

EARLY DIAGENETIC PROCESSES AFFECTING NUTRIENTS IN THE PORE WATERS OF CENTRAL INDIAN OCEAN CORES

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Abstract

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Pore-water nutrients, nitrite, nitrate, phosphate, silicate, pH and solid-phase organic carbon were analysed for one core from the Arabian Sea and three cores from the manganese nodule area in the Central Indian Ocean Basin. Possible denitrification and unoxidized organic carbon are observed in the Arabian Sea core, while nitrification with intermediate low nitrate and high organic carbon are found in the mottled zones of the pelagic cores. Mobilization of iron-host oxyhydroxides and peaked profiles of organic carbon are related to a gradual increase in the phosphate profiles. The asymptotic silica values are less than the 100% saturation levels of amorphous silica and they fall outside the stability field of sepiolite.

Introduction

Pore-water studies play an important role in understanding the diagenetic reactions taking place at the sediment–water interface. Determination of nutrients in the interstitial waters also helps in understanding the nature and sequence of oxidation, the nature of oxidants and their role in organic matter diagenesis. In addition to revealing the nature of dominant diagenetic reactions, detailed pore-water studies also help in estimating the diffusive fluxes and geochemical balances. A number of studies have been undertaken in recent years in the Eastern Equatorial Atlantic, Eastern Equatorial Pacific, the MANOP sites, on the Madeira abyssal plain, on the Nares abyssal plain, in the South Atlantic and in the North Atlantic (Bender et al., 1977; Grundmanis and Murray, 1977; Sayles, 1979; Emerson et al., 1980; Jahnke et al., 1982; Colley et al., 1984; De Lange, 1986) to under-

stand the role of nutrients in early diagenetic processes. Pore-water nutrients have not been studied in sediments of the Indian Ocean (Manheim, 1976). The present investigation is an attempt to study the behaviour of various chemicals across the sediment–water interface and to understand the early diagenetic processes controlling the nutrients in pore waters of one core from the Arabian Sea and three cores from the Central Indian Ocean Basin.

Materials and methods

During cruise 20 of the O.R.V. *Sagar Kanya* one box core from the Arabian Sea and three spade cores from the Central Indian Ocean Basin were collected for the study of interstitial water (Fig.1). Samples were squeezed from the sediments onboard using Manheim's (1966) sediment squeezer.

The box core was subsampled onboard and

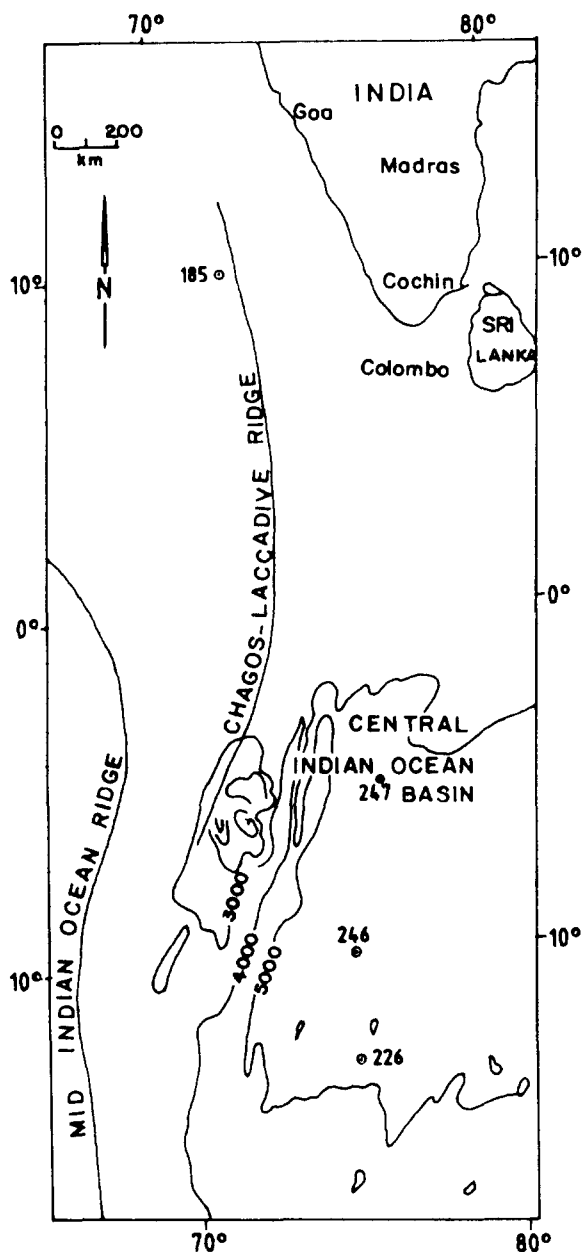


Fig.1. Sampling stations.

squeezing of the sediment was carried out immediately. The interstitial water was collected with the help of a disposable syringe and stored in acrylic bottles in a refrigerator (temperature around 2°C). This subsampled pore water was used for the nutrient analysis onboard. The nutrients, phosphate, silica, nitrate and nitrite, were measured using a

Skalar-400 Auto Analyser (the methods are as described in the Skalar manual). The accuracy of the nitrate and nitrite values is 0.1% for cores 185, 226, and 246 and 8–10% for core 247. For the phosphate in all the cores the accuracy is 2.5–5.0%. The accuracy for silica is 0.82% for cores 226 and 246 and 10% for core 247. The pH was also measured onboard, with Beckman's digital pH meter, immediately after the recovery of the pore water (Table 1).

Organic carbon was determined by the wet oxidation method (El Wakeel and Riley, 1957). The accuracy of the analysis is $\pm 0.01\%$ C.

Core locations and description

A box core (Station 185) was collected in the Arabian Sea on the continental margin at 10.020°N, 71.872°E at a depth of about 2560 m (Fig.1). The total core length is 3.3 m. This core was subsampled at different intervals to obtain the interstitial water. Thirteen subsamples were collected down to a depth of 2.0 m using an acrylic sediment syringe. This core of calcareous ooze shows different shades of grey throughout its length. Colour classification has been carried out following the USGS standard rock colour chart. The top 25 cm of the core represents pelagic yellowish grey foraminiferal ooze. The remaining part of the core has a texture varying from silty clay to clayey silt (Pattan and Rao, pers. commun., 1988).

The other three (spade) cores were collected south of the equator in the Central Indian Ocean Basin. Core 226 was collected at 13.087°S, 75.019°E in the siliceous ooze area (Udintsev, 1975) at a depth of 5270 m. Colour classification revealed pale yellowish brown sediments with intercalations of moderate yellowish brown sediments, this intercalation indicating the effect of bioturbation. The length of the core is 32 cm (Fig.2A).

Core 246 was retrieved at 10°S, 75°E at a depth of 5160 m (Fig.1) and is also from the siliceous ooze area. The core top was slightly compressed due to deeper penetration. Based on the colour classification it has been divided

TABLE 1

pH and nutrients in pore water and organic carbon in sediments

| Depth (cm) | pH | Nitrite ($\mu\text{g}_\text{at}/\text{l}$) | Nitrate ($\mu\text{g}_\text{at}/\text{l}$) | Phosphate ($\mu\text{g}_\text{at}/\text{l}$) | Silica ($\mu\text{g}_\text{at}/\text{l}$) | Organic carbon (%) |
|---|------|---|---|---|--|-----------------------|
| <i>13.087° S, 75.019° E; core 226, depth 5270 m</i> | | | | | | |
| 0-3 | 7.98 | ND | 12.5 | 23.13 | 500 | 0.673 |
| 3-6 | 8.10 | ND | 25.0 | 25.63 | 569 | 0.311 |
| 6-11 | 7.90 | ND | 37.5 | 26.25 | 588 | 0.224 |
| 11-14 | 8.05 | ND | 31.25 | 30.00 | 694 | 0.380 |
| 14-20 | 7.95 | 0.63 | 31.25 | 31.25 | 663 | 0.328 |
| 20-24 | 7.99 | ND | 31.25 | 26.25 | 681 | 0.172 |
| 24-27 | 7.99 | ND | 37.5 | 26.25 | 675 | 0.086 |
| 27-32 | 7.80 | 1.25 | 37.5 | 32.50 | 744 | 0.138 |
| <i>10° S, 75° E; core 246, depth 5160 m</i> | | | | | | |
| 0-3 | 7.90 | ND | 31.25 | 23.13 | 475 | 0.380 |
| 3-6 | 7.82 | ND | 37.50 | 23.13 | 525 | 0.276 |
| 6-11 | 7.72 | ND | 43.75 | 22.50 | 481 | 0.069 |
| 11-15 | 7.80 | ND | 43.75 | 23.75 | 531 | 0.190 |
| 15-18 | 7.79 | 1.25 | 43.75 | 25.63 | 588 | 0.224 |
| 18-25 | 7.82 | 0.63 | 37.50 | 25.63 | 588 | 0.603 |
| 25-30 | 7.82 | ND | 43.75 | 24.38 | 631 | 0.310 |
| 30-35 | 7.59 | ND | 50.00 | 25.63 | 594 | 0.035 |
| 35-40 | 7.59 | ND | 50.00 | 26.88 | 631 | 0.190 |
| 40-44 | 7.60 | ND | 43.75 | 26.88 | 631 | 0.121 |
| <i>5° S, 76° E; core 247, depth 5170 m</i> | | | | | | |
| 0-3 | 7.80 | ND | 31.25 | 20.00 | 394 | 0.535 |
| 3-6 | 8.15 | ND | 25.50 | 23.13 | 456 | 0.380 |
| 6-11 | 8.15 | ND | 31.25 | 25.63 | 550 | 0.414 |
| 11-15 | 8.15 | ND | 25.00 | 23.75 | 513 | 0.310 |
| 15-20 | 8.30 | ND | 25.00 | 23.13 | 475 | 0.310 |
| 20-25 | 7.90 | ND | 25.00 | 25.63 | 544 | 0.396 |
| 25-30 | 8.00 | ND | 25.00 | 24.38 | 544 | 0.258 |
| 30-36 | 7.89 | ND | 25.00 | 23.13 | 544 | 0.207 |
| <i>10.020° N, 71.872° E; core 185, depth 2560 m</i> | | | | | | |
| 0-1 | 8.10 | 9.99 | 35.82 | | | |
| 1-3 | 8.05 | 3.33 | 21.66 | | | |
| 5-8 | 8.05 | ND | 16.66 | | | |
| 10-13 | 8.00 | ND | 16.66 | | | |
| 17-20 | 8.15 | ND | 14.16 | | | |
| 23-26 | 8.15 | ND | 8.33 | | | |
| 41-43 | 7.90 | ND | 9.16 | | | |
| 62-65 | 7.50 | ND | 9.99 | | | |
| 75-78 | 7.80 | ND | 7.49 | | | |
| 90-92 | 7.80 | ND | 4.99 | | | |

ND : Not detectable.

into six zones (Fig.2B). Ten subsamples were taken for interstitial water collection throughout the length of core.

Core 247 was collected at 5°S, 76°E at a depth

of 5170 m (Fig.1), being recovered from the approximate boundary between the terrigenous clay and the siliceous ooze. The core was 36 cm long and the sediment pale brown.

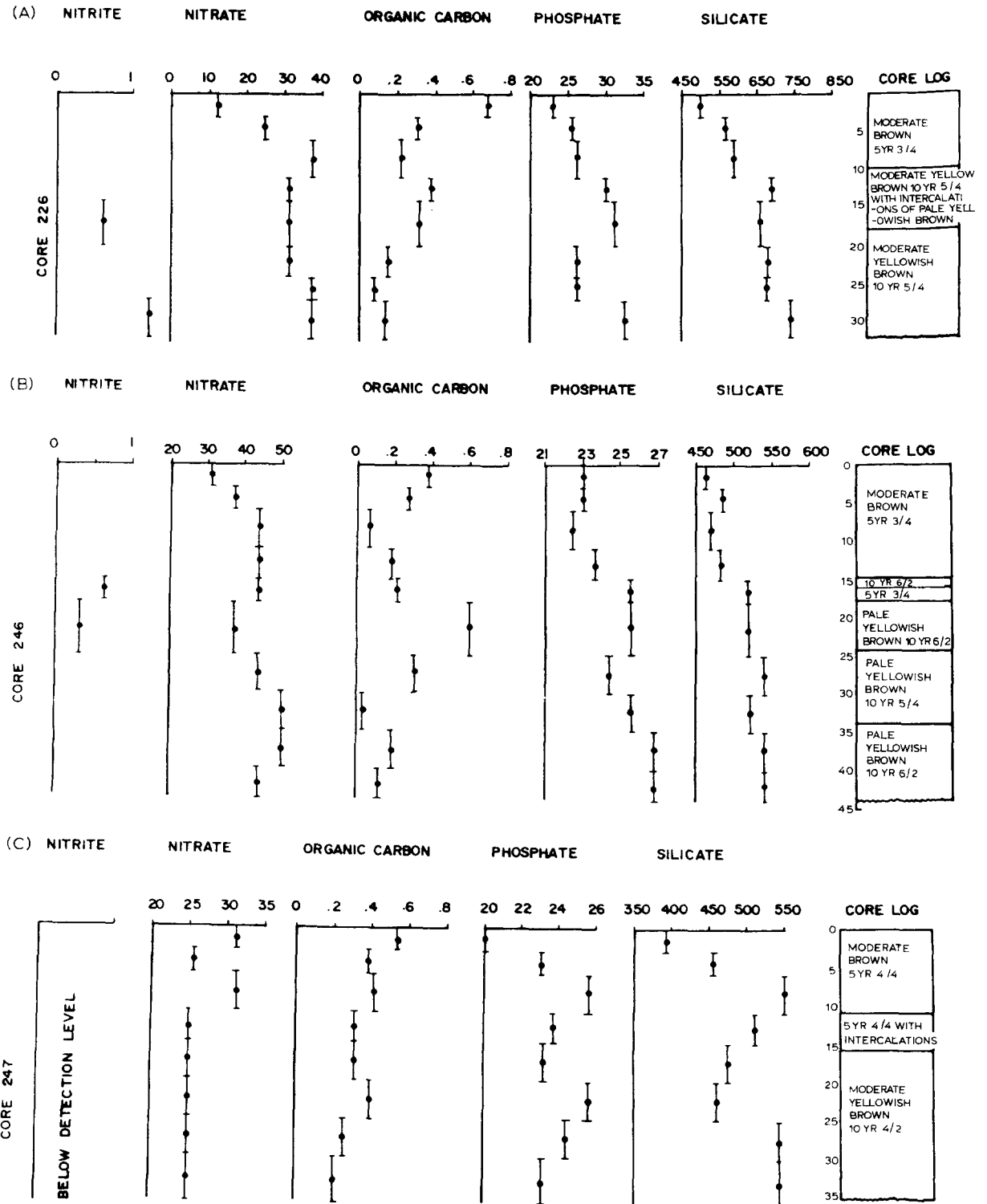


Fig. 2. Plots of pore-water data and organic carbon against depth in core. Core logs also shown. Nutrient data, $\mu\text{g}_\text{N}/\text{l}$; organic carbon, %C. A. Core 226. B. Core 246. C. Core 247.

Between 11 and 16 cm intercalations of dark brown clay were observed, and these appear as mottled structures effected by bioturbation (Fig.2C).

Results

The nitrate concentrations in the siliceous ooze cores 226 and 246 increase steadily downward indicating nitrification. Core 246 shows an increase in nitrate from 30 to 50 $\mu\text{g}_{\text{at}}/\text{l}$ at a depth of 40 cm (Fig.2B). Core 226 shows an even larger gradient from an initial 13 to 38 $\mu\text{g}_{\text{at}}/\text{l}$ at 30 cm (Fig.2A). On the other hand, the nitrate concentrations in core 185 diminish gradually to a value of 5 $\mu\text{g}_{\text{at}}/\text{l}$ at a depth of 90 cm (Table 1).

In most of the samples, nitrite is below detection levels. Nitrite could be detected only in the top few centimetres of core 185 and at intermediate levels in cores 226 and 246.

Dissolved phosphate values increase from the sediment–water interface with depth in all the cores. The extent of the increase varies from 5 to 9 $\mu\text{g}_{\text{at}}/\text{l}$ (Fig.2). Although the trend follows the results from other areas (Froelich et al., 1979; Jahnke et al., 1982; De Lange, 1984) the phosphate concentrations encountered in the present investigation are higher than those reported earlier. The MANOP Pore-Water Chemistry Group have found some discrepancy between phosphate from pore water sampled in situ and that squeezed from cores (Emerson et al., 1980). Bray et al. (1973) have found significant variations in inorganic phosphate due to the oxidation effect.

Pore-water silica profiles show a gradual increase with depth. In all cores from this area the interstitial silica value of the topmost layer is much higher than that in the bottom waters (130 $\mu\text{M}/\text{kg}$) (Spencer et al., 1982). In core 226 the values reach as much as 750 $\mu\text{g}_{\text{at}}/\text{l}$ at a depth of 30 cm (Fig.2A) and in cores 246 and 247 the maximum attained was 631 and 550 $\mu\text{g}_{\text{at}}/\text{l}$ at 27 and 8 cm respectively (Fig.2). The consistent silica increase in all the siliceous ooze cores is as expected and agrees well with the results in other siliceous ooze areas (Shishkina, 1972;

Heath, 1974; Sayles, 1981; Wakefield, 1982). The maximum silicate concentrations range between 545 and 740 $\mu\text{g}_{\text{at}}/\text{l}$ in these samples. The asymptotic values are found below a depth of around 25 cm. The silica values for the calcareous ooze core 185 were quite discrepant owing to long exposure to the atmosphere and are not included in Table 1. Similar observations were made by Setlock (1979) during his experimental work, wherein a considerable difference in silica concentrations was found after warming the cores for 9 h. This was attributed to the desorption of silica from the sediment particles. A similar conclusion was drawn by Fanning and Pilson (1971). It is worthwhile mentioning here that in the same experimental work, no differences were found in the nitrate values.

Amorphous or opaline silica was calculated in the top sediment layers (0–3 cm) following Böstrom et al.'s. (1973) method. The values are 27, 28 and 15% respectively for cores 226, 246, and 247.

The organic carbon content in the solid phase in all three pelagic cores ranges from 0.03 to 0.67%. The depth profiles show a considerable similarity to phosphate and a distinct mirror image concerning the nitrate data.

Discussion

Some of the organic matter, CaCO_3 and opaline silica, which formed in the surface water due to biological productivity settles to the bottom where it undergoes decomposition and dissolution processes. Organic matter decomposition follows a sequence of processes depending upon the availability of the oxidant. This involves the successive utilization of O_2 , NO_3 , MnO_2 , $\text{Fe}(\text{OH})_3$ and SO_4 for organic matter oxidation (Berner, 1982).

Nitrate and nitrite

In the present study, for cores 226 and 246 which totally comprised siliceous oozes, the initial increase in nitrate at the sediment

surface reflects oxygen respiration coupled with nitrification. Both cores have intermediate (10–18 cm in 226 and 18–25 cm in 246) layers showing mottling or bioturbation (Figs. 2A and B). This intermediate layer has a sharp fall in the nitrate values and detectable levels of nitrite, along with increased levels of organic carbon in the solid phase, indicating the effect of bioturbation and oxygen consumption. This shows that oxic and anoxic processes are active at different intervals in the core. The surface layer down to 18 cm in core 246 and down to 10 cm in 226 is undergoing oxic diagenesis while the subsurface layers (10–18 and 18–25 cm) indicate the existence of anoxic conditions and hence denitrification. Thus, one can envisage the process being controlled by the reaction rates and bioturbation of sediments (Berner, 1969, 1980). If we assume that all organic nitrogen is oxidized to NO_3 during O_2 reduction, the stoichiometric model of Emerson et al. (1980) suggests that the NO_3 concentration rises to 51–52 $\mu\text{M}/\text{kg}$. In this study the nitrate concentration rises to levels of 50 and 40 $\mu\text{g}_{\text{at}}/\text{l}$ in a depth range of 35–40 cm for cores 246 and 226 respectively.

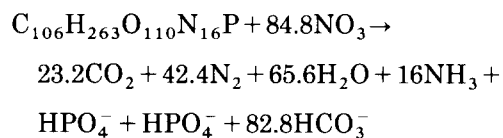
In the areas of siliceous ooze with the presence of significantly high concentrations of oxygen, ammonium is subsequently oxidized to nitrate:



In contrast, denitrification is indicated by decreasing nitrate concentrations and detectable values of nitrite in the top few centimetres of core 185. Increased nitrite, present as an intermediate product in seawater or pore waters, indicates either the oxidation of ammonium and/or the reduction of nitrate (Kiefer et al., 1976; Suess et al., 1980; Jahnke et al., 1982). The low nitrate levels in the interstitial fluid in core 185 may be attributed to biochemical consumption, since nitrate does not form any marine mineral phase (Setlock, 1979). This would indicate in turn that complete oxygen consumption occurred in the pore fluids and nitrate was utilized for organic matter degradation as a secondary terminal electron accep-

tor. Surface biological productivity is high in this area (300–400 t C/km²/yr) (Qasim, 1977) compared to other areas and thus creates a thin oxidising layer, typical over nearshore oxygen-poor sediments (Sholkovitz, 1973). In the same core considerable amounts of undissolvable black particles identified as organic matter were observed during teflon bomb digestion using aqua regia and HF (Pattan and Rao, pers. commun., 1988). This is obviously evidence for suboxic diagenesis, as could be seen from nitrate and nitrite profiles.

Evidently, the process occurring in core 185 could be denitrification as shown by the following reaction (De Lange, 1986), in which case the oxidation of organic matter is, however, incomplete:



Core 247 (in the transitional zone between siliceous ooze and terrigenous sediment) shows a zero gradient of nitrate from a depth of about 10 cm. Hence, there could be no diffusional activity occurring in this zone.

Phosphate

The downward increase in the phosphate profiles can be explained in three ways. One explanation could be the dissolution of phosphorous-bearing solid phases, while the other explanation could be one of anoxic diagenesis. Froelich et al., (1979) have attributed the dramatic increase in dissolved phosphate to the release of phosphate in pore waters during mobilization of iron-host oxyhydroxide coatings. Since the presence of iron oxyhydroxides is explained by the occurrence of iron-rich smectites in this area (Rao and Nath, 1986), it can thus be assumed that the mobilization of iron oxyhydroxides does facilitate an upward diffusive flux of phosphate into the overlying water. De Lange (1984) has correlated the sharp changes in the profile, marked by peaks, with the organic carbon content of the solid

phase. A close relationship can also be seen between the dissolved phosphate concentration and the organic carbon content of the sediment in the present cores (Fig.2).

Silica

The high interstitial values can be explained by the presence of biogenic silica and its dissolution and diffusion upwards from the deep layers. The average biogenic silica value for these three cores is 23%, which agrees with earlier observations (Lisitzin, 1967; DeMaster, 1979). The distribution and variation of dissolved silica in interstitial waters has been related to the productivity of the overlying waters in the North Atlantic (Schink et al., 1974, 1975) but this was contradicted by Sayles (1979) who found concentrations as low as 100–200 μM in the North Atlantic and attributed the silicate mineral equilibrium as a controlling factor. Wakefield (1982) checked the validity of this relationship with productivity by combining his data with those of Bischoff and Sayles (1972), and found no agreement because of the sparse data and marked changes in the productivity. Calculations following the method of Pedersen (1979) yielded average pore-water silica concentrations of 667, 587, 512 and 376 $\mu\text{g}_{\text{at}}/\text{l}$ for cores 226, 246, 247 and 185 respectively. The productivity of the surface waters is 25–50 $\text{t}^3 \text{C}/\text{km}^2/\text{yr}$ for stations 226 and 246, 50–100 t for station 247 and 300–400 t for station 185 (Qasim, 1977). The above values show no significant correlation between average interstitial silica and the primary productivity in the present study, which is in agreement with Sayles (1979).

The observed interstitial silica concentrations at stations 226, 246 and 247 are below the saturation levels with respect to amorphous silica. The experimental saturation levels reported are 1000 μM for acid-cleaned radiolaria (Hurd, 1973) and 830 μM for silica gel (Jones and Pytkowicz, 1973). The asymptotic or near-linear value for core 247 is 544 $\mu\text{g}_{\text{at}}/\text{l}$ at 20 cm and for core 246 at 25 cm it is 631 $\mu\text{g}_{\text{at}}/\text{l}$. These values are about 70% of the expected equilib-

rium solubility for amorphous silica calculated by Wollast (1974). The range of values observed is nearly equal to that reported from the South Atlantic (Sayles, 1981) and these values fall outside the stability field of sepiolite, a magne-

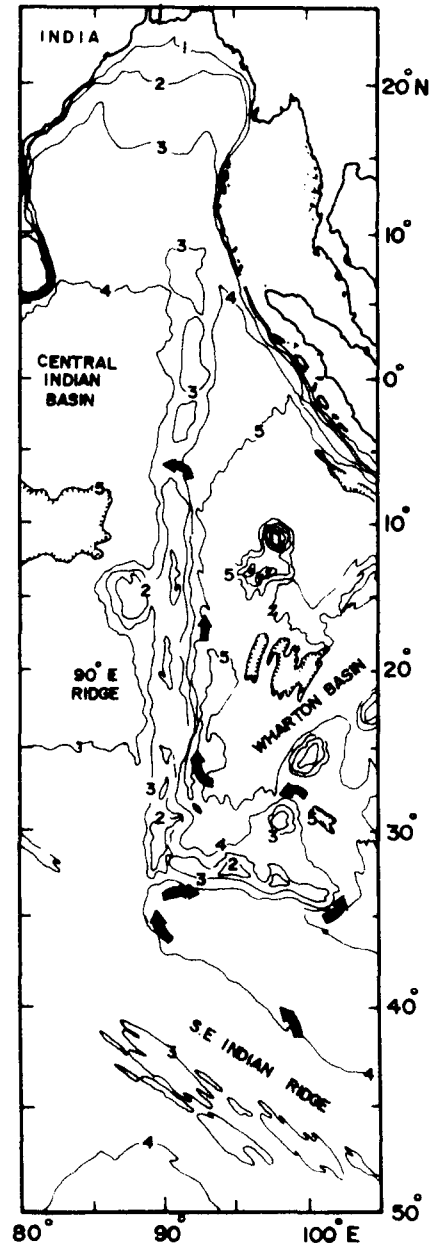


Fig.3. Bathymetric and oceanographic setting of the Eastern Indian Ocean. Arrows indicate the pathways of deep thermohaline currents. After Johnson and Nigrini (1982).

sium silicate which is a surface coating on amorphous silica and radiolarians; hence, the silica in our cores is undersaturated with respect to sepiolite.

Hydrographic effects on silica

Antarctic Bottom Water (AABW) crosses the Southeastern Indian Ridge and flows clockwise around the western margins of the South Australian Basin and Wharton Basin. It continues northwards along the base of the Ninety-East Ridge and enters the Central Indian Basin (Fig.3) (Johnson and Nigrini, 1982). Through these saddles AABW sinks into the basin owing to westward movement controlled by the bathymetry. This water becomes mixed with basin waters and thus the bottom-water temperatures are enhanced in the central and western parts of the basin. The potential temperature at the core locations is 1.03°C (Warren, 1982) at a depth of 4300 m, which is higher than at the Ninety-East Ridge.

Study of Brunt–Vaisala frequencies reveals that the deep-water stability in this region is low. This low stability leads to cyclonic shear, which is observed from the annual mean anomaly of the geopotential thickness of the 3000–4000 m layer (Levitus, 1982). This in turn creates a turbulence. These hydrographic parameters lead to dissolution of biogenic silica and enhanced values of interstitial silica (Lonsdale, 1976; Wakefield, 1982).

Conclusions

Early diagenetic processes control pore-water nutrient levels, as has been demonstrated for one core from the Arabian Sea and three cores from the Central Indian Ocean Basin. The Arabian Sea core has higher concentrations of organic carbon which consumes the oxygen and nitrate as primary and secondary electron acceptors and this leads to denitrification. The two deep-sea cores from the Central Indian Ocean Basin, on the other hand, show nitrification in the lithologically more uniform upper layers. At intermediate depths the bioturbated zones have high con-

tents of organic carbon which is probably not fully decomposed and shows signs of denitrification. One core, which lies on the boundary between terrigenous and siliceous sediments, has no diffusional gradient of nitrate in the deeper layers.

The increasing phosphate values with depth are controlled by the mobilization of iron oxyhydroxides.

The silica values are high but below the saturation levels of amorphous silica. The low stability of the bottom water in this region enhances the dissolution of biogenic silica and leads to the upward diffusive flux of interstitial silica.

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