

## 'NO', a useful tool for the estimation of nitrate deficits in the Arabian Sea

S. W. A. NAQVI\* and R. SEN GUPTA\*

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**Abstract**—The property 'NO', defined as the sum of  $O_2$  and  $8.65 NO_3$ , varies linearly with potential temperature ( $\theta$ ) outside the denitrification zone in the Arabian Sea, but a change in slope of the 'NO'– $\theta$  regression line occurs at  $\theta \approx 15^\circ C$ . The 'NO'– $\theta$  relationships have been utilized to compute the 'original' nitrate concentrations which are combined with the observed data to calculate the nitrate deficits within the denitrification zone. This procedure, which approximately accounts for the 'reserved' nitrate, eliminates the errors associated with the use of the variable  $\Delta AOU:\Delta PO_4$  ratio in earlier methods. In upper layers, the present method yields deficits close to those deduced from the oxidative ratios and  $(NO_3)_r - (PO_4)_r$  relationship. In deep layers (depth  $>500$  m), however, slightly higher values are obtained with the present method, presumably due to the variability of relationships between 'reserved' nutrients. Distribution of nitrate anomaly ( $\Delta N$ ) along a section running from  $15^\circ N, 67^\circ E$  to  $21^\circ N, 63^\circ E$  closely follows the distribution of nitrite at the secondary maximum. Nitrate anomalies in excess of  $8 \mu g\text{-at. dm}^{-3}$  are observed frequently along this section. An intermediate minimum in  $\Delta N$  is observed at some stations at depths where the Subantarctic Mode Water is encountered. This water mass, relatively rich in oxygen, appears to be a source of limited oxygen supply which probably prevents the intermediate layers from becoming completely anoxic.

### INTRODUCTION

NITRATE deficits within the oxygen-deficient layer of the Arabian Sea have been estimated using either the nitrate–salinity relationship (DEUSER *et al.*, 1978) or the oxidative ratios (SEN GUPTA *et al.*, 1976; NAQVI *et al.*, 1982; QASIM, 1982; NAQVI and QASIM, 1983). The two procedures have been evaluated by NAQVI *et al.* (1982) who used the modified procedure of CLINE and RICHARDS (1972). However, the choice of  $\Delta AOU:\Delta NO_3$  and  $\Delta AOU:\Delta PO_4$  ratios necessary for estimating 'original' nitrate concentrations greatly affects the results of the procedure followed by NAQVI *et al.* (1982). The values of  $\Delta AOU:\Delta NO_3$  ratio reported by various workers in the northern Indian Ocean are in good agreement if the values corresponding to very low dissolved oxygen are omitted (NAQVI *et al.*, 1982); the observed ratio (8.65, by moles) is also remarkably close to the theoretical value (8.625) (REDFIELD *et al.*, 1963). On the other hand, the  $\Delta AOU:\Delta PO_4$  ratio varies from 124 (by moles) (NAQVI *et al.*, 1982) to 140 (SEN GUPTA *et al.*, 1976), as compared to the theoretical value of 138 (REDFIELD *et al.*, 1963). Whether the observed variations and the departure from the theoretical value are real or caused by the inconsistency in phosphate data,† is difficult to ascertain. A change in slope of

\* National Institute of Oceanography, Dona Paula, Goa 403 004, India.

† There is considerable inconsistency in phosphate data collected during various cruises in the Arabian Sea. For example, the data collected by F.S. *Meteor* and R.V. *Anton Bruun* during the International Indian Ocean Expedition from the same area differ widely, presumably due to different analytical techniques followed and perhaps also because of errors in standardization.

AOU- $\text{PO}_4$  regression line occurs at  $\text{PO}_4 \approx 2.5 \mu\text{M dm}^{-3}$  (DEUSER *et al.*, 1978; NAQVI *et al.*, 1982), probably as a result of a steep increase in 'reserved' phosphate at this level. Excess phosphate regenerated during denitrification will also contribute to the higher phosphate concentrations. Therefore, lower  $\Delta\text{AOU}:\Delta\text{PO}_4$  ratio is obtained if the high phosphate values are not omitted from the calculation. In any event, the uncertainty in the  $\Delta\text{AOU}:\Delta\text{PO}_4$  ratio will introduce significant uncertainty in the calculation of 'reserved' phosphate. Further, the relationships between the 'reserved' fractions of nitrate and phosphate used for the calculation of  $(\text{NO}_3)_r$  may also vary with depth and geographically. For these reasons, we have attempted to evolve an alternate procedure which does not involve the use of  $\Delta\text{AOU}:\Delta\text{PO}_4$  ratio, but where contribution from the 'reserved' nitrate is considered, albeit approximately.

The use of nitrate tracer 'NO', defined as the sum of  $\text{O}_2$  and  $9 \text{NO}_3$ , was introduced by BROECKER (1974) to delineate water masses with different 'preformed' nitrate contents. For a given parcel of seawater, this property should remain nearly constant as the decrease in dissolved oxygen due to the oxidation of organic matter will be balanced by a corresponding increase in nitrate. The departure of 'NO' from the oxygen saturation value for a given temperature and salinity is a measure of the 'preformed' or 'reserved' nitrate content (BROECKER, 1974). As mentioned earlier, the  $\Delta\text{AOU}:\Delta\text{NO}_3$  ratio is relatively constant in the northern Indian Ocean, 8.65 (by moles). Therefore, we define 'NO' as the sum of  $\text{O}_2$  and  $8.65 \text{NO}_3$ . In this paper, we shall examine the anticipated linear variation of this property with potential temperature ( $\theta$ ), and the application of the 'NO'- $\theta$  relationships, deduced outside the denitrification zone, for the computation of 'original' nitrate concentrations within the denitrifying layer.

#### MATERIAL AND METHODS

Data from several locations in the Arabian Sea have been used in the present study (Fig. 1). Stations 1899 to 1937 were occupied during cruises of R.V. *Gaveshani* in January to February 1981; Stas 2689 to 2695 were occupied by the same vessel in April 1983 and Sta. 85 was occupied during a cruise of R.V. *Sagar Kanya* in June 1983.

Metallic Nansen bottles, TPN Hydrobios, and PVC Niskin water samplers fitted with reversing thermometers were used for collecting samples. Nutrients, dissolved oxygen, and salinity were determined shortly after collection on board ship. Inorganic phosphate was estimated by the method of MURPHY and RILEY (1962), nitrate by the method of MORRIS and RILEY (1963) as modified by GRASSHOFF (1964), and nitrite by the method of BENDSCHNEIDER and ROBINSON (1952). Analyses were performed manually on board *Gaveshani*, but on board *Sagar Kanya*, a SKALAR 6-channel analyzer 5100/1 was used. Dissolved oxygen was determined by the Winkler method as modified by CARPENTER (1965). However, for Stas 2689 to 2695, oxygen estimations from the oxygen minimum layer ( $\sim 150$  to  $1200$  m) were carried out by the colorimetric method of BROENKOW and CLINE (1969). The manganous sulphate and alkaline iodide solutions used in the colorimetric method were deoxygenated initially with nitrogen, but halfway through the cruise a leakage in the valve emptied the cylinder, and the degassing was performed by keeping the reagents under reduced pressure for half an hour before and during collection. This mode of deoxygenation might not have given the best results, but the oxygen values obtained were consistently low, generally below  $3 \mu\text{M dm}^{-3}$ . Salinity was measured with an AUTOSAL model 8400 salinometer.

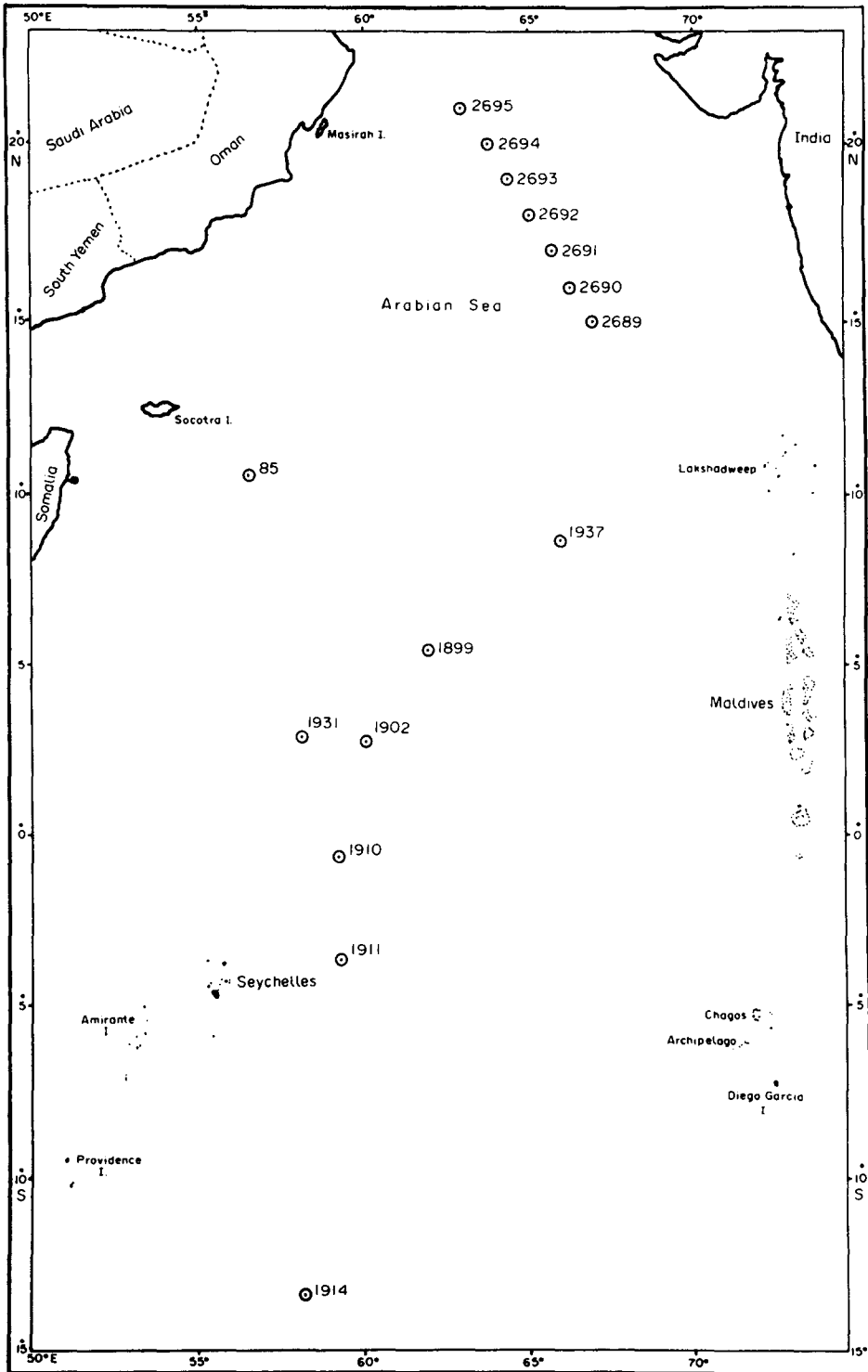


Fig. 1. Location of stations.

## RESULTS AND DISCUSSION

A plot of 'NO' vs  $\theta$  for Stas 1899 to 1937 and 85 is presented (Fig. 2). As the denitrification zone in the Arabian Sea lies north of about 11°N latitude (LUKASHEV, 1980; NAQVI *et al.*, 1982; NAQVI and QASIM, 1983; SEN GUPTA and NAQVI, 1984), the stations selected to deduce the 'NO'– $\theta$  relationships are all located south of this latitude. For the  $\theta$  range of interest, which covers the oxygen-deficient layer in the Arabian Sea, 'NO' correlates linearly with  $\theta$ , but a change in slope occurs at  $\theta \approx 15^\circ\text{C}$ . The 15°C isotherm could be taken to represent the base of a strong upper thermocline (Dr. J. S. SASTRY, personal communication). Hence a change in slope at this level merely reflects the transition from cold waters of the main thermocline, mostly derived from high southern latitudes and with high 'preformed' or 'reserved' nitrate, to warm surface waters with low 'reserved' nitrate. The 'NO'– $\theta$  regression line closely follows the curve for oxygen saturation at higher temperatures, but a steep increase in 'reserved' nitrate below the thermocline results in a progressively larger departure of 'NO' from the oxygen saturation value as the temperature decreases (Fig. 2). The lines giving the two-way least squares fits for the two layers are represented by the following equations:

$$'NO' = 429.81 - 12.4416\theta \quad (\pm 9.17) \quad 3 \leq \theta < 15 \quad (1)$$

$$'NO' = 290.66 - 2.9288\theta \quad (\pm 8.67) \quad 15 \leq \theta \leq 26. \quad (2)$$

The numbers in parentheses give the sample standard deviation from the regression,  $\sigma = \sqrt{|\Sigma(y-\hat{y})^2/(n-2)|}$ , where  $y$  and  $\hat{y}$  are the observed and predicted values of 'NO' respectively.

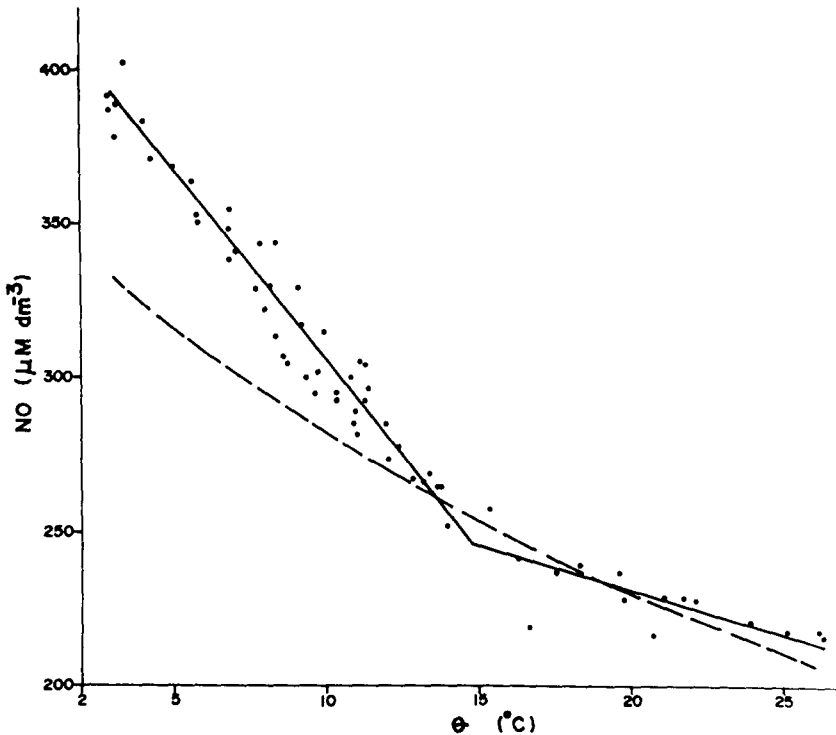


Fig. 2. Plot of 'NO' vs potential temperature ( $\theta$ ) outside the denitrification zone. The straight lines represent the two-way linear least-squares regression between 'NO' and  $\theta$ , with a change in slope at  $\theta \approx 15^\circ\text{C}$ . The broken curve gives the oxygen saturation for 35‰ salinity.

To use the above relationships within the denitrification zone, it should be assumed that for a given temperature, the 'reserved' nitrate will remain constant. This assumption may not be *strictly* valid, since the fraction of water masses originating in the Red Sea and Persian Gulf (characterized by low concentrations of 'reserved' nutrients) will increase progressively northward in the Arabian Sea. However, the assumption should be *nearly* so because the northward decrease in 'reserved' nitrate is accompanied by an increase in temperature. Considering that equations (1) and (2) have been deduced using data from stations located widely apart, the relatively small scatter in Fig. 2, perhaps, justifies the assumption. The quantity 'NO' equals to the oxygen saturation value plus 8.65 times the concentration of 'reserved' nitrate (BROECKER, 1974). The oxygen saturation varies with both salinity and temperature. However, equations (1) and (2) give the variations with temperature, ignoring the effect of salinity. This may introduce a slight error because salinity is known to increase steadily northward at any given level in the Arabian Sea (WYRTKI, 1971). The error, however, is small considering that a difference of  $1 \times 10^{-3}$  in salinity will result in a difference of 0.03 to 0.05  $\text{cm}^3 \text{dm}^{-3}$  in oxygen saturation for the ranges of salinity and temperature encountered within the area of study. The difference is negligible, being of the same order as the accuracy of oxygen estimation by the Winkler method.

Equations (1) and (2) have been used to compute 'NO' from potential temperature within the oxygen-deficient layer of the Arabian Sea. The nitrate anomaly has been calculated with the equation:

$$\Delta N = \frac{(\text{NO} - \text{O}_2)}{8.65} - (\text{NO}_3 + \text{NO}_2). \quad (3)$$

Although ammonia was determined at some locations following the indophenol blue method of KOROLEFF (1969), the results were not very reliable due to high blanks. The upper limit on ammonia within the denitrifying layer of the Arabian Sea could, however, be placed at  $1 \mu\text{M dm}^{-3}$  (DEUSER *et al.*, 1978), and the concentrations are rather uniform below the thermocline (NAQVI and QASIM, 1983). For these reasons ammonia has not been considered in equation (3).

In addition to nitrite, the other intermediate products of denitrification are nitric and nitrous oxides. However, the batch culture experiments indicate that NO and  $\text{N}_2\text{O}$  do not accumulate during marine denitrification, unlike nitrite (BARBAREE and PAYNE, 1967; GOERING and CLINE, 1970). This is further supported by field observations in the eastern tropical North Pacific (ETNP). COHEN and GORDON (1978) observed a pronounced minimum in  $\text{N}_2\text{O}$  profiles within the oxygen-deficient layers of the ETNP. The minimum in  $\text{N}_2\text{O}$  coincided with the secondary nitrite maximum, and the  $\text{N}_2\text{O}$  concentrations within the denitrifying layer correlated negatively with the concentration of nitrite and the enrichment of  $^{15}\text{N}$  [a measure of denitrification intensity as  $^{14}\text{NO}_3$  is utilized preferentially during denitrification (CLINE and KAPLAN, 1975)]. Cohen and Gordon concluded that the denitrification in the ETNP results in a net consumption of  $\text{N}_2\text{O}$ , unlike the denitrification in soils which leads to a net production of  $\text{N}_2\text{O}$  (HAHN and JUNGE, 1977). As pointed out by NAQVI *et al.* (1982) and NAQVI and QASIM (1983), the intermediate stages of denitrification in the Arabian Sea and the ETNP differ slightly in that nitrite accumulates at relatively higher levels in the former region, although the maximum amounts of denitrified nitrogen in the two regions are about the same. This has been attributed to a high rate of primary production in the Arabian Sea; preferential reduction of nitrate to nitrite and the consequent greater accumulation of intermediate nitrite appears to occur when organic substrate levels are high (ANDERSON *et al.*, 1982). How these

differences affect the distribution of  $N_2O$  within the denitrifying layer of the Arabian Sea is not known. There is no report so far on the variations of  $N_2O$  in the North Indian Ocean. However, it seems reasonable to assume that  $N_2O$  levels in the Arabian Sea should be of the same order of magnitude as those observed in the ETNP [subsurface concentrations reported by COHEN and GORDON (1978) in the ETNP range between  $<10$  and  $>40$   $nM\ dm^{-3}$ , with minimum and maximum values occurring at the depths of the secondary nitrite maximum and at the lower portion of the thermocline, respectively]. Contributions of  $N_2O$  and  $NO$  to the marine denitrification budget may be considered as negligible due to the extremely low concentrations of these constituents in seawater, and hence equation (3) could be taken to give the terminal reduction to gaseous nitrogen.

A comparison of the results obtained with equations (1) to (3) (designated hereafter as Method 1) with those by the procedure outlined by NAQVI *et al.* (1982) (designated as Method 2) is presented in Fig. 3 for Sta. 2690. Excess phosphate regenerated during denitrification would lead to apparently high deficits in Method 2. The maximum value of  $\Delta N$  observed at Sta. 2690 was  $9.2\ \mu g\text{-at.}\ dm^{-3}$ . Assuming that the  $\Delta NO_3:\Delta PO_4$  ratio maintained during denitrification is 94.4:1 (RICHARDS, 1965), a maximum of  $0.097\ \mu M\ dm^{-3}$  of phosphate will be regenerated during denitrification. As the 'reserved' nitrate in Method 2 is computed from the equation:

$$(NO_3)_r = 13.7 (PO_4)_r - 8.9 \quad (4)$$

the associated errors in the computed values of  $(NO_3)_r$  and  $\Delta N$  will be  $1.33\ \mu M\ dm^{-3}$  and

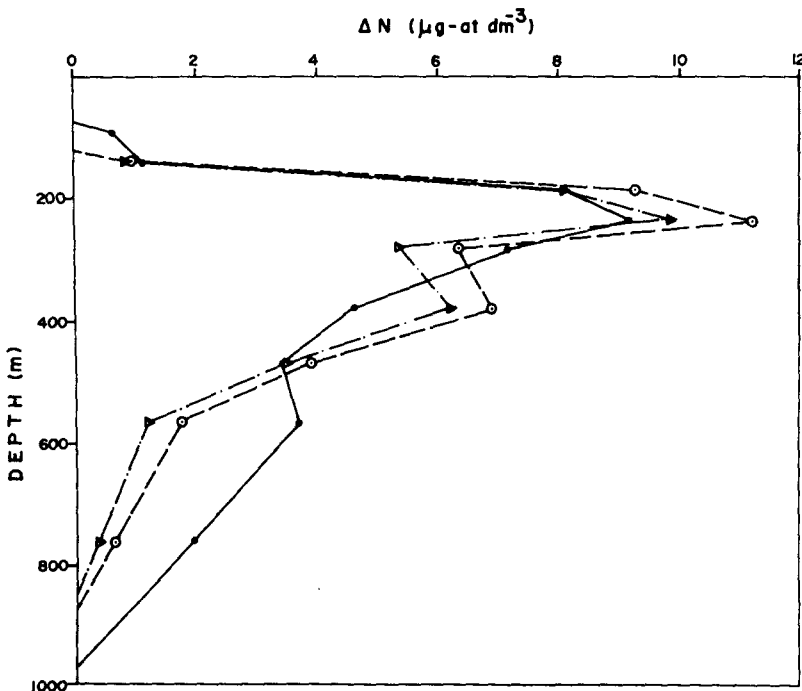
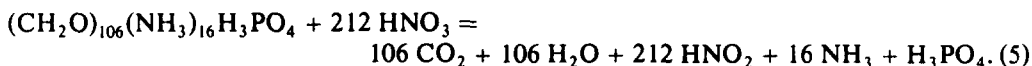


Fig. 3. Profiles of nitrate anomaly ( $\Delta N$ ) constructed for Sta. 2690 with (a) the present method, solid circles; (b) the method of NAQVI *et al.* (1982), open circles; and (c) same as (b) but corrected for phosphate released during denitrification, triangles.

$1.33 \mu\text{g-at. dm}^{-3}$ , respectively. Some phosphate will also be released during the reduction of nitrate to nitrite.



However, as only 1 mole of phosphate is released with the production of 212 moles of nitrite by this reaction, the phosphate added by this step will not exceed  $0.024 \mu\text{M dm}^{-3}$  for nitrite concentrations up to  $5 \mu\text{M dm}^{-3}$ . Therefore, the contribution of this reaction has been ignored and the  $\Delta\text{N}$  values estimated with Method 2 have been corrected only for the excess phosphate regenerated during denitrification, using the  $\Delta\text{N}$  values calculated from Method 1 for this purpose. The results so obtained are also presented in Fig. 3.

The deficits obtained with the two procedures agree very well in the upper layers when the correction for excess phosphate is made (Fig. 3). In deeper layers (depth  $>500$  m), however, the agreement is not so good, with Method 2 yielding consistently lower deficits. The same method also gives negative values reaching up to  $-8 \mu\text{g-at. dm}^{-3}$  at all stations at depths  $>1000$  m, indicating that the 'original' nitrate concentrations derived from this procedure are too low for deep layers. This discrepancy may arise from error in the calculation of 'reserved' nitrate from the relationship between the 'reserved' fractions of nitrate and phosphate, which probably shows variations with depth.

Because the present method involves fewer steps in computations, it is much simpler and probably more accurate and consistent than Method 2, as the uncertainties associated with variable  $\Delta\text{AOU}:\Delta\text{PO}_4$  ratio and errors in phosphate measurements have been eliminated. The sensitivity of Method 2 to errors in phosphate arises from a high slope of the  $(\text{NO}_3)_r - (\text{PO}_4)_r$  regression line (equation 4); an error of  $0.1 \mu\text{M dm}^{-3}$  in phosphate will result in an error of about  $1.4 \mu\text{g-at. dm}^{-3}$  in  $\Delta\text{N}$ . The accuracy of both procedures ultimately will depend upon the accuracy of nitrate measurements, but the overall uncertainty in  $\Delta\text{N}$  computed by the present procedure should not exceed 1 to  $2 \mu\text{g-at. dm}^{-3}$ .

Nitrate anomalies calculated with equations (1) to (3) for Stas 2689 to 2695 frequently exceed  $8 \mu\text{g-at. dm}^{-3}$  at depths between 150 and 600 m (Fig. 4). A maximum value of  $13.7 \mu\text{g-at. dm}^{-3}$  occurs at 189 m at Sta. 2691. The distribution of  $\Delta\text{N}$  closely follows that of nitrite at the secondary maximum (Fig. 5), but the absolute values of  $\Delta\text{N}$  do not always correlate with the concentration of secondary nitrite, in conformity with the results of NAQVI *et al.* (1982). Highest values of  $\Delta\text{N}$  are associated with minima in nitrate, and the deficits extend to progressively deeper layers from south to north. The integrated nitrate anomaly also increases northward, ranging from  $3.02 \text{ g-at. m}^{-2}$  at  $15^\circ\text{N}$  to  $7.31 \text{ g-at. m}^{-2}$  at  $21^\circ\text{N}$ , with an average of  $4.67 \text{ g-at. m}^{-2}$  (Table 1). The northward increase is consistent with the results of NAQVI *et al.* (1982) and QASIM (1982), but the mean value is much lower than that reported by QASIM (1982), for the northern Arabian Sea ( $14.29 \text{ g-at. m}^{-2}$ ). While SEN GUPTA *et al.* (1980) estimated an average value of  $5 \text{ g-at. m}^{-2}$  at  $15^\circ\text{N}$ , NAQVI *et al.* (1982) arrived at an average of  $3.11 \text{ g-at. m}^{-2}$  between  $12$  and  $15^\circ\text{N}$  latitudes. These values compare favourably with the present results.

DEUSER (1978) estimated the total 'standing crop' of denitrified nitrogen in the Arabian Sea to be  $4 \times 10^{12}$  g. Assuming the residence time of the Persian Gulf core layer in the Arabian Sea to be 3 to 30 years, the rate of denitrification in the Arabian Sea was estimated to lie between  $10^{11}$  and  $10^{12} \text{ gN y}^{-1}$ , representing 1 to 10% of the global denitrification. Based on an average integrated nitrate anomaly of  $3.144 \text{ g-at. m}^{-2}$ , and assuming the mean residence time of the denitrifying layer to be 30 y, NAQVI *et al.* (1982) calculated the rate of denitrifica-

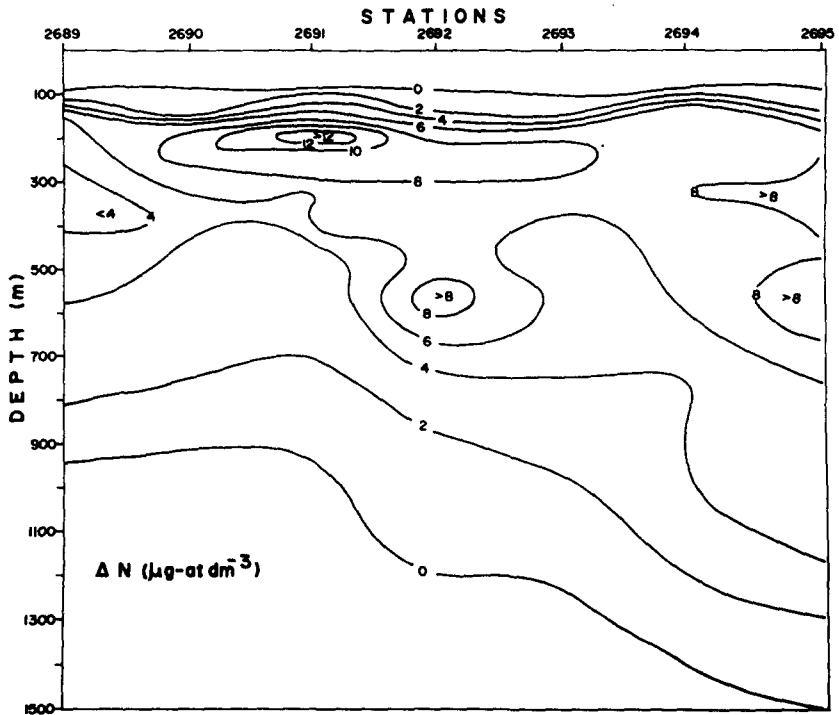


Fig. 4. Distribution of nitrate anomaly along a section running from  $15^{\circ}\text{N}$ ,  $67^{\circ}\text{E}$  to  $21^{\circ}\text{N}$ ,  $63^{\circ}\text{E}$

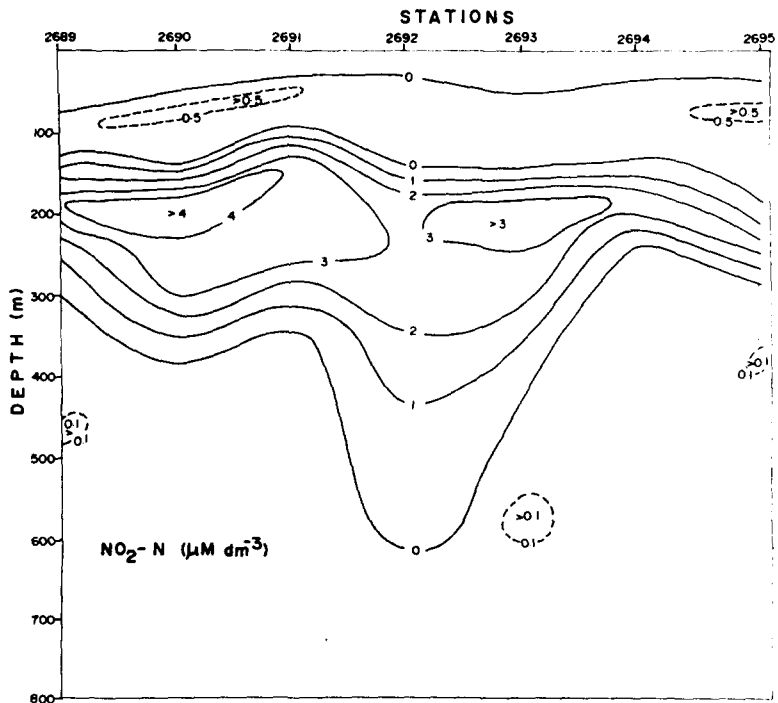


Fig. 5. Distribution of nitrite-nitrogen along the same section as in Fig. 4.



Table 1. *Integrated nitrate anomaly in the Arabian Sea*

Station	2689	2690	2691	2692	2693	2694	2695
Integrated $\Delta N$ (g-at. m <sup>-2</sup> )	3.02	3.08	3.65	4.97	4.53	6.11	7.31

tion in the Arabian Sea to be  $3.2 \times 10^{12}$  g y<sup>-1</sup>, approximately 5% of the global denitrification rate. Our average of the integrated nitrate anomaly is about 50% higher than the corresponding value obtained by NAQVI *et al.* (1982), presumably due to more intense denitrification in our area of study. Consequently, the rate of denitrification in the Arabian Sea should be higher than estimated previously. However, in view of various uncertainties inherent in the calculations, all such estimates at best should be taken as first approximations. For example, in addition to the systematic errors in the computations of nitrate deficits, the choice of residence time affects the results greatly. Our data are not sufficient to arrive at a more precise estimate; however, it can be concluded that the Arabian Sea may contribute up to 10% to the global denitrification budget.

An interesting and hitherto ignored aspect of  $\Delta N$  distribution in the Arabian Sea is the occurrence of two maxima at some stations (2689, 2692, and 2695). The maxima are separated by a layer of relatively low  $\Delta N$ . Similar features are discernible in the  $\Delta N$  profiles published by NAQVI *et al.* (1982) and NAQVI and QASIM (1983), especially pronounced in the southern sector of the denitrification zone. Although the seasonal and patchy processes of primary production may produce the observed features, a more likely origin of the intermediate  $\Delta N$  minimum appears to be a physical phenomenon. The depths where the minima in  $\Delta N$  are observed, roughly coincide with the depths (~400 m) of occurrence of the Subantarctic Mode Water characterized by an oxygen maximum (WYRTKI, 1971, 1973; SEN GUPTA and NAQVI, 1984). However, as the intermediate oxygen maximum and  $\Delta N$  minimum do not occur everywhere in the region, it appears that the advection of Subantarctic Mode Water to the northern Arabian Sea is not continuous. The discontinuity may be brought about by eddies; evidence for the appreciably deep vertical extension of the eddies in the northern Arabian Sea has been provided by KESAVA DAS *et al.* (1980). Intermittent supply of dissolved oxygen, albeit limited, associated with the advection of the Subantarctic Mode Water to the stagnant, oxygen-depleted intermediate waters of the Arabian Sea perhaps prevents these waters from becoming completely anoxic.

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