

On the occurrence of ferromanganese micronodules from the sediments of the Bengal Fan: a high terrigenous sediment input region

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Received 22 March 1994; accepted 14 September 1994

Abstract

Studies of 13 samples (10 surface sediments and 3 cores) from across the middle fan region of the Bay of Bengal reveal the presence of micronodules (area 0.22 million km²) over the entire bay. These micronodules have two dominant morphological forms: botryoidal in the eastern bay and spheroidal in the central and the western regions of the bay. The abundance of the micronodules was found to be higher in the eastern region of the bay. In the cores, the micronodules were generally found to decrease in abundance down-core. The core from the eastern region had micronodules down to deeper levels than the western side.

Todorokite is the manganese mineral present in the micronodules. Associated mineral phases are quartz, feldspar and phillipsite. These micronodules have high Mn (19.20–32.13%), rather low Fe (1.20–1.83%), Ni (0.10–1.14%), Zn (0.10–0.30%) and Co (0.01–0.04%), and their Mn/Fe ratio ranges from 10.55 to 25.42. The contents of Mn, Ni, Cu and Zn, and the Mn/Fe ratio decrease down-core.

Based upon the variations in the abundance, morphology, Mn/Fe ratio of the micronodules, and the elemental distributions and the O/Mn ratio in the sediments, remobilization of Mn from deeper levels and its precipitation at the upper surfaces of the sediment column, in an oxic environment, is suggested. The results of the present study also suggest that a high rate of sedimentation is not a limiting factor for the formation of micronodules, although it might affect their growth.

1. Introduction

Ferromanganese nodules are among the important authigenic constituents of marine sediments. Macronodules are generally present at the surface, whereas micronodules are distributed within the upper few centimetres of the sediment column. The important factors for the formation of nodules are: (1) availability and precipitation of Mn under favorable redox conditions; (2) seed-

ing material; and (3) a low sedimentation rate [1–14]. Based upon mass balance equations, Bender et al. [1,2], Hishida and Uchio [6] and Stoffers et al. [14] have shown a negative correlation between the rate of sedimentation and the formation and abundance of these nodules and, as such, the reported occurrence of macro- or micro-ferromanganese nodules are mainly from the areas of low sedimentation (a few mm/ky) [7,15–21]. The occurrence of micronodules has, so far,

not been reported in sites having a very high terrigenous input, particularly in the fan-canyon systems of the world.

The Bengal Fan, together with its eastern lobe, the Nicobar Fan, is one of the largest of its kind in the world [22,23]. As part of integrated surveys of the Bay of Bengal Fan, 13 box cores were collected from the middle fan region of the bay during cruise numbers 31 and 72 of the ORV *Sagar Kanya* (Fig. 1). The present work is based upon the studies of these cores and reports the occurrence of manganese micronodules in this area, characterized by high terrigenous input. Ferromanganese micronodules are, hitherto, un-

reported from such a depositional environment. This study also presents the temporal and spatial distribution, morphogroups, bulk mineralogy, chemical composition and genesis of the micronodules from the Bay of Bengal.

2. Study area

The Bay of Bengal is one of the largest terrigenous depositional sites of the world oceans (annual suspended sediment input about 2000 million t). The major physiography of the bay has been described by Emmel and Curray [23] and

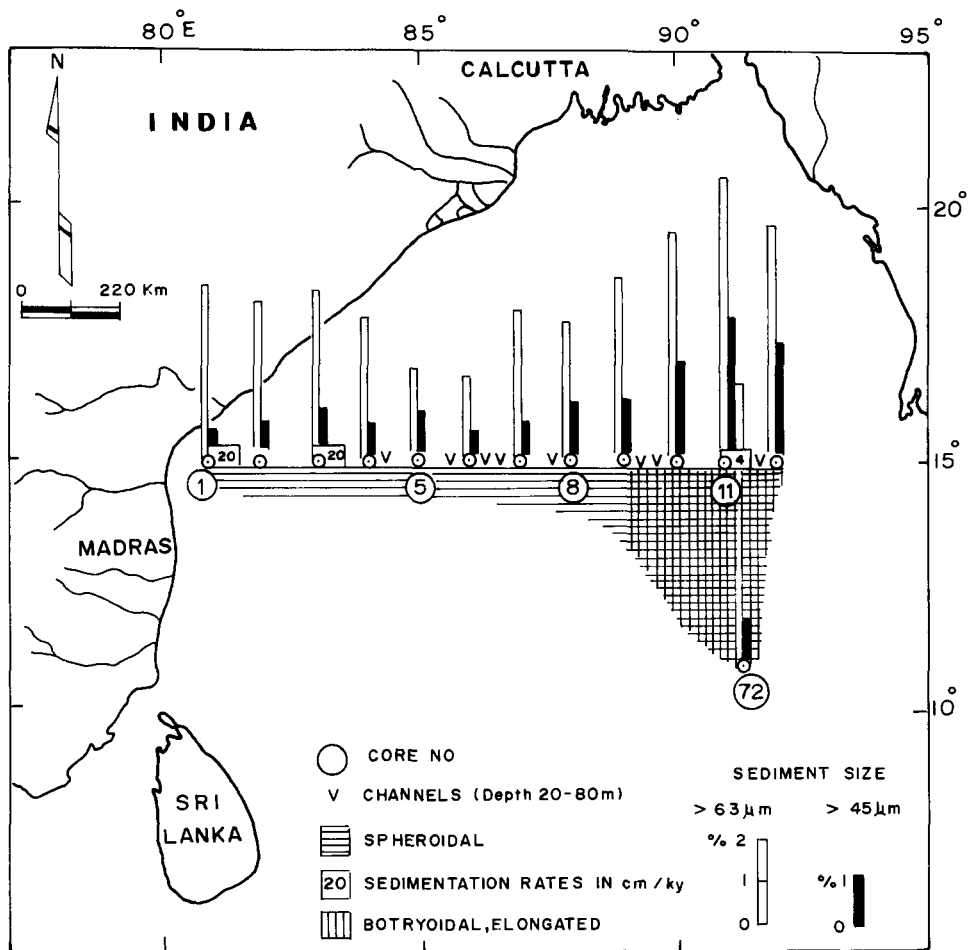


Fig. 1. Location of the sampling stations, surface abundance (in > 45 and $63 \mu\text{m}$ fractions) and spatial distribution of morphogroups of the micronodules in the sediments of the Bay of Bengal. Holocene rate of sedimentation based upon earlier studies [39,40] are also shown.

Curry and Moore [24]. A major portion of the floor of the bay is covered by the Bengal Fan, a terrigenous depositional feature resulting from sediment input from perennial rivers. Prominent among these are the Ganga, the Brahmaputra and their tributaries in the Himalayas, in the north; the Mahanadi, the Krishna, the Godavari, and the Cauvery, in the west from peninsular India; and the Irrawadi, in the east, from Burma. The fan is reported to be about 2800–3000 km long, 830–1430 km wide and sediments over 16 km thick carpet the floor of the northern Bay [22,23]. The study area is located in the middle fan region [22], which has active channels with levees. The area is reported to have been an area of sedimentation since the beginning of Miocene [25].

3. Materials and methods

13 box cores (generally 30 cm × 30 cm × 6 m) were subsampled at 5 cm intervals, washed to remove salts, dried at 60°C and preserved for laboratory analyses. These subsamples were subjected to grain size separation at 1 phi intervals using standard methods [26]. The separated > 45 μm fractions (sand and coarse silt) were examined under a binocular microscope and their components were determined. The abundance of the micronodules was obtained in the > 45 and 63 μm fractions of 10 g dried samples, and a minimum of 300 grains were counted in each sample to calculate the abundance of various morphogroups. Due to the limitation of the small quantity of the micronodules in the sediment, only bulk mineralogy on the hand-picked, powdered micronodules was obtained, using a Phillips Diffractometer Model PW 1840 (using a Ni-filtered Cu K_α radiation).

The hand-picked micronodules from selected levels of cores 1 and 11, from the eastern and the western regions of the bay, were impregnated in a cold setting resin, polished, coated with gold and analyzed for selected major and trace elements using an electron microprobe (model Jeol JS-840). To enhance the accuracy, on each micronodule eight spots were analyzed and average concentration (sigma values 0.01–0.03) of these analysis

were reported. For detailed morphological studies, the micronodules were studied under a scanning electron microscope. In all, over 100 micronodules were studied.

The chemical analysis of the sediments of cores 1 and 11 was carried out using a Phillips XRF (model 1480). The accuracy of the results of the chemical analysis was checked against 20% repeat samples, and also, against the standard SDO 1-3 and was found to be better than 5%. The concentrations of the metals in the sediments are presented on a CaCO₃-free basis.

The oxidation state of the sediments of core 1 and 11 was determined following the Winkler iodimetric titration method, as described in Murray et al. [27]. The O/Mn ratio as MnO_x was calculated using the formula:

$$x = 1.0 + 0.5 \frac{[(\text{total oxidizing equivalent}) - (\text{total Fe equivalent})]}{(\text{total moles of Mn})}$$

The accuracy of the analysis (determined from 20% repeat samples) is less than 4%. Also, a

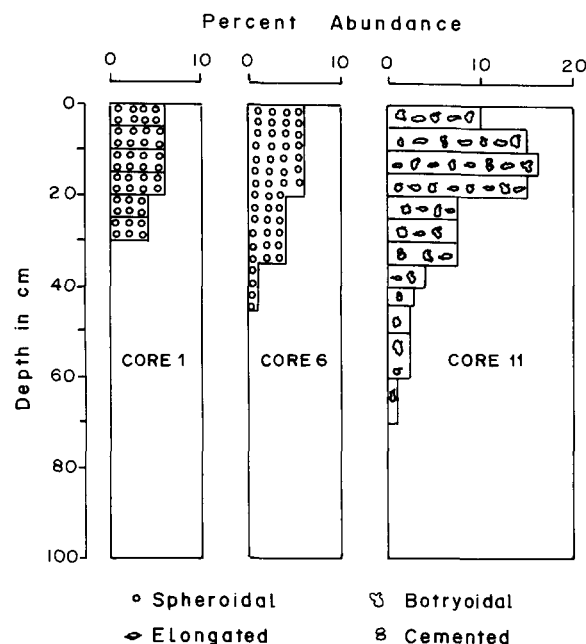


Fig. 2. Variations in the down-core abundance of the micronodules in the cores from the eastern, the western and the central bay (cores 1, 6 and 11). For locations of the cores see Fig. 1.

routine checkup on reagent grade $\text{MnSO}_4 \cdot 2\text{H}_2\text{O}$ gave O/Mn values between 1.02 and 1.04.

4. Results

The spatial distribution of the micronodules in the fan sediments of the Bay of Bengal is shown in Fig. 1. These results suggest that between the eastern and the western bay the micronodules are present over a 1100 km long transect. Their pres-

ence is also recorded about 400 km southwards (Fig. 1). The study of the cores from the eastern, the western and the central bay (Figs. 1 and 2) also confirms their presence at subsurface levels. The abundance of the micronodules in these cores generally follows a decreasing down-core trend. The levels down to which these micronodules occur in these cores were found to be highly variable (Fig. 2). In the eastern bay the micronodules were recorded at deeper levels in the core (78 cm) than in the western bay (30 cm). The

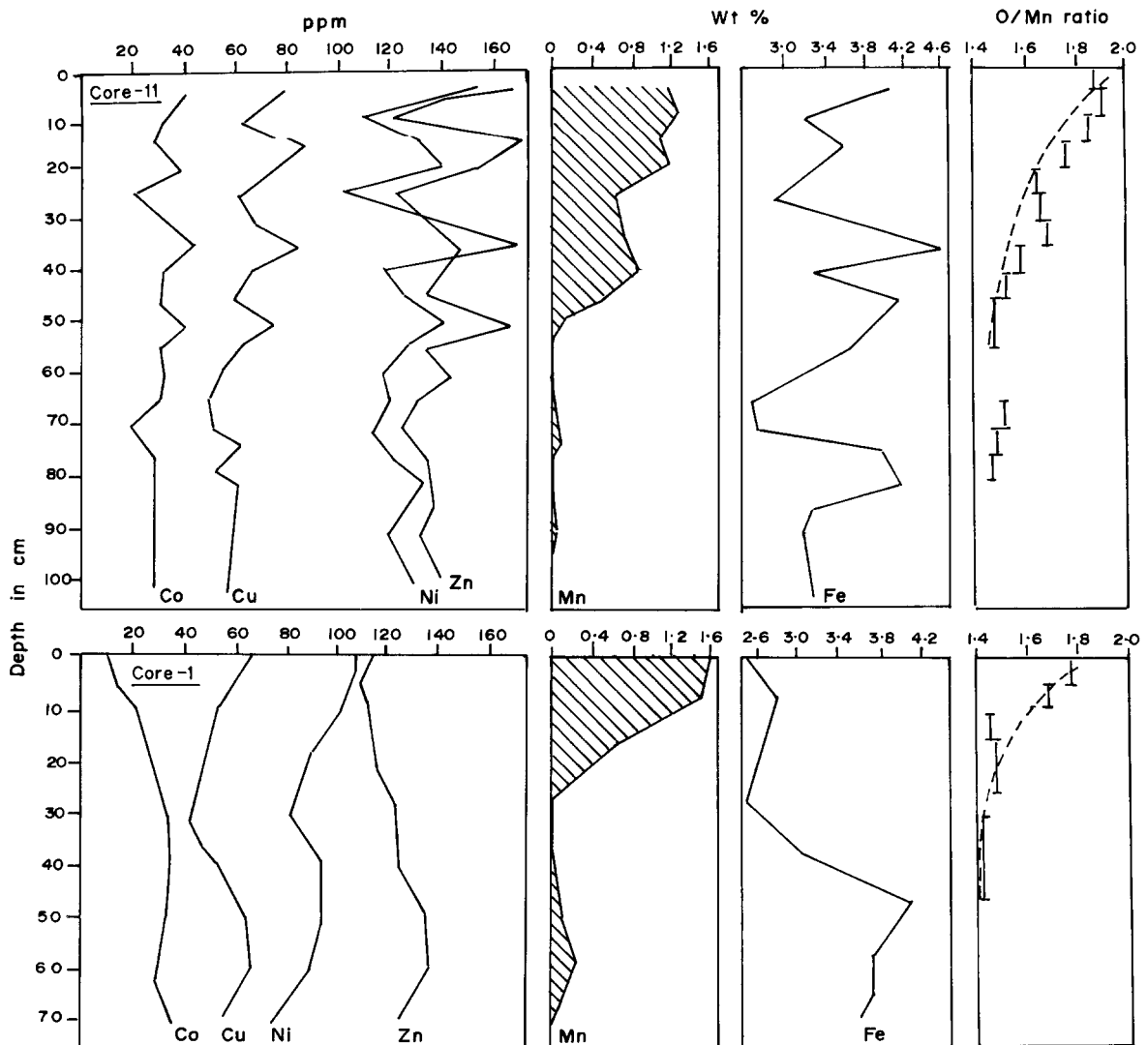


Fig. 3. Down-core variations in the concentrations of the Mn, Fe (percent), Co, Cu, Ni, Zn (ppm) and O/Mn ratio in the cores from the eastern and the western regions of the bay (cores 1 and 11).

Table 1

Chemical composition of micronodules (obtained from polished sections of micronodules on an electron microprobe) from cores 1 and 11 sections of cores 1 and 11

Level (cm)	Elements										Mn/Fe ratio
	Si	Ca	Co	Cu	Ni	Zn	Mn	Fe	Ti	Al	
<i>Core 11</i>											
0–5 (<i>N</i> = 5)	1.03	1.14	0.01	1.07	1.14	0.30	32.13	1.26	0.01	1.96	25.42
5–10 (<i>N</i> = 5)	0.92	1.16	0.02	1.01	1.07	0.22	31.36	1.45	0.01	1.88	21.66
10–15 (<i>N</i> = 5)	0.87	1.17	0.01	0.93	0.98	0.22	29.17	1.33	0.01	1.88	21.97
15–20 (<i>N</i> = 5)	0.82	1.10	0.04	0.91	0.93	0.22	28.07	1.42	0.01	1.99	19.82
20–25 (<i>N</i> = 5)	0.74	1.07	0.02	0.76	0.71	0.17	24.36	1.67	0.01	1.94	14.62
30–35 (<i>N</i> = 5)	0.72	1.17	0.02	0.65	0.68	0.16	22.54	1.43	0.01	1.99	15.77
40–45 (<i>N</i> = 4)	0.65	1.09	0.03	0.48	0.42	0.12	21.47	1.83	0.01	1.95	11.72
55–60 (<i>N</i> = 4)	2.67	.92	0.03	0.34	0.22	0.10	19.20	1.82	0.01	1.88	10.55
<i>Core 1</i>											
0–10 (<i>N</i> = 5)	0.65	1.93	0.01	0.43	0.36	0.20	24.19	1.07	0.01	1.38	22.65
10–20 (<i>N</i> = 5)	0.67	2.01	0.02	0.29	0.28	0.20	22.51	1.30	0.01	1.21	17.31
20–30 (<i>N</i> = 5)	0.83	1.69	0.02	0.17	0.10	0.10	19.83	1.62	0.01	1.18	12.21

All values in weight percent. For locations of the cores, refer to Fig. 1. Note enrichment of Mn, Ni, Zn and Cu in the upper 0–20 cm.

surface sediments also have variations in the abundance of micronodules, and cores 11 and 12 from the eastern bay have a higher abundance of micronodules (> 9%; Fig. 1) than in the western bay (3%; Fig. 1).

The morphology of the micronodules is vari-

able. In the eastern bay the micronodules have a predominantly botryoidal-elongated morphology with irregular surfaces. Cementing of two nodules, and occasionally a mammilliated form is also observed. The morphology of the micronodules in the western and the central regions is

Table 2

Average chemical composition of micronodules from Bay of Bengal, Central Indian Ocean Basin and Pacific Ocean

Element	Bay of Bengal		Central Indian Ocean Basin ^a	Pacific Ocean ^b	World average ^c
	A	B			
Ti	0.10	0.01	0.18	–	0.69
Mn	24.10	25.21	35.02	19.81	16.18
Fe	1.10	1.46	1.38	10.97	6.47
Al	2.04	1.74	0.66	–	1.47
Ca	1.90	1.33	1.64	–	2.2
Mg	1.30	–	2.40	–	1.56
Ni	0.08	0.64	1.44	0.42	0.48
Co	0.01	0.02	0.16	0.12	0.30
Cu	0.05	0.65	1.62	0.43	0.25
Zn	0.15	0.14	0.37	0.11	–
Mn/Fe (range)	–	(10.55–25.42)	(5.00–101)	(1.95–5.37)	–

All values in weight percent. A: Obtained from digesting bulk nodules in aqua regia. B: Obtained from electron microprobe analysis (*N* = 52) of micronodules. ^a Adapted from Pattan [21]. ^b Adapted from Stoffers et al. [14]. ^c Adapted from Poppe et al. [16].

predominantly spheroidal; botryoidal, elongated or cemented morphogroups are conspicuously absent.

In photomicrographs of the polished sections of the micronodules, todorokite is observed. X-ray diffraction of the bulk micronodules also indicates that todorokite is the dominant manganese mineral phase in all the 9 samples analyzed from the eastern, the central and the western bay. The δ MnO₂ phase was conspicuous by its absence. Associated mineral phases are quartz, feldspar and phillipsite. A similar association of the minerals was also observed in the micronodules from the Central Indian Basin (CIB) and the Pacific [7,20].

The results of the chemical analyses of the micronodules and sediments of the cores are presented in Fig. 3 and Tables 1 and 2. From these results it is observed that Mn contents of these micronodules are highest in the top 0–20 cm sections (32.13–29.19% for core 11 and 24.19–22.51% for core 1) and have a down-core decreasing trend. Mn contents are much reduced at the deeper levels (19.20% at 55–60 cm in core 11 and 19.83% at 20–30 cm in the core 1; Table 1). The concentrations of Cu, Ni and Zn show similar down-core decreasing trends but Fe and Co contents have inverse relations to these metals (Table 1). Ti values are constant and Al, Si and Ca do not have a clear trend. However, of cores 1 and 11, the micronodules of core 1 (western bay) have higher Ca contents. The Mn/Fe ratio of the micronodules also varies. This ratio is higher in the upper 20 cm sections (25.42–19.82), and decreases with depth in both the cores (from 15.77 to 10.55 from the 30–35 to the 55–65 cm sections of core 11; Table 1).

The results of bulk chemical analysis of the micronodules (obtained after digesting 100 mg of bulk, powdered micronodules in aqua regia and elements analyzed on ICP–AES Plasma-400), in general, agree with the average elemental concentration obtained from the electron microprobe data (Tables 1 and 2). These results also suggest that the micronodules of the Bay of Bengal, on an average, contain mostly Mn (range 32.13–19.20%), as well as Ni (1.14–0.10%); Co (0.04–0.01%); Zn (0.30–0.10%) and Fe (1.83–1.20%).

The Mn/Fe ratio in the micronodules is also high (25.42–10.55). Compared to the deep sea micronodules of the other regions of the world's oceans (Table 2), these micronodules are poorer in Mn, Cu and Ni, and have values of Fe comparable to that of CIB (Table 2). The micronodules of the Pacific Ocean, on the contrary, have a ten-fold higher Fe, resulting in a lower Mn/Fe ratio (Table 2).

5. Discussion

As indicated earlier, the important parameters which control the formation of the micronodules are: (1) the availability of Mn; (2) an oxidizing environment; (3) a low sedimentation rate; and (4) the availability of the seeding material [1–4,7–12,28,29]. The observed presence of micronodules over a sizable area in the middle fan region of the Bay of Bengal confirms that the environment for the formation of these micronodules is widespread and suitable conditions do prevail, despite the high rate of sedimentation in this region.

6. Sources of Mn

Three accretionary modes: (1) hydrogenous precipitation; (2) oxic diagenesis; and (3) suboxic diagenesis; have been suggested for Mn precipitation in marine environments [7–15,21]. At the MANOP sites, based upon studies of pore water chemistry and O/Mn ratio, the existence of four environments has been identified [8,9,12,29]: (1) the oxidized zone; (2) the manganese oxidation zone; (3) the manganese reduction zone; and (4) the iron reduction zone. From Mn contents of pore water, it is suggested that during early diagenesis Mn(IV) present in the 'Mn reducing zone' dissolves as Mn(II), mobilizes upwards and precipitates as Mn(IV) in the 'Mn oxidizing zone' in the upper surfaces of the sediment column [8,9,29–31]. The oxidized zone is, however, most important for hydrogenous precipitation.

Earlier studies [11,21,31–35], using variations in Mn, Fe and Cu + Ni + Co contents of macro/micro nodules, have delineated the field

of nodules precipitated in hydrogenous, oxic and suboxic environments. The results of the chemical analysis of the micronodules from different levels of the cores 1 and 11 were plotted in the ternary diagram [31–33]. The micronodules of 0–40 cm levels of core 11 fall within the oxic domain, while the micronodules of the deeper subsurface (> 40 cm levels of core 11) show some shift towards the suboxic zone (Fig. 4). However, for core 1 the micronodules from the 20–30 cm levels fall close to the suboxic domain. From these results it is inferred that the processes which contribute to the formation of these micronodules are oxic-suboxic diagenesis.

The abundance of micronodules in the three cores from the central, the eastern and the western bay shows a down-core decreasing trend and their abundance is greatest in the upper 20 cm

sections of all the cores studied (Fig. 2). These results confirm a surface enrichment of Mn in the form of micronodules in the upper 20 cm levels. The O/Mn ratio of the sediments in the 0–20 cm portion of cores 1 and 11 is significantly high (1.89–1.64 in core 11 and 1.79–1.69 in core 1) and reduces considerably in the sections below 20 cm (1.49 at 65–70 cm in core 11 and 1.41 at 40–50 cm in core 1; Fig. 3). There is also an enrichment of Mn (~ 1.0–1.5%) in the sediments of cores 1 and 11 in the upper 0–20 cm (Fig. 3). This enrichment corresponds well with the higher abundance of the micronodules in these sections of the cores (Figs. 2 and 3). Kalthorn and Emerson [29], from O/Mn ratios and porewater chemistry, have reported that an O/Mn ratio of 1.60, and above, correlates with a reduction in porewater Mn contents and an enrichment of solid phase

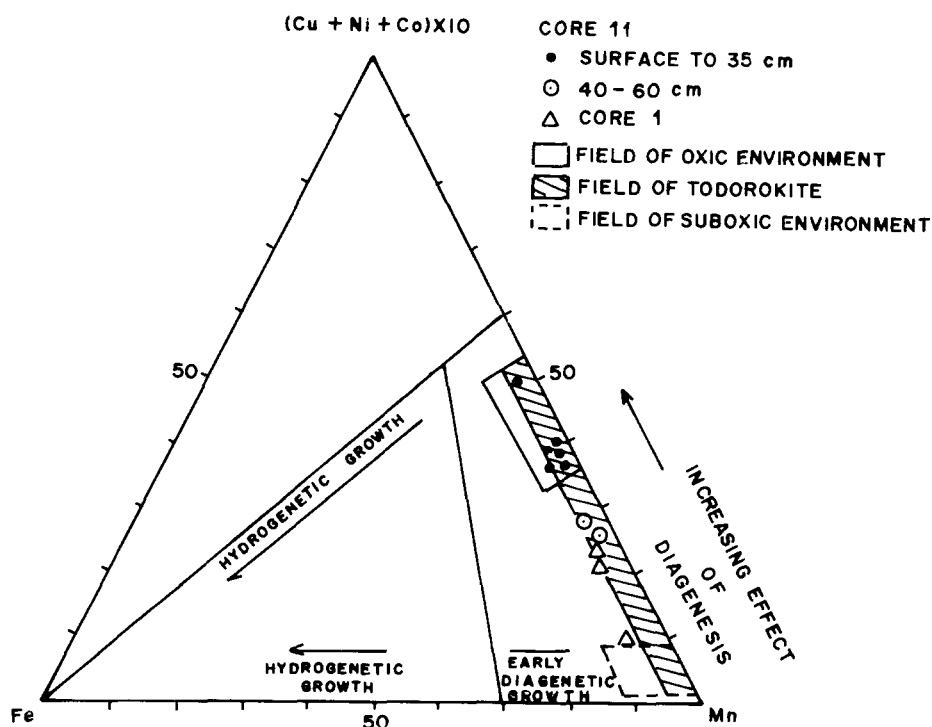


Fig. 4. A plot of the composition of the micronodules of cores 11 and 1 in a triangular diagram modified from [11,32,33]. Samples from the upper surfaces (0–20 cm for core 1 and 0–40 cm for core 11) mostly occupy the oxic zone in the todorokite region. A shift in the down-core samples (> 45 cm in core 11) toward the suboxic domain is also observed. This shift is more pronounced for the micronodules of the 20–30 cm level from core 1.

Mn in the sediments. These characteristics correspond to the 'Mn oxidation zone' [9,29]. The bottom water dissolved oxygen content of the study area is $\sim 160 \mu\text{mol l}^{-1}$ [34], similar to that of the Pacific and the Central Indian Ocean ($160 \mu\text{mol l}^{-1}$ and $160\text{--}180 \mu\text{mol l}^{-1}$ respectively; [21]), which suggests a similar availability of O_2 . In the upper 20 cm levels, the high O/Mn ratio (> 1.60), enrichment of the solid Mn phase, and the corresponding higher abundance of the micronodules in the sediment column, and enrichment of Mn in the micronodules, lead us to identify these sections as the 'Mn oxidation zone'. The Mn contents in the sediments of the cores below 20 cm levels, particularly in the deeper levels (below 25 cm), show a negative curve, and the abundance of micronodules and the O/Mn ratio is also much reduced (< 1.5 ; Figs. 2 and 3). These parameters correspond well with the characteristics of the 'Mn reducing zone' [9].

The elemental distribution in the micronodules from the different levels has been evaluated to ascertain the above referred zones. In the upper 20 cm, the enrichment of Ni (0.22–1.14% and 0.10–0.36% for cores 11 and 1, respectively, Table 1), which is reported to be produced in the Mn reduction zone and consumed within the Mn oxidation zone [9], clearly confirms the influence of diagenetic mobilization from the deeper levels and its precipitation in the upper surfaces. Mn and associated metals, that is, Cu and Zn, are also observed to follow the similar trend and to be enriched in the 0–20 cm levels (Mn oxidation zone; Table 1). These observations suggest that the micronodules of this region undergo dissolution in the deeper levels (Mn reduction zone) and dissolved Mn(II), together with Ni, Cu and Zn, migrates upwards [35–37] and precipitates in the Mn oxidizing zone (having a O/Mn ratio > 1.6 ; Fig. 3). From these results it is inferred that oxic diagenesis is the controlling process for Mn precipitation and the formation of the micronodules in the Bay of Bengal. The micronodules of the upper surfaces (0–20 cm) represent the accreting micronodules, whereas the micronodules of the deeper subsurface undergo diagenetic dissolution and reduce in abundance.

Earlier studies [20–28] have suggested that the

marine manganese oxides are, in general, poorly crystallized and have large vacancies in their structures. This may pose some problems for the identification of these minerals. Significantly, todorokite observed in the polished sections of micronodules, and also identified by distinct reflections at 9.65 and 4.82 Å and weak reflections at 2.42 and 1.40 Å in X-ray diffractograms [28], is the major phase in the micronodules. δMnO_2 is conspicuously absent. Todorokite and δMnO_2 are the mineral phases reported to be produced as a result of oxic–suboxic diagenesis and hydrogenous processes, respectively [10,17,30,31,38]. The observed presence of todorokite in the micronodules of the bay, therefore, implies that these micronodules are formed by oxic diagenesis.

7. Rate of sedimentation

The rate of sedimentation in the area, as expected in a fan-canyon system, is high, ranging from 4 to 20 cm ky^{-1} (Fig. 1) [39,40]. The eastern zone, in the vicinity of cores 11 and 12, is reported to have the lowest sedimentation rate, 4 cm ky^{-1} [40]. The organic carbon content of the sediments of the core 1 varies between 0.44% and 0.41% for the 0–40 cm levels. The nodule-bearing sediments of the CIB have a comparable range of organic carbon (ranges between 0.67% and 0.38%; [41]). Enrichment of Mn in the surface sediments from the southern California Borderland, characterised by high sedimentation rates, has been reported [42]. However, the occurrence of micronodules from a high terrigenous sedimentation site has not been documented so far. The occurrence of micronodules in the bay, having 40- to 200-fold higher sedimentation than the nodule-bearing regions of the Pacific or CIB (although the organic carbon and dissolved oxygen contents are similar), is contrary to earlier observations [1,2,7,8,15,16,20,21]. The presence of the micronodules in the region is thus intriguing and, perhaps, suggests that the rate of sedimentation, although important for the growth, is not a limiting factor for their formation. In addition, the large variations in the magnitude of the sedimen-

tation within the study area also provide an opportunity for understanding the influence of sedimentation on the growth of micronodules.

Contrasting morphogroups of the micronodules have been observed in the areas characterized by high and low sedimentation rates. The micronodules of the eastern bay have dominant botryoidal, elongated and cemented morphogroups with irregular surfaces. The western and the central bay, however, have dominant spheroidal morphogroups and a complete absence of other types. Stoffers et al. [7] have reported that the latter morphogroups are produced by the deposition of Mn(IV) in biological tests which have been subsequently dissolved in situ. To confirm this, SEM studies of the polished sections of spheroidal and botryoidal groups were undertaken. The botryoidal group (confined to the eastern bay only) has a uniform deposition of todorokite and no concentric layering (alternately rich in Mn and Fe — an important feature observed in the micronodules of the CIB and Pacific; [7,14,21]) was observed. In the western bay (characterised by the presence of the spheroidal group) todorokite is observed to have replaced the outer shell and deposition continues within the shell. The micronodules of the core from the western bay have a higher Ca content than those from the eastern Bay (Table 1). To confirm the presence of biological tests, if still undissolved, 100 micronodules from the study area were treated with 5% acetic acid. The test revealed the occurrence of dissolution pits on the surfaces of the micronodules and a reduction in the mean size from 60 μm to 45 μm in 80% of the micronodules of the central and the western regions of the bay only. This reduction in size of the spheroidal morphogroups is associated with the dissolution of the biological component and confirms the deposition of Mn(IV), as todorokite, in carbonate tests. The botryoidal group, which has a larger mean size, does not show any reduction in the mean size after the acid treatment. This, together with the characteristics of deposition of todorokite in the polished section of botryoidal micronodules, implies that the development and growth of botryoidal, elongated or cemented forms represent an advanced stage of micronod-

ule formation rather than the spheroidal ones. As these morphogroups are characteristically confined to the areas of high and low sedimentation, these variations may be attributed to the influence of sedimentation.

The western and the central regions of the bay are characterized by a high sedimentation (20 cm/ky; Fig. 1) and the burial of the micronodules is much faster in these regions than in the eastern bay. The O/Mn ratio between cores 1 and 11 varies significantly and is much higher down to the deeper layers in the eastern bay (1.89–1.69 between 0 and 35 cm levels for core 11) than in the western bay (1.79–1.60 for 0–20 cm levels in the core 1). This ratio also reduces more rapidly in core 1 than in core 11 (< 1.46 below the 30 cm level in core 1; 1.41 at the 80 cm level in core 11). An early, deep burial of the micronodules associated with higher sedimentation will move the micronodules from a Mn oxitic to Mn suboxic/reducing environment much faster and will, therefore, either retard the precipitation of Mn(IV) released through diagenesis in the deeper subsurface or initiate an early dissolution of Mn [7–10,28–30]. Thus, an early, deeper burial of the micronodules will limit their growth.

Slow sedimentation, on the other hand, enhances growth of the micronodules because accreting micronodules will spend more time in a favorable Mn oxidation environment (0–35 cm). This is reflected by variations in the spatial abundance of the micronodules, the distribution of the morphogroups, and also by the cementation of the micronodules in the eastern bay. The occurrence of the micronodules in the eastern bay down to the deeper levels (78 cm) compared to the western bay (only 30 cm) further suggests either enhanced growth or better endurance of these micronodules against diagenetic dissolution in areas of low sedimentation.

8. Conclusions

From the above discussion, it is evident that the micronodules of the Bay of Bengal are formed from the diagenetic mobilization of Mn from

below the 20 cm level and its precipitation in the upper sediment column under oxic conditions.

Based upon the abundance of micronodules, the O/Mn ratio of the sediments in the eastern and the western bay, and the distributions of botryoidal, elongated and spheroidal morphogroups, it is inferred that the formation of the micronodules is controlled by the availability of the manganese and oxic environment and their growth depends mainly upon the sedimentation rate.

Acknowledgements

The authors are grateful to the Director, Dr. E. Desa, and to R.R. Nair, National Institute of Oceanography, Dona Paula, Goa, for providing helpful comments and discussion on the manuscript. Thanks are also due to Drs. P.K. Verma, N.H. Hashimi, B. Nagender Nath, A. B. Valsangkar, J.N. Pattan, S. Iyer and M. Shyamprasad for the fruitful discussions, and to Mr. N.V. Ambre, K. Ramani, G. Prabhu, P.G. Mislanker and V.M. Date for their help with various analyses and for photographic work. [MK]

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