

Nitrous oxide in the western Bay of Bengal

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Abstract

Extensive observations on nitrous oxide (N_2O) in the atmosphere and waters of the western Bay of Bengal during March–April 1991 yield surface saturations and atmospheric fluxes ranging from 89.3 to 213.9% (mean 125.2%), and from -0.10 to $10.67 \mu\text{mol } (N_2O)/\text{m}^2/\text{d}$ (mean $0.65 \mu\text{mol}/\text{m}^2/\text{d}$), respectively. The overall N_2O flux from the Bay of Bengal is estimated to be 0.027 – 0.077 Tg N/yr which is substantially lower than that computed for the Arabian Sea (0.19 – 0.28 Tg N/yr). Lower surface saturations and consequently smaller air–sea fluxes in the Bay of Bengal may result from strong stratification caused by the immense river runoff. The computed vertical exchange coefficient ($0.16 \text{ cm}^2/\text{s}$) at the top of the thermocline is about $1/3$ of the corresponding value ($0.55 \text{ cm}^2/\text{s}$) in the Arabian Sea. The associated smaller nutrient fluxes to the euphotic zone may be significantly responsible for the lower productivity of the Bay of Bengal. As expected, marked accumulation of N_2O occurs in subsurface layers of the Bay of Bengal with the total inventory of excess nitrous oxide (ΔN_2O) estimated as $\sim 5.4 \text{ Tg N}$. Since the Bay of Bengal is not an active water-column denitrification site, there appears to be no loss of N_2O through bacterial reduction. Vertical profiles of N_2O are characterized by a pronounced maximum at ~ 200 – 300 m which intensifies northward. The N_2O distribution seems to be influenced by the subsurface circulation. The relationship between ΔN_2O and the apparent oxygen utilization (AOU) is linear in three ranges: at $\text{AOU} < 150 \mu\text{M}$, from $\text{AOU} \approx 150 \mu\text{M}$ to the N_2O maximum (depth $\approx 300 \text{ m}$), and at depths $> \sim 1000 \text{ m}$. Within the depth range ~ 300 – 1000 m , however, ΔN_2O and AOU are not significantly correlated due to large changes in N_2O concentrations associated with small variations in the ambient oxygen levels.

1. Introduction

The Bay of Bengal may be one of the most important oceanic areas in terms of the cycling of nitrous oxide (N_2O) because it experiences a severe and widespread depletion of dissolved oxygen at mid-depths (Wyrki, 1971). However, unlike the Arabian Sea, the redox conditions within the oxygen minimum layer in the Bay of Bengal are just above those required to support denitrification (Rao et al., 1994–this issue). Such environments are ideally suited for an enhanced production of N_2O (Codispoti et al., 1992, and references

therein), and so the Bay of Bengal is expected to be a major reservoir of the oceanic N_2O .

Some recent studies undertaken in the Arabian Sea (Law and Owens, 1990; Naqvi and Noronha, 1991) and the eastern equatorial Indian Ocean (Butler et al., 1989) revealed high degrees of supersaturation of N_2O in surface waters and, consequently, large fluxes to the atmosphere. However, the absence of data from the Bay of Bengal has so far prevented a reliable estimation of the extent of N_2O emissions from the northern Indian Ocean as a whole. Such estimates are needed for a better quantification of the global oceanic N_2O flux in

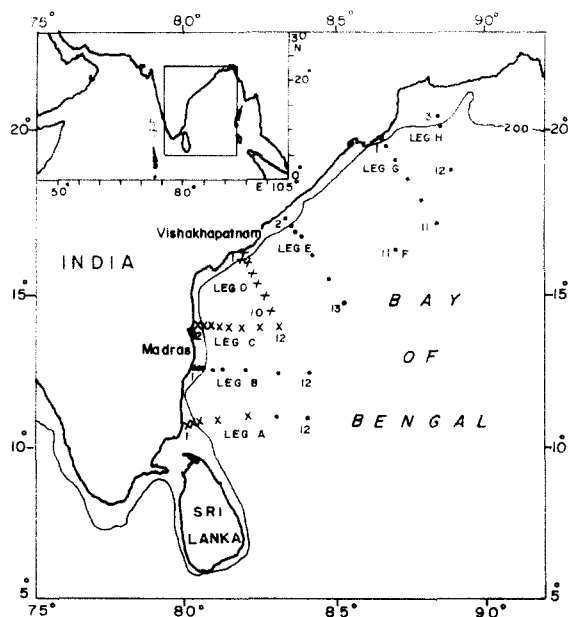


Fig. 1. Station locations. ● = stations where depth profiles of N_2O were obtained; × = stations where only surface observations were made.

view of the growing realization of the importance of the areas containing low-oxygen environments in determining the size of the overall oceanic N_2O flux (see Codispoti et al., 1992, for an overview).

In this paper, we present results of the first whole water column measurements of N_2O together with simultaneous measurements of oxygen and nutrients in the northeastern Indian Ocean. The data on nutrients and oxygen are described and discussed in detail elsewhere (Rao et al., 1994-this issue). Hydrography and circulation during the period of our study have been investigated by Shetye et al. (1993).

2. Material and methods

2.1 Sampling and analyses

Extensive measurements of N_2O in seawater and air were made during the 63rd cruise of ORV *Sagar Kanya* undertaken during the pre-monsoon period (March–April 1991). The station locations are shown in Fig. 1. Vertical profiles of N_2O were generated generally covering the entire water

column at 27 stations. At 21 other stations, the observations were restricted to the sea surface. Analyses of atmospheric N_2O were also carried out at most stations.

Air samples were collected in 1–1.2 l glass flasks from the bow of the ship immediately before arrival at the stations. Water samples were collected with Niskin samplers mounted on a CTD-rosette assembly (Seabird Electronics Inc., USA). Sub-samples (0.25–1.2 l) for the N_2O analysis were taken in ground-glass stoppered bottles. These samples were poisoned using saturated $HgCl_2$ solution (2 ml for 1 l of the sample), and stored in a refrigerator. The extraction of dissolved N_2O was carried out in the same bottles after removing a small amount of the sample to allow the gas dispersion tube to be introduced. A correction was made for the headspace above the liquid in the gas-stripping system.

N_2O analyses were performed on board within 3 days of collection following the procedure of Rönner (1983) as described by Naqvi and Noronha (1991). A Perkin-Elmer gas chromatograph (model 3920B) equipped with an electron capture detector containing 10 mCi ^{63}Ni foil was used for the analyses. A standard gas mixture of 2500 ppm N_2O in N_2 (Alphagaz, France) was used for the calibration. Replicate analyses ($n = 10$) of a sample with a N_2O content of 45.9 nM yielded a coefficient of variation of $\pm 4.2\%$.

Sub-samples for the analyses of nutrients and oxygen (and occasionally for salinity measurements to evaluate the functioning of the CTD sensor) were also taken from the same cast as above and analyzed following standard procedures (Rao et al., 1994-this issue).

2.2 Computation of surface saturation and atmospheric flux

Measurements of N_2O in atmospheric samples gave an average mole fraction of N_2O in the air over the western Bay of Bengal as 303 ppb, which compares very well with recent measurements elsewhere over the oceans (Khalil and Rasmussen, 1988; Butler et al., 1989; Oudot et al., 1990). This value was utilized to compute the equilibrium concentrations of N_2O in the surface waters [$(N_2O)_{eq}$]

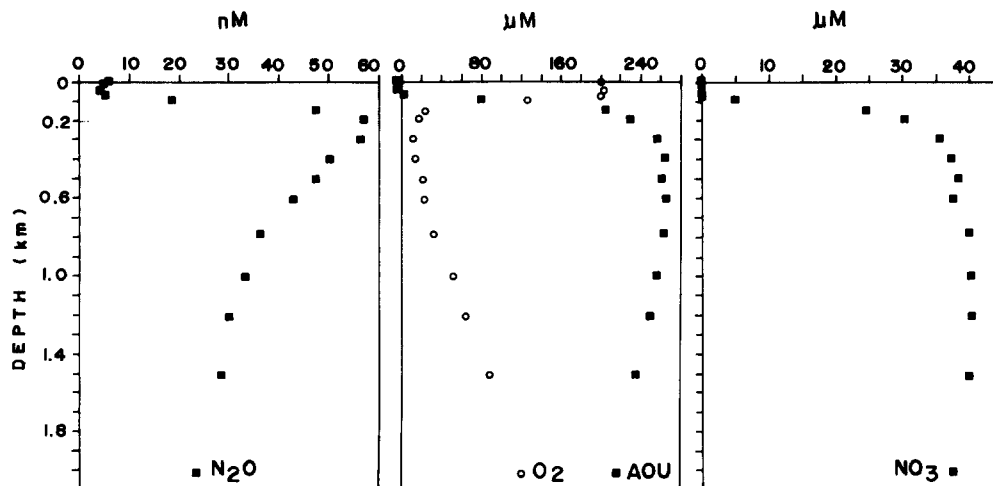


Fig. 2. A typical vertical profile of dissolved N_2O in the Bay of Bengal (Station A-11; latitude $10^\circ 59.83'N$, longitude $82^\circ 56.66'E$). Profiles of dissolved oxygen, AOU and nitrate are also shown for comparison.

from the solubility equation of Weiss and Price (1980). The equilibrium concentrations were then combined with the corresponding observed values $[(N_2O)_{obs}]$ to estimate the extent of surface saturation and the N_2O fluxes (F) to the atmosphere:

$$F = V[(N_2O)_{obs} - (N_2O)_{eq}] \quad (1)$$

The piston velocity, V , was computed from the Liss and Merlivat (1986) wind speed-transfer velocity relationship, assuming similar transfer velocities for carbon dioxide and N_2O as the molecular diffusivities of these gases are nearly equal (Broecker and Peng, 1974).

3. Results and discussion

3.1 Surface saturations and atmospheric fluxes

Surface waters in the Bay of Bengal were found to be generally supersaturated with N_2O except at 5 stations where slight undersaturation was observed. The degree of saturation varied from 89.3 to 213.9% with a mean ± 1 SD of $125.2 \pm 25.4\%$. The highest supersaturation occurred at station C-2, followed by station C-3 (161.2%). High saturation anomalies at these stations were associated with weak coastal

upwelling as evident from the physical, chemical and biological data. For example, surface water at C-3 was approximately $1.5^\circ C$ cooler as compared to the offshore stations, and the $26^\circ C$ isotherm lay at ~ 20 m at C-2, ~ 30 m at C-3, and at depths > 50 m at other stations. Similarly, significantly higher nutrient concentrations were observed within the euphotic zone at C-2 and C-3 leading to high chlorophyll concentrations (Dr. H.R. Gomes, pers. commun., 1991). Also, a pCO_2 exceeding $400 \mu atm$ was observed at the sea surface at C-3. These observations show how slight changes in the hydrographical conditions can bring about potentially large changes in the fluxes of greenhouse gases across the air–sea interface in the northern Indian Ocean.

The mean saturation anomaly observed in the Bay of Bengal (25%) is substantially higher than the global average (4%) obtained by Weiss (1978) from the analyses of 4200 samples collected from various oceanic areas. Butler et al.'s (1989) observations in the eastern equatorial Indian Ocean and the western Pacific during an El Niño year led to an even lower value (2.5%). However, their results from the eastern equatorial Indian Ocean (saturation anomaly $\sim 37\%$) compare well with our data. When compared to results from the Arabian Sea, where the N_2O supersaturations average around 86% (Law and Owens, 1990; Naqvi

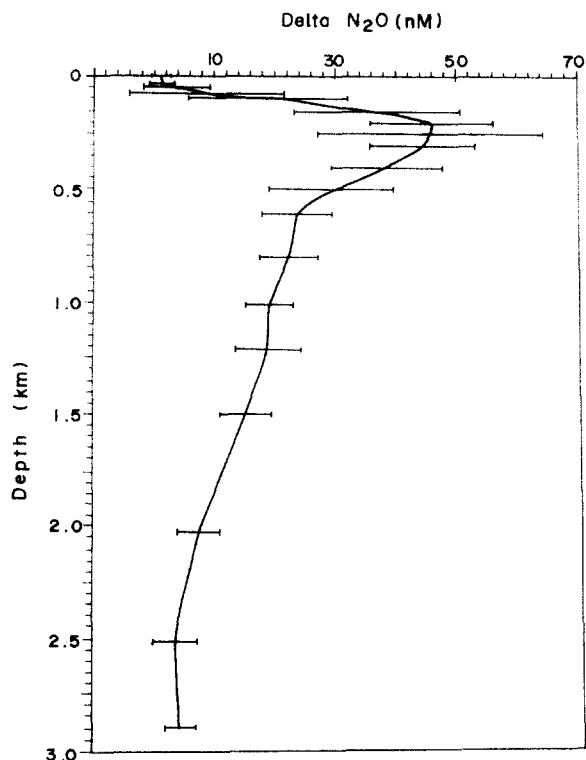


Fig. 3. Average profile of excess N_2O (nM) in the Bay of Bengal. The error bars represent 1 SD ($n = 27$ stations).

and Noronha, 1991), the saturation anomaly in the Bay of Bengal is much lower.

The flux of N_2O from the ocean to the atmosphere based on monthly means for March and April from the climatology of Hastenrath and Lamb (1979) (wind data in digital form made available by Prof. S. Hastenrath) ranged from -0.10 to $10.67 \mu\text{mol } (N_2O)/\text{m}^2/\text{d}$, with an average of $0.65 \pm 2.00 \mu\text{mol}/\text{m}^2/\text{d}$. This is much smaller than the atmospheric fluxes computed for the Arabian Sea [$4.46 \pm 2.60 \mu\text{mol}/\text{m}^2/\text{d}$ by Naqvi and Noronha (1991) and 8.64 ± 4.32 to $15.51 \pm 7.66 \mu\text{mol}/\text{m}^2/\text{d}$ by Law and Owens (1990)]. Reflecting the combined effect of a smaller supersaturation and weaker winds, this would correspond to an overall emission rate of N_2O from the Bay of Bengal (area $4.087 \times 10^6 \text{ km}^2$) of 0.027 Tg N annually. However, this most likely is an underestimate since the mean winds during March–April are quite weak and not representative of the whole year. In order to fix an

upper limit on the extent of N_2O emissions from the Bay, we have used a mean annual wind speed of 5 m/s to arrive at the net N_2O flux of 0.077 Tg N/yr . Thus, it would appear that unlike the Arabian Sea (which occupies about 50% more area than the Bay of Bengal), where the overall N_2O flux has been estimated to range between 0.19 and 0.28 Tg N/yr (Law and Owens, 1990; Naqvi and Noronha, 1991; Naqvi et al., 1993), the Bay of Bengal is a much weaker source of atmospheric N_2O .

Assuming that N_2O concentrations within the surface layers were regulated by turbulent fluxes from the thermocline, and that a steady-state existed whereby the N_2O input from the subsurface to surface waters was balanced by its escape to the atmosphere, we estimated the vertical exchange coefficient required to sustain the computed atmospheric N_2O flux. Using data from all the stations we determined the average N_2O concentrations at 7 depths between 30 and 250 m and fitted these to a polynomial to determine the N_2O gradient at the 100 m level as $0.468 \mu\text{mol}/\text{m}^4$. Combining this value with the computed N_2O flux, the vertical diffusivity was calculated as $0.16 \text{ cm}^2/\text{s}$. This value is less than $1/3$ of the corresponding value ($0.55 \text{ cm}^2/\text{s}$) for the Arabian Sea (Naqvi and Noronha, 1991). A slower rate of exchange between the surface and subsurface waters in the Bay of Bengal as implied by the N_2O data is perfectly compatible with a much stronger near-surface vertical stratification owing to the near-estuarine character of the Bay. A more vigorous vertical mixing in the Arabian Sea probably sustains larger fluxes of nutrients to the euphotic zone, thereby contributing to column productivity rates that are on average a factor of three higher than those in the Bay of Bengal (Qasim, 1977).

3.2 N_2O distribution at depth

Unlike most parts of the northern Arabian Sea, where two maxima in N_2O are observed separated by a minimum that is associated with the secondary nitrite maximum (SNM), vertical profiles of dissolved N_2O in the Bay of Bengal generally exhibit a single pronounced maximum (e.g. Fig. 2). Since the oxygen concentrations in the Bay of Bengal approach but do not reach suboxia, as evident

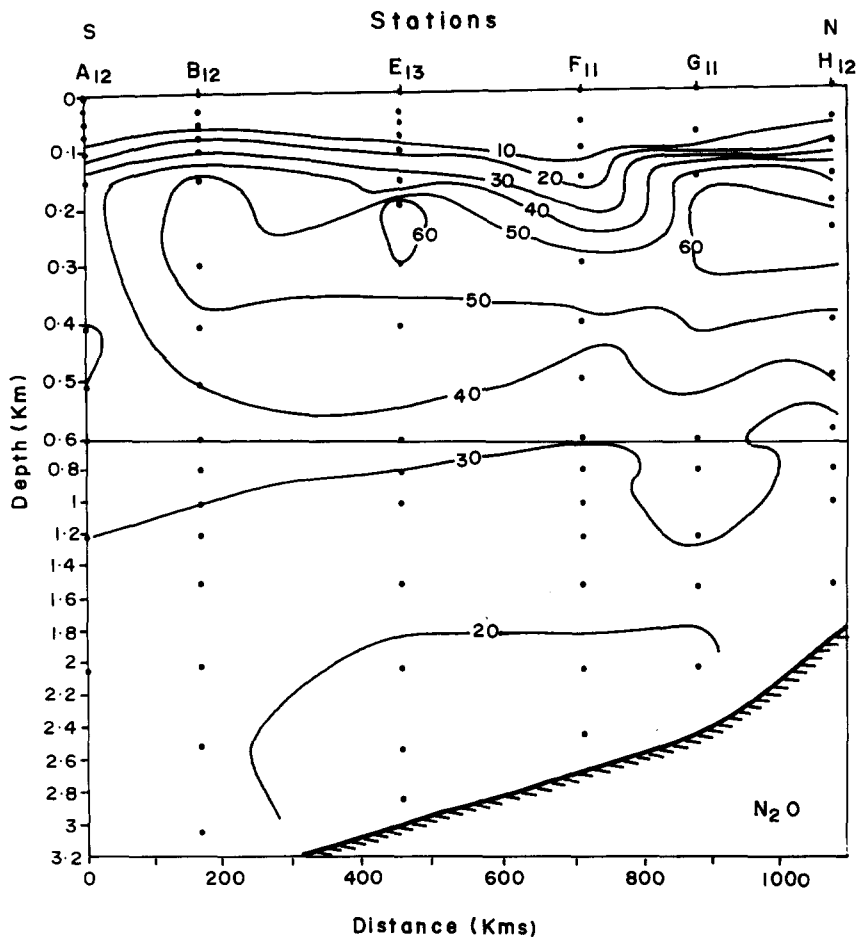


Fig. 4. Distribution of N_2O (nM) along a transect oriented approximately in N–S direction.

from the absence of the SNM (Rao et al., 1994—this issue), it is very unlikely that any loss of N_2O through bacterial reduction to molecular nitrogen occurs within the water column in this region. The maximal N_2O concentrations are among the highest observed in the oceans. As stated earlier, this is expected since the Bay of Bengal offers ideal conditions for N_2O accumulation in the water column. In order to estimate the total amount of ΔN_2O (that is the quantity in excess of the corresponding saturation value), we have determined the average profile of ΔN_2O (Fig. 3) and integrated the area under the profile, yielding an inventory of 1.32 g N/m^2 . The overall inventory of excess N_2O for the Bay of Bengal amounts to $\sim 5.4 \text{ Tg N}$.

Spatial variability of dissolved N_2O has been

investigated through vertical sections along a transect oriented roughly in the N–S direction and three transects (Legs B, E and G) normal to the Indian coast (Figs. 4–7). Concentrations of N_2O within its maximum exhibit a steady increase towards the north. The highest concentration measured during the cruise ($\sim 80 \text{ nM}$) was at 200 m depth at Sta. G-9 (Fig. 7); this was associated with very low ($< 4 \mu\text{M}$) oxygen concentrations (Rao et al., 1994—this issue). Lower oxygen concentrations, often reaching the detection limit of the Winkler procedure (but still not low enough to trigger denitrification) coupled with higher rates of biological productivity (Qasim, 1977) and higher particle fluxes as measured by the sediment traps (Ittekkot et al., 1991) might result in the

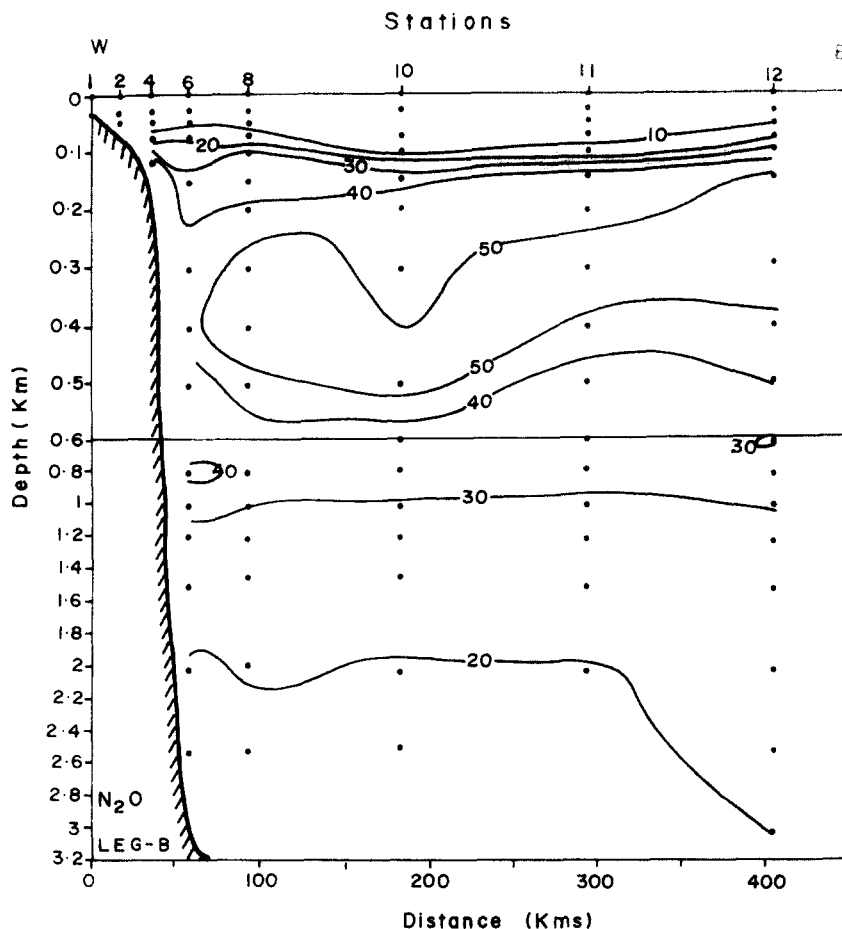


Fig. 5. Distribution of N_2O (nM) along Leg B.

observed enhanced N_2O accumulation in the northern Bay of Bengal. The sections oriented normal to the coast reveal the occurrence of lower N_2O concentrations off the continental margin than in the offshore waters, particularly along Legs B and G (Figs. 5 and 7). Similar decreases observed off the west coast of India have been attributed to the possible consumption of N_2O within the continental shelf/slope sediments (Naqvi and Noronha, 1991). However, a more effective renewal of intermediate waters off the continental margin could also account for the lower N_2O content of these waters (Dr. L.A. Codispoti, pers. commun., 1992). It may be pointed out that a western boundary current (WBC) was found to occur in the Bay of Bengal during the period of our observations

(Shetye et al., 1993). This current, forming a part of the seasonal subtropical gyre, carried warmer and more saline waters northward, and extended to the depth of the oxygen minimum as evident from the vertical sections of temperature, salinity, and density (Shetye et al., 1993). Its presence is also corroborated by the occurrence of invariably higher oxygen concentrations at the stations located just off the shelf break (Rao et al., 1994—this issue). Significantly, the highest N_2O concentrations along Leg G (Fig. 7) were observed closer to the continental margin as compared to Leg E (Fig. 6). This might have resulted from an offshore movement of the WBC along Leg G (Shetye et al., 1993). Given that the coastal circulation in the Bay of Bengal undergoes large seasonal changes, these

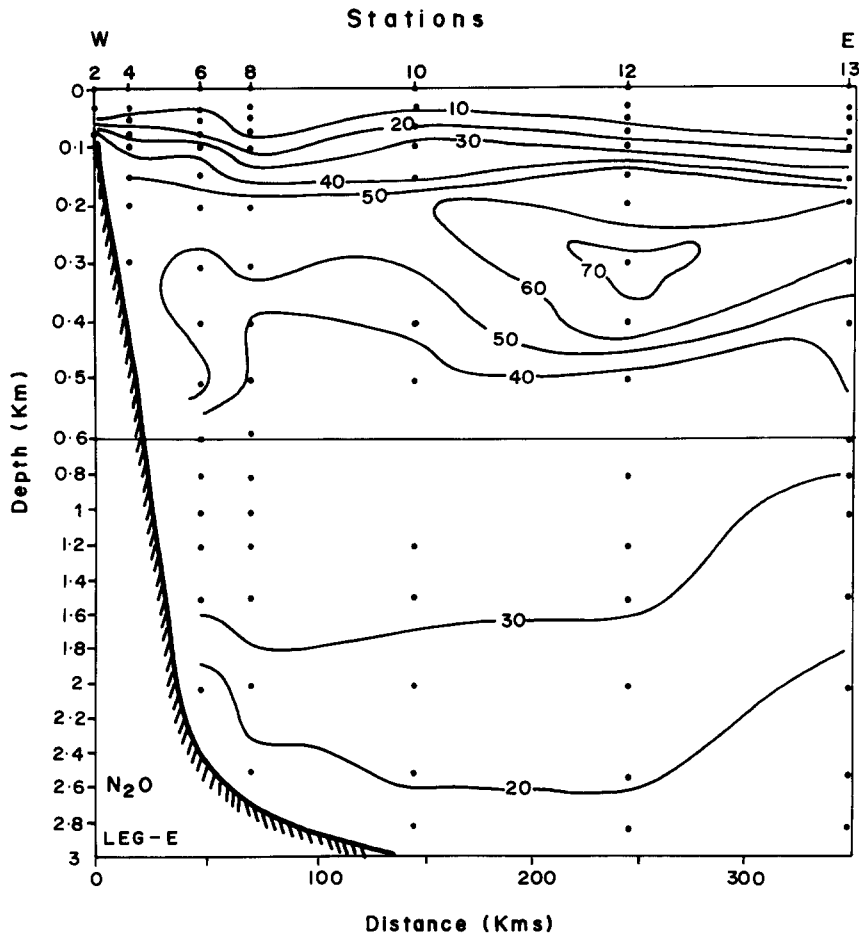


Fig. 6. Distribution of N_2O (nM) along Leg E.

observations would imply that appreciable short-term changes in the N_2O distribution may occur in the intermediate waters of the region.

3.3 N_2O – O_2 relationship

Vertical profiles of N_2O and O_2 in the Bay of Bengal show striking negative correlations (e.g. Fig. 2) as in other oceanic areas (Yoshinari, 1976; Cohen and Gordon, 1978; Elkins et al., 1978; Rönner, 1983; Codispoti and Christensen, 1985; Cline et al., 1987; Butler et al., 1989; Oudot et al., 1990; Law and Owens, 1990; Naqvi and Noronha, 1991). However, a plot (Fig. 8) between the apparent N_2O production (ΔN_2O) and the apparent oxygen utilization (AOU) shows that

distinct linear relationships between ΔN_2O and AOI may exist within three ranges: at $AOI < 150 \mu M$, from $AOI \approx 150 \mu M$ to the N_2O maximum (depth ≈ 300 m), and at depths $> \sim 1000$ m. The correlation between ΔN_2O and AOI was statistically insignificant in the depth range ~ 300 – 1000 m.

A change in slope of the ΔN_2O –AOI regression line seen at $AOI \approx 150 \mu M$ has also been reported from the Arabian Sea, albeit at slightly higher AOI ($197 \mu M$). It probably arises from the aforementioned enhancement in production of N_2O in the non-reducing oxygen-depleted waters. At present, there is considerable disagreement in the literature as to the exact mechanism(s) responsible for this increase. The low oxygen concentrations have

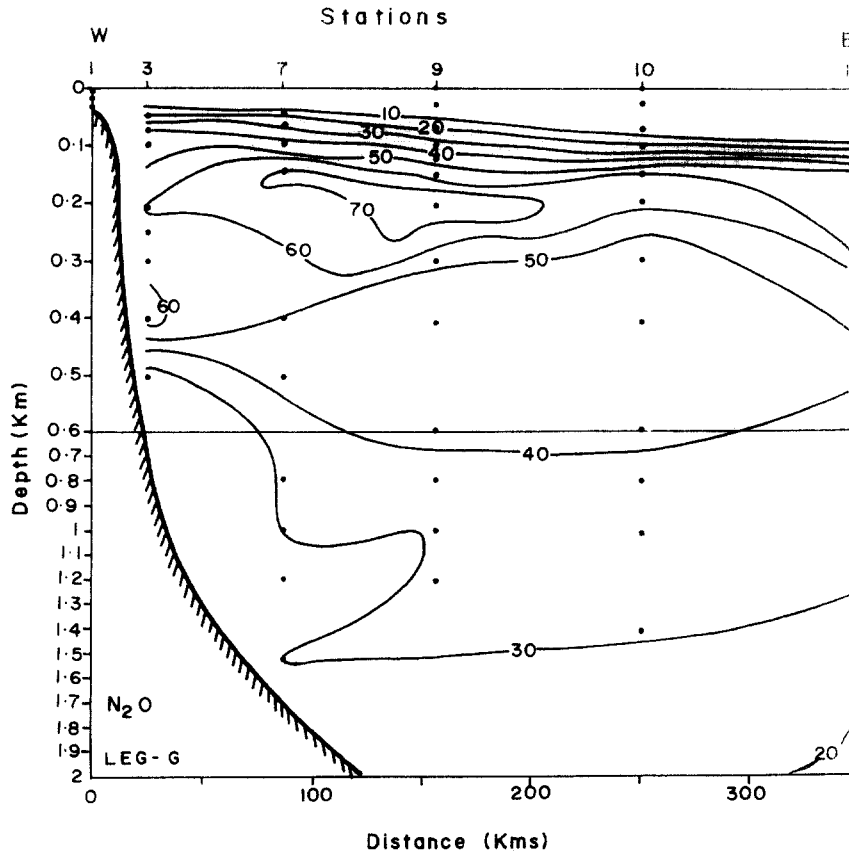


Fig. 7. Distribution of N_2O (nM) along Leg G.

been known to cause a higher yield of N_2O during nitrification (Goreau et al., 1980). This has been widely believed to be the principal reason for the accumulation of N_2O in the low-oxygen waters (Cohen and Gordon, 1978; Elkins et al., 1978; Codispoti and Christensen, 1985). On the other hand, it has also been observed that, at oxygen concentrations slightly above those prevailing within the cores of the suboxic zones ($> \sim 3 \mu M$), a net production of N_2O may occur through denitrification under carbon limiting conditions (Betlach and Tiedje, 1981). The oxygen levels in the Bay of Bengal may perfectly suit this pathway, but the relative significance of nitrification and denitrification in the N_2O production cannot be evaluated from the available data. It should be pointed out here that recent results on the isotopic composition ($^{15}N/^{14}N$ and $^{18}O/^{16}O$) of the oceanic N_2O challenge the prevailing views about its origin

(Kim and Craig, 1990; Yoshida et al., 1989; Naqvi, 1991; Naqvi and Noronha, 1991; Codispoti et al., 1992). Obviously, more work is required to settle the current disagreement concerning the mechanisms of N_2O production in the ocean.

An anomalous aspect of the N_2O – O_2 relationship in the Bay of Bengal is that over a wide depth range (~ 300 – 1000 m), ΔN_2O and AOU are apparently unrelated. This primarily results from the sensitivity of N_2O production to small changes in the ambient oxygen concentrations when the latter fail to near-zero levels. Thus, while the N_2O concentration varies greatly (with the peak values associated with the oxygen minimum at 200–300 m), the AOU remains more or less constant within the above depth interval (Fig. 2).

As stated above, a linear correlation between ΔN_2O and AOU also exists at depths > 1000 m. The deep waters in the Bay of Bengal could be

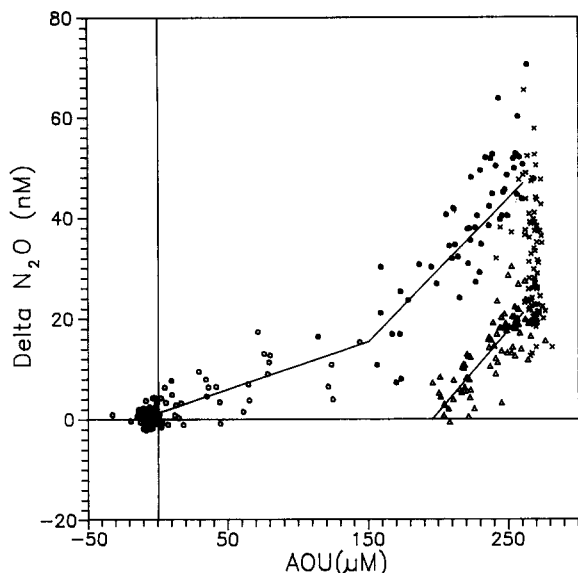


Fig. 8. Relationship between the apparent N_2O production (ΔN_2O) and apparent oxygen utilization (AOU). The straight lines represent least-square fits for three ranges: (1) $AOU < 150 \mu M$ (\circ); (2) $AOU \sim 150 \mu M$ to N_2O maximum (depth ~ 300 m; \bullet); and (3) depths $> \sim 1000$ m (Δ). The correlation was insignificant within the depth range ~ 300 – 1000 m (\times).

formed through binary mixing involving the bottom water and the North Indian high-salinity intermediate water (Rao et al., 1994-this issue). Hence their N_2O content may be determined to a large extent by the mixing of the high- N_2O intermediate water with the low- N_2O bottom water. It may be noted here that the N_2O levels in deep waters are substantially lower than those expected from the observed AOU. One may expect ΔN_2O of about 20 nM for an AOU of 200 μM (Broecker and Peng, 1982); in contrast, N_2O concentrations in the deepest waters are very close to the corresponding saturation values. This clearly indicates that N_2O in deep waters may not be as inert as believed so far, and its consumption (probably within the sediments) could account for the observed anomaly.

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