

Calcium : chlorinity ratio and carbonate dissolution in the northwestern Indian Ocean

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(Received 22 March 1982; in revised form 20 August 1982; accepted 20 October 1982)

Abstract—Analysis of 128 water samples from the northwestern Indian Ocean gave an average calcium : chlorinity ratio of 0.02164 ± 0.000009 . An identical ratio (0.02165) was obtained using the data from selected *Meteor* stations in the region. The ratio in the northern Indian Ocean is significantly higher than the oceanic averages. The northern Indian Ocean, occupying only 3% of the global oceanic area, receives about 9% of the total global river runoff annually. It is suggested that the high calcium flux from the rivers in a relatively small area increases the Ca : Cl ratio in the upper layers of the region. The ratio in the deep waters in the region is also higher than in other oceanic areas, presumably due to a high rate of calcium removal at the surface by biological activity and its subsequent input to the bottom waters. A more rapid upward movement of deep waters in the Indian Ocean than in the Pacific and Atlantic oceans appears to result in a faster upward flux of calcium in the region. The ratio increases below the lysocline.

The calcite saturation depth lies between 2500 and 3000 m. Carbonate content of the sediments, however, shows no appreciable decrease above 4000 m, which supports kinetic models for CaCO_3 dissolution. The data support the Broecker–Takahashi model (*Deep-Sea Research*, 25, 65–91, 1978), indicating that variations in the lysocline depth can be accounted for largely by variations in carbonate ion concentration. The “critical carbonate ion concentration” for the region is $90 \pm 5 \mu\text{M kg}^{-1}$. It is concluded that calcite dissolution does not occur above 4000 m.

INTRODUCTION

THE MAJOR constituents of seawater generally bear a constant ratio with chlorinity, but in some areas the ratios may differ significantly (CULKIN, 1965; WILSON, 1975). Studies on the calcium : chlorinity ratio in several parts of the northern Indian Ocean have yielded consistently high values (SEN GUPTA, NAIK and SINGBAL, 1978; NAQVI and REDDY, 1979; NORONHA, MORAES and SEN GUPTA, 1981). In this paper we examine factors that increase the ratio in the region and that control the dissolution of CaCO_3 in the oceans, to explain the observed trends in the variations of the Ca : Cl ratio with depth.

MATERIAL AND METHODS

The study is largely based on observations during the 86th and 87th cruises of R.V. *Gaveshani* in the northwestern Indian Ocean in January to February 1981. The data collected by R.V. *Meteor* in 1965 in the Arabian Sea have also been used for comparison. The station locations are given in Fig. 1.

Water samples during the *Gaveshani* cruises were collected from standard depths with metallic Nansen bottles and Niskin samplers. Sediment samples were obtained with a

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The analytical procedures followed during *Meteor* cruises are described in the cruise reports (DIETRICH, DÜING, GRASSHOFF and KOSKE, 1966).

Computation of the degree of calcite saturation

Carbonate ion concentration was computed from the alkalinity and pH data. Apparent dissociation constants used were those determined by MEHRBACH, CULBERSON, HAWLEY and PYTKOWICZ (1973) for carbonic acid and by LYMAN (1957), as expressed by LI, TAKAHASHI and BROECKER (1969), for boric acid. The pressure effects were calculated according to CULBERSON and PYTKOWICZ (1968). The apparent solubility product (K'_{sp}) of calcite and its pressure dependence were calculated according to INGLE (1975). The ionic product was divided by the apparent solubility product to obtain the degree of saturation

$$\Omega = (\text{Ca}^{2+})(\text{CO}_3^{2-})/K'_{sp}(P).$$

We estimate the pH measurements to be accurate to within at least ± 0.02 pH units, which corresponds to an uncertainty in the computed value of Ω of $\pm 5\%$ due to pH and $\pm 1\%$ due to alkalinity at a depth of about 2000 m. EDMOND and GIESKES (1970) reported that the greatest uncertainty in the computed values of Ω arises from the uncertainty in the apparent solubility product (K'_{sp}). The values of K'_{sp} for calcite used by us (INGLE, 1975) are in close agreement with the values of MORSE, MUCCI and MILLERO (1980). Further, the effect of pressure on K'_{sp} as reported by INGLE (1975) has been confirmed by MILLERO (1982). Thus, the uncertainty in K'_{sp} has been considerably reduced. The errors associated with the dissociation constants are small (EDMOND and GIESKES, 1970). Therefore, the overall uncertainty in Ω calculated by us should be well within $\pm 10\%$.

RESULTS AND DISCUSSION

The average calcium (g kg^{-1}) to chlorinity ($\times 10^3$) ratio for all samples analyzed by us was 0.02164 with a standard error of ± 0.000009 and an average calcium concentration of 420.6 mg kg^{-1} . A remarkably similar ratio (0.02165) was obtained using the *Meteor* data; the calcium concentrations were converted to weight ratios for the calculations. The present value is close to that reported by NAQVI and REDDY (1979) for the Laccadive Sea (0.02168), but it is considerably higher than the oceanic average values of 0.2126 by CULKIN and COX (1966) and 0.02128 by RILEY and TONGUDAI (1967). SHILLER and GIESKES (1980) obtained a ratio of 0.02109 for standard seawater. Thus, the present ratio is higher than the world oceanic average by 1.7% and that of standard seawater by 2.6%. Still higher ratios were reported by SEN GUPTA *et al.* (1978) (0.02229) and NORONHA *et al.* (1981) (0.02224) from areas in the northern Indian Ocean where massive dilution by river runoff occurs. Numerous reports with consistently high Ca : Cl ratios from the northern Indian Ocean rule out the possibility of systematic error, and it can be concluded that the ratio is indeed higher in the region than in other areas.

The values of the Ca : Cl ratio for all stations have been pooled and averaged for different depth ranges (Table 1) to reduce the bulk of the data and minimize the effects of random errors. In general, the *Meteor* and *Gaveshani* data agree well, with the *Meteor* values being slightly higher for the deep layers. Lowest values are in the first few hundred metres, after which the ratio increases and becomes fairly constant from 1000 to 4000 m. *Meteor* data show an increase below 4000 m.

The northern Indian Ocean is closed to the north by land masses, which influence its

Table 1. Average Ca (g kg⁻¹):Cl (×10³) ratio for various depth intervals

Depth range (m)	Gaveshani data		Meteor data	
	Ca:Cl	No. of samples	Ca:Cl	No. of samples
0–200	0.02158	54	0.02156	108
201–400	0.02162	14	0.02163	28
401–600	0.02170	9	0.02166	28
601–800	0.02168	8	0.02167	15
801–1000	0.02170	8	0.02166	15
1001–1500	0.02168	9	0.02172	30
1501–2000	0.02169	7	0.02174	24
2001–2500	0.02171	3	0.02173	17
2501–3000	0.02169	6	0.02173	15
3001–3500	0.02168	4	0.02171	15
3501–4000	0.02171	5	0.02170	9
>4000	0.02169	1	0.02175	10

climate and give rise to unusual oceanographic phenomena. A number of rivers flow into this area; the annual runoffs from the Brahmaputra, Ganges, and Irawadi have been estimated to be 630, 490, and 440 km³, respectively (BAUMGARTNER and REICHEL, 1975), and the contribution of other rivers along the Indian coast is about 720 km³ y⁻¹ (RAO, 1977). A value of 175 km³ y⁻¹ can be calculated for the Indus (LEOPOLD, 1962) bringing the total runoff from these rivers to about 2500 km³ y⁻¹. Assuming about 500 km³ y⁻¹ to be the runoff from all other rivers discharging into the region in the absence of reliable data, a total of about 3000 km³ is obtained. This is about 9% of the total estimated annual global river runoff (about 32,000 km³, GARRELS and MACKENZIE, 1971; LIVINGSTONE, 1963). The total area of Arabian Sea, Bay of Bengal, Andaman Sea, Persian Gulf, and Red Sea is 11.2 × 10⁶ km², approximately 3% of the total area of the oceans (SVERDRUP, JOHNSON and FLEMING, 1942). Thus, about 9% of the global runoff from the rivers occurs into the northern Indian Ocean, only 3% of the total oceanic area. The total dissolved calcium flux from the rivers has been estimated to be 495 × 10¹² g y⁻¹, with an average calcium concentration in river water of 14.6 mg l⁻¹ (MARTIN and MEYBECK, 1979). The flux of dissolved calcium from rivers into the northern Indian Ocean is about 45 × 10¹² g y⁻¹. Such a large flux should increase the Ca:Cl ratio in the region, at least in the surface layers. The bulk of the runoff is in the Bay of Bengal and probably explains the higher Ca:Cl ratio for the bay and for the Andaman Sea (SEN GUPTA *et al.*, 1978; NORONHA *et al.*, 1981). As the subsurface waters in the northern Indian

Table 2. Average Ca:Cl ratio in the upper 200 m for individual Gaveshani stations

Station No.	Location	Ca:Cl
1937	8°40'N; 66°00'E	0.02155
1896	6°48'N; 67°17'E	0.02163
1899	5°28'N; 61°58'E	0.02171
1902	2°46'N; 59°59'E	0.02165
1910	0°34'S; 59°16'E	0.02166
1911	3°39'S; 59°18'E	0.02154
1917	10°01'S; 57°05'E	0.02147
1914	13°26'S; 58°12'E	0.02148

Ocean are formed by excessive evaporation in coastal areas, the riverine influence could also affect the ratio in the subsurface layers.

The average Ca:Cl ratios in the upper 200 m of individual *Gaveshani* stations (Table 2) generally decreased southward; the two southernmost stations show the lowest ratios (0.02147 and 0.02148). Evidently the river runoff significantly increases the ratio in the upper layers of the northern Indian Ocean.

The Ca:Cl ratios in the surface layers are lower than in the deeper layers (Table 1), probably the result of biological uptake of CaCO_3 by organisms in the upper layers. The rates of CaCO_3 precipitation are not accurately known, but it is a productive area (QASIM, 1977, 1982), and $1 \text{ to } 2 \times 10^{-3} \text{ g CaCO}_3 \text{ cm}^{-2} \text{ y}^{-1}$ appears to be reasonable. This would remove calcium by $45 \text{ to } 90 \times 10^{12} \text{ g y}^{-1}$, the same order as the annual flux from rivers. The flux from rivers into other areas of the Indian Ocean should be much lower; about 65% of the runoff in the Indian Ocean is in the northern sector. Solution of calcium in the deeper layers and its upward flux should maintain a steady supply of calcium to the surface layers to compensate for its removal by organisms.

It has long been known that the deep waters contain more calcium relative to chlorinity than do surface waters (e.g., WILSON, 1975; TSUNOGAI, YAMAHATA, KUDO and SAITO, 1973; SHILLER and GIESKES, 1980). Tsunogai *et al.* studied the variations in the ratio in the Pacific Ocean and found a maximum of 0.02134 in deep waters of the North Pacific, about 1.1% higher than in surface waters of the tropical and subtropical Pacific. Our value for the deep waters (>2000 m) (0.02170) is about 1.7% higher than that for the deep Pacific. The deep water masses in the two oceans are of Antarctic origin, and so they would be expected to have similar 'preformed' calcium. Hence the higher ratios in the deep northern Indian Ocean are probably the result of processes peculiar to the region.

Three samples were recently collected close to the bottom in the Central Indian Ocean, using a new technique that involves linking the triggering mechanism of a deep-sea grab to that of a water sampler fixed just above the grab. The samples were analyzed for calcium (Table 3) but alkalinity could not be determined. The Ca:Cl ratio for the near-bottom samples is anomalously high and the enrichment of calcium, calculated by using the average ratio of 0.02164, ranges from 3.42 to 4.85%.

SHILLER and GIESKES (1980) examined the processes controlling the distribution of calcium in the oceans. They identified two fluxes that could contribute calcium to deep waters, one resulting from the low temperature alteration of the basalt and basal sediments, the other due to the hydrothermal basalt alteration at ridge crests and fracture zones. While the former flux is not great enough to increase the deep-water calcium measurably, the latter could contribute an extra $5 \mu\text{M kg}^{-1}$ of calcium to the bottom waters of the North Pacific (SHILLER and GIESKES, 1980). The contribution by such processes should be of the same order in the Indian

Table 3. Calcium enrichment in the near-bottom samples from the Central Indian Ocean

Station	Depth (m)	Calcium (mg kg^{-1})	Salinity ($\times 10^3$)	Ca(g kg^{-1}): Cl($\times 10^3$)	Ca enrichment (%)
2394 (5°00.7'S; 76°59.3'E)	5050	430.6	34.76	0.02238	3.42
2396 (5°00.4'S; 79°00.7'E)	5200	433.9	34.54	0.02269	4.85
2397 (3°59.9'S; 79°00.7'E)	4980	429.0	34.42	0.02252	4.07

Ocean, and hence would not account for the anomalously high calcium in the near-bottom waters. As the three samples came from well below the lysocline, it is apparent that the high ratios are caused by the solution of CaCO_3 from the underlying sediments. A high rate of CaCO_3 supply from the surface layers and its subsequent solution probably account for the enrichment of the bottom waters.

WARREN (1981) estimated the net northward volume transport below 2000 m for the three deep boundary-current systems of the Indian Ocean to be about $16 \times 10^6 \text{ m}^3 \text{ s}^{-1}$ at 18°S latitude. Using this value he calculated the average vertical velocity near the 2000-m level to be $4.0 \times 10^{-5} \text{ cm s}^{-1}$, compared to $0.8 \times 10^{-5} \text{ cm s}^{-1}$ for the South Atlantic and $1.3 \times 10^{-5} \text{ cm s}^{-1}$ for the Pacific. The Indian Ocean value is thus three times that of the Pacific and five times that for the South Atlantic. Warren suggested that the upward movement of water is much more rapid in the Indian than in the other two oceans. Consequently, the upward flux of calcium added to the bottom waters due to CaCO_3 solution would also be much higher in the Indian than in the Pacific or Atlantic oceans. Therefore, we suggest that

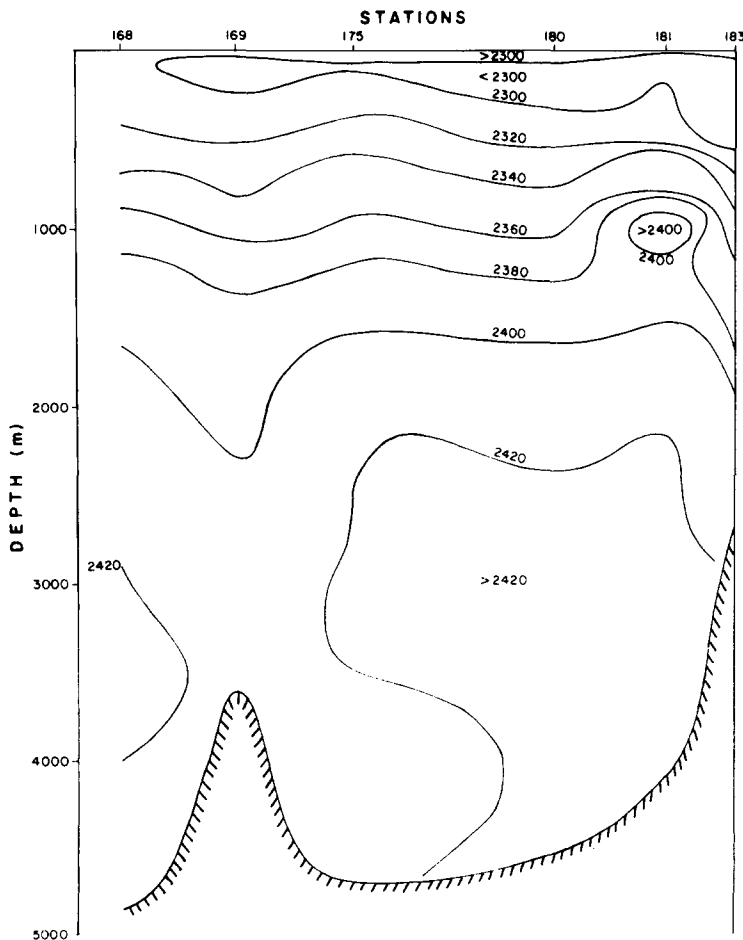


Fig. 2. Distribution of normalized alkalinity ($\mu \text{ eq kg}^{-1}$) along a *Meteor* section.

the high Ca:Cl ratio in the deep waters of the northern Indian Ocean is due to input of calcium by excessive stripping at the surface and a more rapid upward flux than in other oceanic areas.

The distribution of normalized alkalinity ($A_N = \text{Alk} \times 35/S$) along a *Meteor* section is given in Fig. 2. The deep alkalinity data from the northern Indian Ocean are comparable to similar data from the North Pacific (see Fig. 2 of SHILLER and GIESKES, 1980). This is surprising considering that if the higher calcium in the deep waters of the northern Indian Ocean is due to CaCO_3 solution, the alkalinity should also be higher by 0.3 to 0.4 meq kg^{-1} . However, no such increase in alkalinity is observed. It is unlikely that our data are systematically too high for calcium or too low for alkalinity, as the *Meteor* and *Gaveshani* data agree quite well. We cannot explain the discrepancy but suggest that it could arise from changes in alkalinity by processes other than CaCO_3 solution.

Degree of CaCO_3 saturation and CaCO_3 solution in deeper layers

Calcite saturation profiles at seven stations are given in Fig. 3. Surface waters are several fold supersaturated with respect to calcite, and the degree of saturation decreases sharply within the thermocline. The supersaturation extends to considerable depths and a crossover to undersaturation is observed between 2500 and 3000 m.

There are two earlier reports on variations in the degree of CaCO_3 saturation with depth in the Indian Ocean (NAQVI and REDDY, 1975; INGLE, 1975). Based on the Indo-Soviet Monsoon Experiments (ISMEX) 1973 data, NAQVI and REDDY (1975) observed that the saturation depth is about 2600 m in the equatorial Indian Ocean and about 1000 m in the northern Arabian Sea. Undersaturation at shallower depths in the latter region is probably due to high total CO_2 content of the intermediate layers (NAQVI and REDDY, 1975) resulting from a high rate of photosynthesis at the surface and limited ventilation at intermediate depths in the region (NAQVI, NORONHA and REDDY, 1982; QASIM, 1982). INGLE (1975) published saturation profiles along a *Meteor* section running from the Somali coast to the southwest coast of India; they show undersaturation between 1000 and 2000 m, with the saturation depth decreasing from southwest to northeast. Our results are, therefore, consistent with those of NAQVI and REDDY (1975), but they indicate that the saturation depth in the Indian Ocean is slightly deeper than reported by INGLE (1975). The discrepancy is difficult to account for, because the same constants were used for the calculation of Ω in both studies. We suspect the discrepancy might arise from the procedures for calculating the *in situ* pH.

The percent CaCO_3 of sediments at 3 stations in the Arabian basin and 16 stations in the Somali basin is given in Fig. 4. A sharp decrease in CaCO_3 content occurs between 4400 and 4800 m. The three stations in the Arabian basin showed relatively low CaCO_3 content in the sediments. KOLLA, BE and BISCAYE (1976) reported that the carbonate critical depth (depth below which the carbonate content is <10%) is about 4800 m in the Arabian basin and >5100 m in the Somali basin. They found that the lysocline (both 'foram' and 'sedimentary') in the region starts at about 4000 m. The data indicate that the carbonate critical depth and the lysocline lie far below the saturation depth in the Indian Ocean as in other oceanic areas (PYTKOWICZ, 1970; EDMOND, 1974).

Morse and Berner studied the kinetics of solution of calcite at constant degrees of saturation (MORSE and BERNER, 1972; BERNER and MORSE, 1974) and found that near equilibrium, similar to the conditions prevailing in the sea, the rate of solution accelerated sharply at a critical degree of saturation ($\Omega = 0.31$ to 0.67), which increased with the phosphate concentration. They concluded that at such a critical degree of saturation the monomolecular

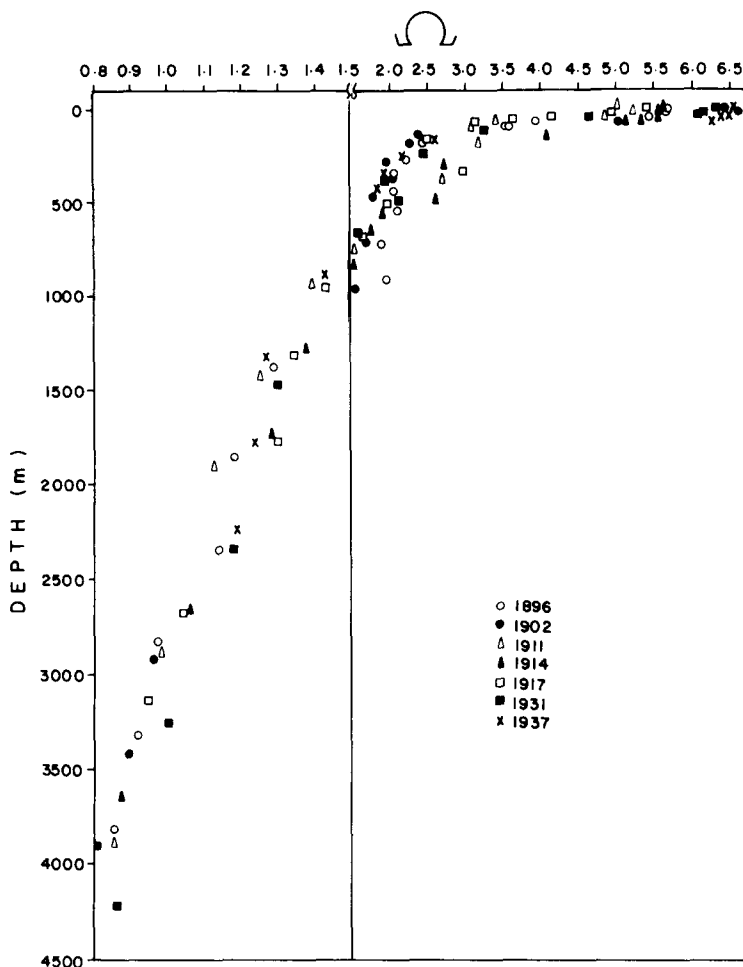


Fig. 3. Vertical distribution of the degree of calcite saturation.

steps at the crystal surface penetrate between the adsorbed inhibitor ions such as phosphate, thereby bringing about solution, and that the lysocline in the oceans is due to this mechanism.

EDMOND (1974) compared the levels of 50% saturation with the lysocline depth in the Pacific Ocean and found good agreement between the two in the Central Pacific, but not in the southwest Pacific and the Antarctic regions. Instead, he observed a correspondence between the lysocline and the upper limit of turbidity as evidenced by the nephelometer profiles, prompting him to suggest that turbulence could be the dominant factor in the solution of calcite in deep-oceanic basins. In the South Pacific, however, the top of the nepheloid layer does not always coincide with the lysocline (KOLLA *et al.*, 1976). Moreover, for turbulence to be the dominant factor, as suggested by EDMOND (1974), the dissolution should be diffusion-controlled, i.e., the layer adjacent to the solid surface should be saturated with CaCO_3 , and the diffusion from the layer to the bulk solution, which will increase with turbulence, should determine the rate of solution. Such does not appear to be the case because BERNER and MORSE (1974) observed that solution could be diffusion-controlled at low pH

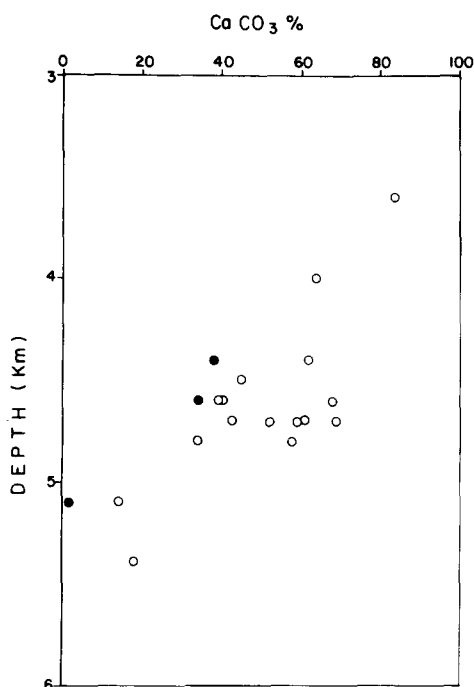


Fig. 4. Percent CaCO_3 in the sediments: solid circles for Stas 1894, 1896, and 1898 in the Arabian basin; open circles for stations in the Somali basin.

(<4.0); at low and moderate degrees of undersaturation, the concentration in the layer adjacent to the surface does not build up to saturation value and the surface reaction is, therefore, rate-limiting.

BROECKER and TAKAHASHI (1978) introduced the concept of critical carbonate ion concentration. They studied the variations in carbonate ion concentrations with depth in several areas in the Pacific and Atlantic and determined the carbonate ion concentration at lysocline depth. The data fitted the equation

$$(\text{CO}_3^{2-})_{\text{crit.}}^{\text{calc.}} (\mu\text{M kg}^{-1}) = 90 \exp[0.16(D - 4)]. \quad (1)$$

A similar equation for aragonite was also deduced

$$(\text{CO}_3^{2-})_{\text{crit.}}^{\text{arag.}} (\mu\text{M kg}^{-1}) = 120 \exp[0.15(D - 4)], \quad (2)$$

where D is the lysocline depth in km.

It was observed that the lysocline depth can be explained within 200 m in terms of the carbonate ion concentration alone, without considering bottom-water turbulence and phosphate content. To check the applicability of the model in the Indian Ocean, we plotted carbonate ion concentration vs depth for all stations deeper than 1000 m together with the curves given by equations (1) and (2) (Fig. 5). Below 1000 m there is an almost linear relation between the carbonate ion concentration and depth. The North Indian Deep Water and North Indian Bottom Water have higher carbonate ion contents than the overlying water masses. A high rate of oxidation of organic matter probably results in a higher total CO_2 content of intermediate layers, which suppresses the carbonate ion concentration, so the carbonate ion concentration

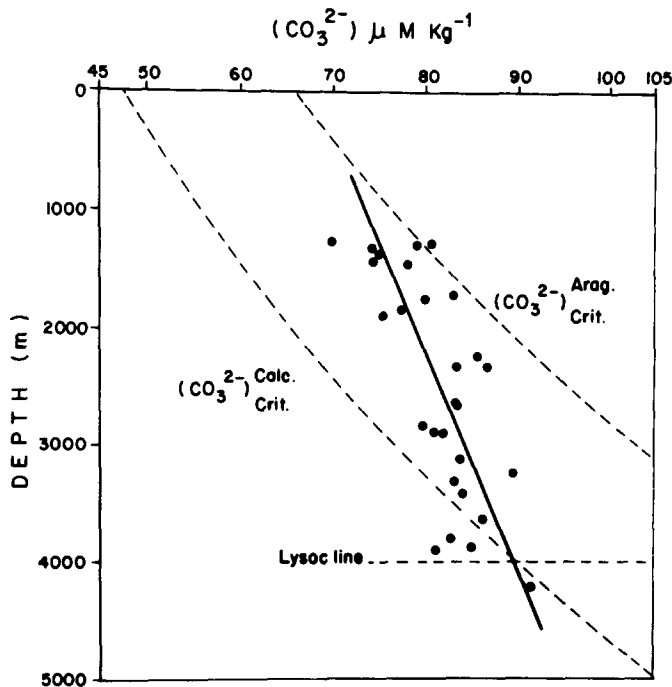


Fig. 5. Plot of carbonate ion concentration vs depth: the curves give the 'critical carbonate ion concentration' for calcite and aragonite, and the straight line represents the two-way linear least-squares regression between the carbonate ion concentration and depth. Lysocline depth from KOLLA *et al.* (1976).

increases with depth. The linear least-squares regression between carbonate ion concentration and depth intersects the curve for critical carbonate ion concentration for calcite at 4000 m, exactly the depth of the lysocline in the Arabian and Somali basins (KOLLA *et al.*, 1976). Although our results indicate that the 'sedimentary' lysocline may be a little deeper, we prefer to use the values given by Kolla *et al.* because their results are based on a large number of samples. Thus, there is an excellent agreement between the lysocline depth as predicted by the Broecker-Takahashi model and that observed in the region. The critical carbonate ion concentration for the area is $90 \pm 5 \mu\text{M kg}^{-1}$, a value comparable with other oceanic areas (BROECKER and TAKAHASHI, 1978).

The variations in calcium are insignificant compared to those in the carbonate ion concentration, and so the critical carbonate ion concentration actually corresponds to the critical degree of saturation. Therefore, the dominant factor controlling deep-sea carbonate dissolution should be the degree of saturation; waters with higher undersaturation (lower carbonate ion concentration) should dissolve more carbonate. This appears to be the case because the compensation depth in the Somali basin is deeper than in the Arabian basin (KOLLA *et al.*, 1976; Fig. 4), where deeper waters are more undersaturated.

The degree of saturation at the lysocline depth in our area of observations ranges between 0.7 and 0.8, slightly higher than the value experimentally determined by MORSE and BERNER (1974). However, the critical degree of saturation is likely to vary depending upon factors such as the extent of inert coating and the magnesium content of the tests.

Figure 5 indicates that the carbonate ion concentrations are less than the critical values for aragonite below about 1000 m. The aragonite tests may therefore dissolve above the lysocline. Indeed the dissolution of pteropod tests has been observed at relatively shallow depths (BERGER, 1970), but such dissolution is probably much less and therefore would increase the Ca : Cl ratio only slightly. Intense CaCO₃ solution will occur only below the lysocline, evident by the increase in the Ca : Cl ratio below 4000 m (Table 1) and in the near-bottom samples (Table 3).

Acknowledgements—We wish to express our gratitude to S. Z. QASIM, V. V. R. VARADACHARI, H. N. SIDDIQUIE, and C. V. G. REDDY who extended their kind support at various stages of the work. The computations were by our colleague K. ARAVIND GHOSH.

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