Naqvi and Shailaja 1993). Naqvi and Shailaja (1993) have also suggested the possible involvement of suspended organic matter transported laterally from the continental margins.

Well-defined particle maxima have been known to occur within the cores of the oxygen minima and nitrite maxima in the eastern tropical Pacific (Pak *et al* 1980; Garfield *et al* 1983; Spinrad *et al* 1989). Recent optical measurements showed that a similar turbid layer occurs in the Arabian Sea as well, as exemplified by the beam attenuation profiles in figures 8 and 10 (Naqvi *et al* 1993). The intermediate particle maximum in the Arabian Sea does not appear to be confined to a specific water mass as it occurs over wide depth and density ranges. Instead it shows a remarkable association with the SNM, both in its horizontal and vertical extents. Significantly, the particle maximum does not exhibit any systematic onshore-offshore gradients expected from an offshore transport of the bottom nepheloid layers, the mechanism suggested by Pak *et al* (1980) for its occurrence off Peru; indeed, it seems to intensify offshore (Naqvi *et al* 1993), consistent with the aforementioned trend in denitrification. As the particle maximum is associated with the maxima in ETS activity and in particulate protein (figure 10), it probably has a large living organic component. Ducklow's (1993) observations reveal that a pronounced bacterial biomass maximum also occurs at about the same depth (figure 8). This has led to the suggestion that, as in the Peruvian upwelling zone, the bacteria may directly account for the increased turbidity at these depths. And since the anomalies in beam attenuation show excellent correlation with the nitrate deficit and nitrite (Naqvi *et al* 1993), it would appear that most of the bacteria within the SNM should be engaged in the nitrate reduction/denitrification pathways.

If the intermediate nepheloid layer in the Arabian Sea, as indeed in other similar environments, mainly arises from an increase in bacterial biomass, then the quasi-horizontal transport of suspended matter may not be very important in providing the additional carbon required to sustain the high respiration rates. A more plausible mode of input of the biodegradable organic carbon is the advective and diffusive transports of DOC from surface layer. Improvements in the analytical techniques, namely the development of a high temperature catalytic oxidation (HTCO) method for estimating DOC in seawater, have led to the recognition of a large pool of organic carbon present as DOC that escaped detection by the wet oxidation technique (Sugimura and Suzuki 1988). More recent results by Martin and Fitzwater (1992) reinforce this view, although a correlation between the DOC and oxygen consumption has not been validated. However, Ogawa and Ogura (1992) found that the DOC concentrations estimated by the HTCO are only slightly higher than those obtained from the wet oxidation method. One important inference that could be made from these rather conflicting results obtained following apparently the same technique is that the DOC may be differentially labile in space. Large vertical gradients observed by Sugimura and Suzuki (1988) and the steep horizontal gradient seen by Martin and Fitzwater (1992) appear to testify to the active involvement of DOC in some geographical areas (Toggweiler 1992). Following a HTCO method using nickel and cobalt oxide as catalyst, appreciable vertical and horizontal gradients in the DOC concentrations have been observed by Kumar *et al* (1990) in the Arabian Sea as well. Although the DOC–AOU correlation observed by these authors was not very significant in pooled data, better correlations are evident when the data are treated separately for the northern, central and southern Arabian Sea. Significantly, the lowest DOC values ($\sim 60\,\mu$M) occurred within the denitrifying layer. Considered together with the evidence for the inadequate vertical fluxes to meet the mid-depth carbon demand, these observations strongly suggest that denitrification could be dominantly fuelled by DOC. It seems quite possible that the DOC is more effectively utilized by bacteria under denitrifying conditions leading to their proliferation. An implication of these results is that, given the short renewal time of the denitrifying layer, the labile fraction of the DOC should be characterized by short regeneration times.

6. Nitrous oxide cycling

Considerable attention is being paid in recent years to understand the fate and fluxes of nitrous oxide (N_2O) in the ocean, especially within the low-oxygen environments where its turnover appears to be very rapid (Codispoti *et al* 1992). Two reports have appeared in the literature on the cycling of this important greenhouse gas in the Arabian Sea (Law and Owens 1990; Naqvi and Noronha 1991). The results are in excellent agreement and suggest that the Arabian Sea serves as a significant source

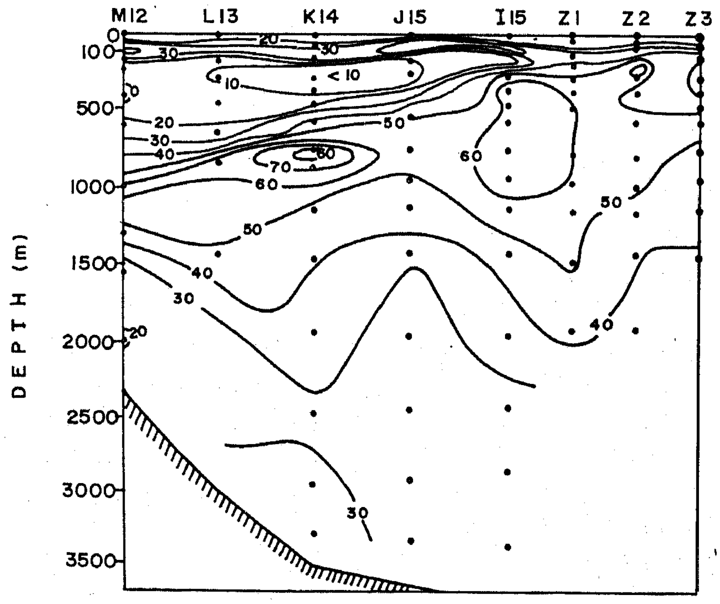

Figure 12. Distribution of nitrous oxide (nM) along the same transect as in figure 6 (from Naqvi and Noronha 1991).

of nitrous oxide to the atmosphere (0·30–0·44 Tg y^{-1}). Apparently, the large atmospheric fluxes are caused by a high rate of N_2O pumping to the surface layer as a result of upwelling and vertical mixing (Naqvi and Noronha 1991).

High nitrous oxide concentrations in subsurface waters of the Arabian Sea result from an enhanced production of N_2O in the low-oxygen waters, as long as the latter do not experience denitrification (Naqvi and Noronha 1991). Vertical profiles of N_2O generally exhibit a single maximum coinciding with the oxygen minimum (Yoshinari 1976). The onset of denitrification, however, greatly affects the N_2O distribution as shown in figure 12. The pronounced minimum in N_2O embedded between two maxima obviously reflects N_2O consumption within the core of the denitrifying layer since the N_2O minimum coincides with the SNM (figure 7). The denitrifying layer acts as a sink for the oceanic N_2O, as do the reducing shelf/slope sediments; the combined strength of these sinks in the Arabian Sea has been estimated as > 2 Tg N y^{-1} (Naqvi and Noronha 1991). The denitrifying layer also insulates low-oxygen, high N_2O waters of the lower part of the oxygen-deficient zone from upper layers which ventilate part of their N_2O to the atmosphere.

7. Some other consequences of reducing conditions

As stated earlier, despite the rapid renewal of intermediate waters, suboxic conditions develop at mid-depths in the Arabian Sea. This is partly due to the fact that the waters responsible for renewal may be originally oxygen-depleted (Swallow 1984), and partly because of high respiration rates evident from the ETS data. However, the waters advecting from the south ensure sufficient supply of nitrate to the Arabian Sea. Calculations included in section 3 suggest that denitrification may account for about 1/3 of the total carbon respired within the oxygen-deficient zone. Thus, the large nitrate flux could be important in preventing the development of complete anoxia (i.e., sulphate reduction). However, as stated earlier, the lowered redox potential affects the cycling of several other redox elements.

Saager *et al* (1989) investigated depth profiles of dissolved manganese at several locations and that of dissolved iron at one location in the Arabian Sea. Their results (figure 13) reveal the occurrence of broad maxima in concentrations of both Mn and Fe within the oxygen-poor zone, attributed to *in situ* dissolution of Fe–Mn from particulate matter and/or lateral inputs of these metals mobilized within the reducing continental shelf/slope sediments. Interestingly, the greatest accumulation of Mn was found to occur at the periphery (Sta. 7) or outside (Sta. 9) the denitrification zone beneath the highly productive waters of the western Arabian Sea. It was ascribed to different mechanisms of Mn enrichment. While the *in situ* dissolution might dominate within the denitrifying waters of the central Arabian Sea, lateral inputs from the continental margin into the Red Sea core layer during its transport towards the north may be more important in the western Arabian Sea (Saager *et al* 1989). However, it must be noted that the salinity maximum corresponding to the Red Sea water was not prominent off Oman (German and Elderfield 1990), which is in conformity with the previous results (Naqvi *et al* 1982). An association of the Mn maximum (at $\sigma_\theta = 27{\cdot}2$) with the Red Sea water may thus be questioned. An alternate explanation of the above anomaly could be that the larger enrichment in the western Arabian Sea may be caused by Mn mobilization within the microreducing sites provided by

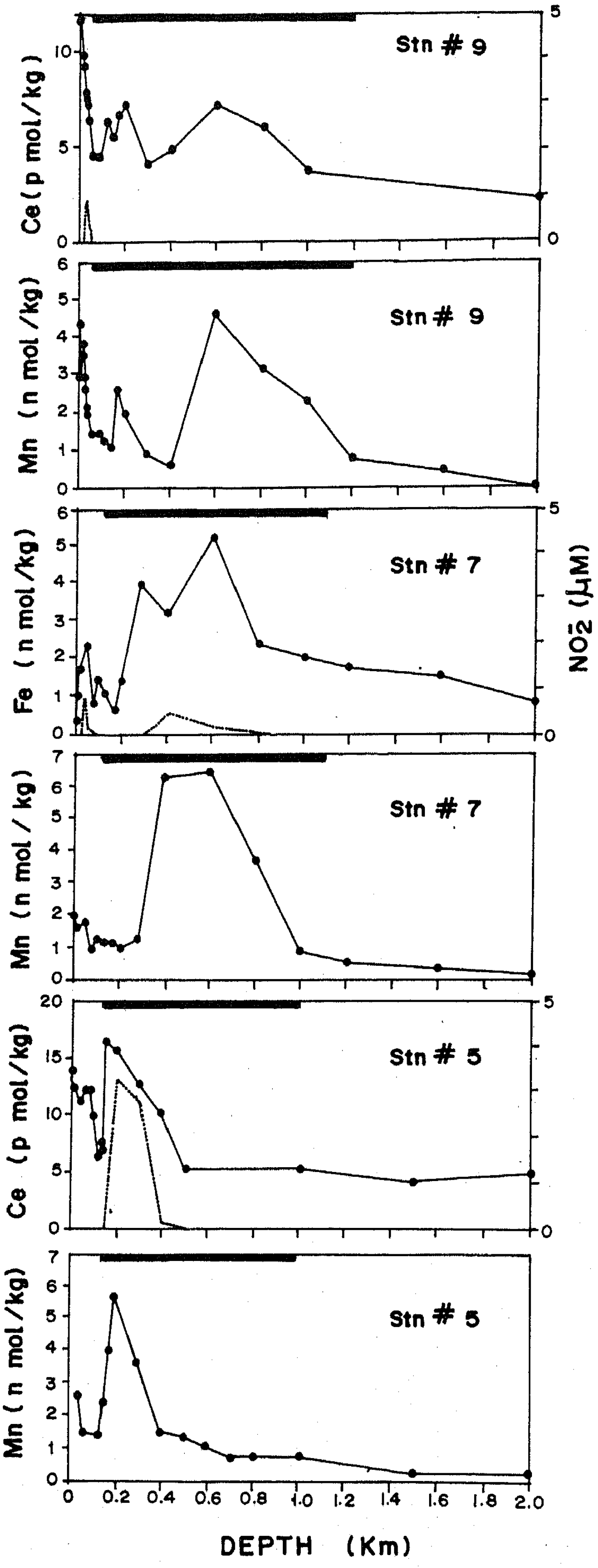

Figure 13. Vertical profiles of Mn, Fe and Ce at selected *Charles Darwin* stations (Sta. 5: 14° 30′N, 67°E; Sta. 7: 21° 16′N, 63° 22′E; Sta. 9: 23° 30′N, 59°E). Nitrite profiles (dotted curves) have been included for comparison. The solid bar on the right side of each figure denotes the oxygen minimum zone ($O_2 < 20\,\mu M$) [data from Saagar *et al* (1989) and German and Elderfield (1990)].

the particle aggregates. The western Arabian Sea offers ideal conditions for the development of such sites as the ambient oxygen levels approach but do not reach suboxia, and the export production is extremely high. On the other hand, as discussed earlier, the denitrifying layer underlies surface waters which are much less productive. Therefore, although the conditions within the bulk of the water column are denitrifying, Mn available for reduction may be much less.

Results of an investigation of the rare earth elements (REE) are in general conformity with the above report: German and Elderfield (1990) found a three fold enrichment of dissolved cerium in the suboxic waters as compared to the overlying oxygenated layer (figure 13), ostensibly caused by the reduction of Ce(IV) to Ce(III). The other strictly-trivalent REE, by comparison, showed a very modest enrichment of only 10–20%, which was attributed to the adsorption/desorption processes associated with precipitation and dissolution of ferromanganese oxyhydroxides. As in case of Mn, Ce enrichment was not limited to the denitrifying zone, although the Ce maximum was sharper and more intense in the denitrifying waters; it also occurred at shallower depths (figure 13). However, their results also show that Ce and Mn distributions in the Gulf of Oman could be somewhat decoupled presumably due to minor changes in oxygen concentrations, thus highlighting the delicate biogeochemical poise that exists in the northwestern Indian Ocean and the sensitivity of various redox systems to the ambient oxygen levels.

8. Future work

Our knowledge of the biogeochemical cycling within the coupled carbon and nitrogen systems, particularly water column denitrification, in the northwestern Indian Ocean has improved greatly since the mid 1970s. The work carried out so far has uncovered several unusual aspects of the biogeochemistry of this oceanographically unique part of the oceans, but it has also raised many new questions. The issues that need to be addressed have been identified by Smith *et al* (1991). Here only a few points will be emphasized concerning the observational as well as modelling work that should be pursued in future.

Given the large physical, biological and chemical changes that occur within the Arabian Sea, the observational coverage of this region has been highly uneven in both space and time. For example, very few data are presently available from the most productive regions (western and northern Arabian Sea) especially during the southwest monsoon. Several international programmes under the JGOFS currently underway or to be undertaken shortly are expected to remedy this situation. Creation of a good data base, adequate for providing a measure of the spatial and temporal changes, should get the highest priority.

Some recent results from elsewhere in the oceans highlight the importance of denitrification within sediments in the overall nitrogen cycling (Christensen *et al* 1987; Devol 1991). No attempt has been made to measure sedimentary denitrification rates in the Arabian Sea. These can be expected to be significant particularly within marginal sediments which are in contact with the oxygen-minimum layer. Benthic chamber studies similar to those planned as a part of the US JGOFS Program should be extended to the eastern Arabian Sea to quantify this important and yet unknown term in the nitrogen budget.

There has been only one modest attempt so far to model denitrification in the Arabian Sea: Anderson *et al* (1982) used a one dimensional (vertical) approach to simulate nitrate and nitrite distributions in the oceanic suboxic zones including the Arabian Sea. In an area where the horizontal processes cannot be ignored (particularly in view of the observations which show that the sinking fluxes may be insufficient to meet the organic carbon demand within the oxygen minimum zone), one-dimensional models have very limited utility. However, more realistic modelling would await a larger data base particularly on subsurface circulation in this region.

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