

POLLUTION STUDIES IN THE ARABIAN SEA*

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The oceans are the final sink for a wide variety of wastes, which can be natural in origin, industrial or man-made, and receives them through land drainage, industrial discharge and atmosphere. The seas around India receive a considerable amount of discharge of domestic sewage and industrial wastes. If we assume our coastal population as 150 million generating 1.6 m³/day/person liquid waste and 1.7 kg/day/person solid waste the coastal regions of India would then receive 88 km³ liquid waste and 93 million tonnes of solid waste every year. Our marine environment can also be expected to have received about 25% of 250 000 tonnes of pesticides, manufactured in India between 1970 and 1976, and 110 000 tonnes of synthetic detergents which we consume annually. Of the total annual world marine transport of 1700 million tonnes of crude oil and its products about 990 million tonnes or approximately 58.3% of the total are carried across the Arabian Sea. It can also be expected that about 5% of all the wastes discharged inland annually finds its way ultimately to the seas around our country.

In this report, we have attempted to summarise the observations carried out during cruise XVI of RV GAVESHANI in the Arabian Sea devoted mainly to pollution studies. This cruise was the first of its kind to be undertaken by any research vessel in Indian waters. The broad objectives set for the cruise were to assess the present status of marine pollution and to collect baseline information on the 'health' of the Arabian Sea.

Two transects in the NE-SE direction along the 100 and 1000-fathoms isobaths between Bombay and Trivandrum were selected (Figure 1) with a view to survey the nearshore area of the west coast of India as well as part of the oil tanker route from the Gulf countries, across the Arabian Sea, to Far East and Japan. The studies were planned to carry out observations on the baseline levels of petroleum

* Contributions in this paper are equal from all the authors. S.P. Fondekar was responsible for petroleum hydrocarbons; T.W. Kureishy for chlorinated hydrocarbons; S.Y.S. Singbal for the heavy metals and Sujata Sanzgiri for mercury, all under the overall guidance of R. Sen Gupta.

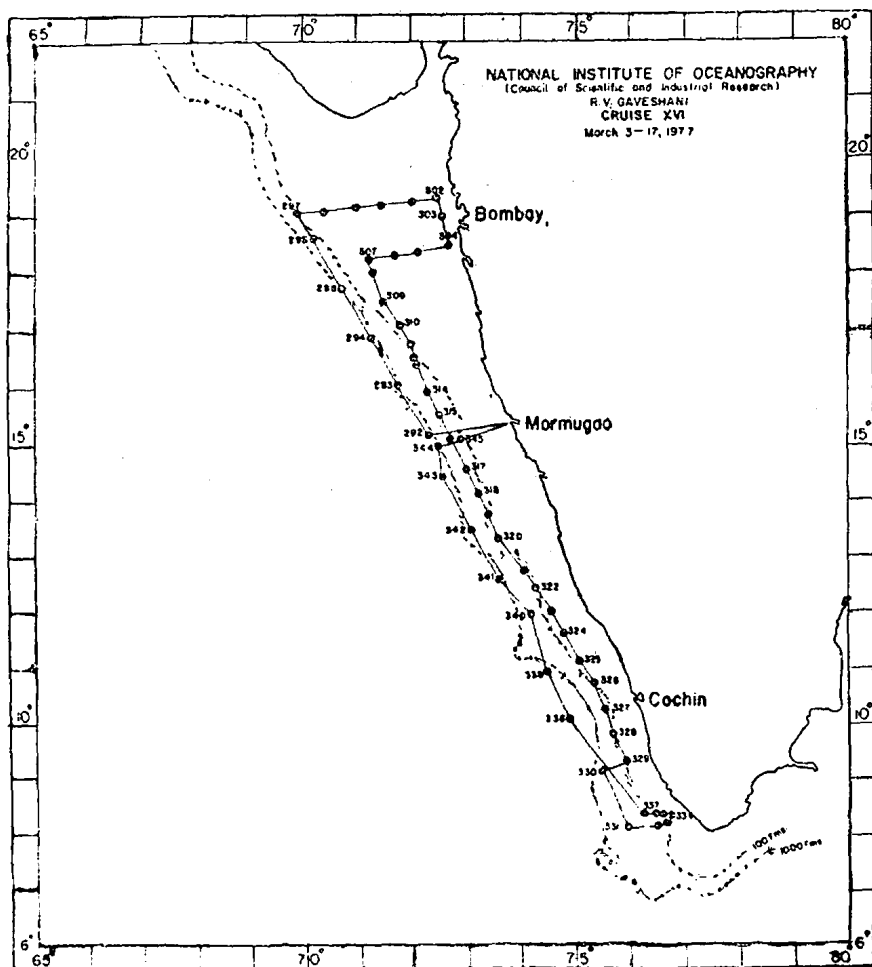


Fig. 1. Network of observation stations.

hydrocarbons, chlorinated hydrocarbons and several heavy metals *e.g.* Hg, Cu, Fe, Zn, Co, Ni, and Mn.

PETROLEUM HYDROCARBONS

Hydrocarbons are minor but ubiquitous components of the aquatic environment. Their primary natural sources are biosynthesis of aquatic organisms and terrestrial organic matter (Wakeham, 1976). Because of man's increasing dependence on petroleum and its products, the inputs of petroleum-derived hydrocarbons in the inland waterways, coastal areas and the ocean itself often may have considerable effect on aquatic ecology. The oil spilled annually at sea has been variously estimated as 2.2 to 6.1 million tonnes (Revelle *et al.* 1971; Goldberg and Grant-Gross,

1971; Farrington, 1977), which is equivalent to about 0.4% of the total world transport across the sea. Transport of crude oil and its products across the Arabian Sea amounts to 990×10^6 tonnes/year (Wong *et al.*, 1976). This is about 58.3% of the total marine transport. Of this, 340×10^6 tonnes are carried to Far East and Japan. The effect of transport of this amount is of direct concern to India. If the oil spill has a relationship with its total transport, then the Arabian Sea should be highly contaminated.

Petroleum hydrocarbon concentrations in the Arabian Sea varied from 0.12 to 2.44 mg/l (average 1.20 mg/l) at the surface (Figure 2), and from 0.44 to 2.39 mg/l (average 1.04 mg/l) at 10m (Table 1). The

Table 1. Concentrations of Petroleum Hydrocarbons at 10m Depth

<i>Station No.</i>	<i>Concentration (mg/l)</i>
295	0.64
297	2.39
303	1.49
310	1.89
315	0.52
321	0.84
326	0.44
330	1.01
334	0.76
337	0.69
344	0.74

average of all the values was 1.14 mg/l. Our values were higher than those from the other oceanic regions (Levy, 1971; Barbier *et al.* 1973; Keizer *et al.* 1977). Those analyses were conducted by Gas-Liquid Chromatography while we did ours by UV-spectrophotometry observing all the necessary precautions in sampling and analyses.

To analyse the data, we divided our area of study as north and south of 15°N latitude. Our transects were quite close to the oil tanker route north of 15°N while south of this latitude we moved closer to the Indian coast.

North of 15°N

This region is full of shipping traffic carrying petroleum and its products and also general cargo. In this region concentrations of petroleum hydrocarbons varied from 0.52 to 2.44 mg/l (average 1.50 mg/l). It was observed that along the 100-fathom isobath, the concentration is

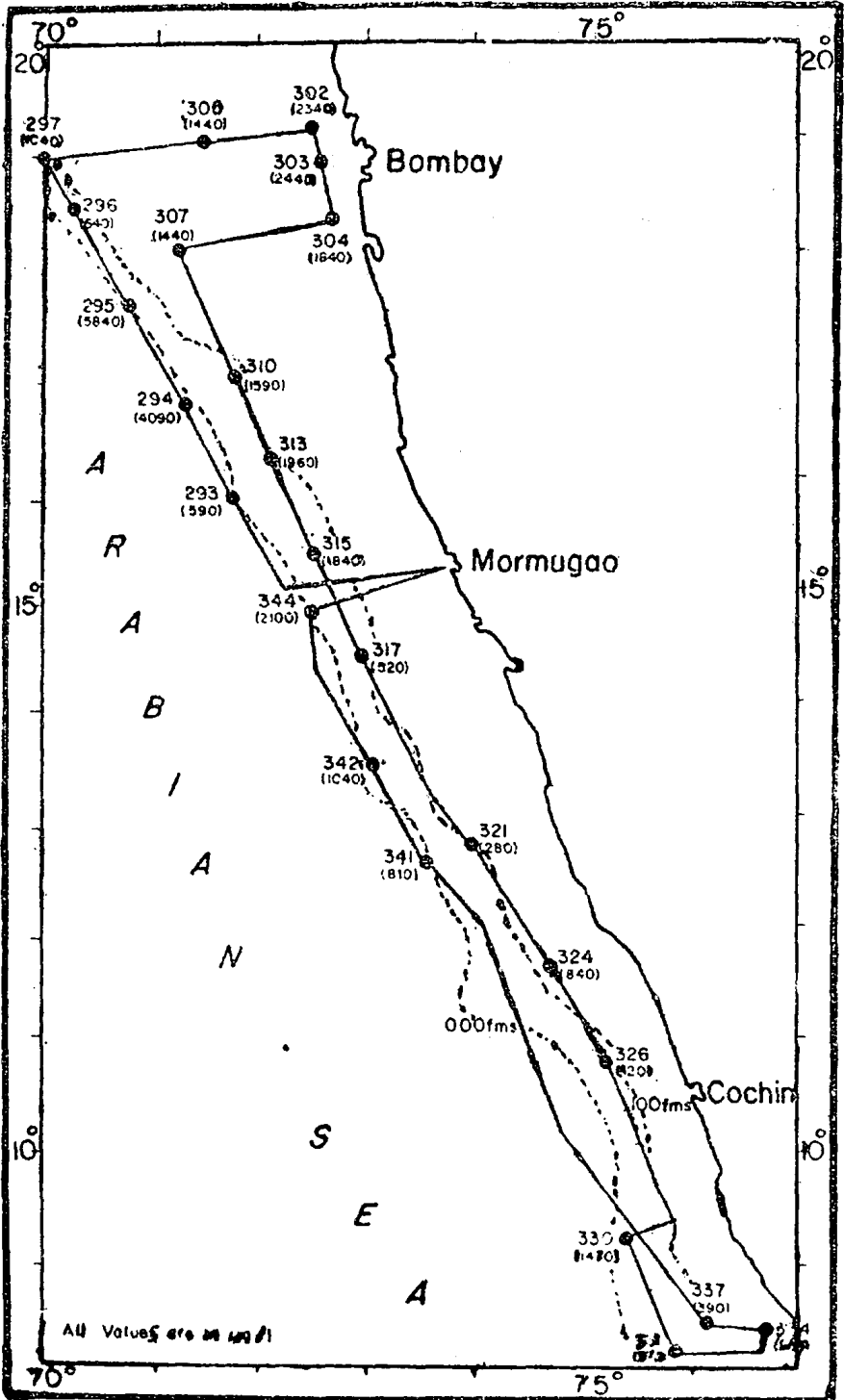


Fig. 2. Concentrations of Petroleum Hydrocarbons (in brackets) in the surface waters of the observation stations in $\mu\text{g/l}$.

greater (average 1.06 mg/l) than along the 1000-fathom isobath. This is possibly because of the closeness to the coast; which receives petroleum-derived pollutants from land, discharges from fishing trawlers, harbours and other industrial activities. Thus these higher values can be directly related to human activities.

Off Bombay (stations 300, 302, 303, 304 and 307), the surface concentrations were high and varied between 1.44 and 2.44 mg/l (average 1.90 mg/l). This region being near the coast and adjacent to one of the busiest ports and industrial areas of the country, the concentrations may be expected to be more than in other regions. This area is also close to Bombay High and some natural seepage can also be expected to occur here.

South of 15°N

This region does not appear to be very much affected by tanker traffic as compared to north of 15°N. Concentrations varied from 0.12 to 2.10 mg/l (average 0.80 mg/l). In this area, the concentrations were found to be less along the 100-fathom isobath (average 0.58 mg/l) than those along the 1000-fathom isobath (average 1.16 mg/l). In other words, the condition in this region was found to be reverse of what was observed north of 15°N, where the concentration was greater along the 100-fathom isobath. The 1000-fathom isobath is not very far from the tanker route. Also some hydrocarbon can be expected to be released by microbial degradation.

At station 344, the concentration of hydrocarbon at the surface was 2.10 mg/l, which is the highest value in this region. This may be due to the presence of tar-like material, which was observed floating on the surface at the time of sampling.

Petroleum and its products change their character in a short time in the sea. C₁ to C₁₂ hydrocarbons are the first to evaporate followed by fractions upto C₂₄ (Farrington *et al.*, 1976). The heavier fractions above these are almost undecomposable. Dissolution, chemical oxidation, volatilization, biodegradation, light-induced reactions are the other processes; which contribute to change the nature of oil in the sea (Keizer and Gordon, 1973). All these factors have to be considered in assessing petroleum hydrocarbon concentrations in the sea.

As mentioned earlier, our values are higher than those reported from other oceanic areas. Whereas oceanic values range from 0.1 mg/l, we obtained a range of 0.12 to 2.44 mg/l with 53% of the values lying above 1 mg/l. This might be the effect of the transport of large volume of oil across the Arabian Sea.

CHLORINATED HYDROCARBONS

The insecticide DDT (Dichloro-diphenyl trichloroethane) is a member of a class of organic compounds known as 'chlorinated hydro-

carbons', which are used in agriculture, health and commerce, because of their toxicity to insects. Since it is a compound, which is quite persistent, it ultimately finds its way to the ocean. High concentrations of DDT found in the marine environment has led to a total ban of this in USA, Europe and Japan and restricted the use of other chlorinated hydrocarbons (Harvey, 1974). There is, however, no data available on their levels of concentration from the seas around India. India is a predominantly agricultural country. Insecticides and pesticides have been in use in abundance in agriculture and pest control since many years. On an average about 34 000 tonnes of pesticides and insecticides are used every year in India for agricultural purposes. The cumulative effect of these chlorinated hydrocarbons on our coastal waters can be expected to be considerable.

Chlorinated hydrocarbons are, in general, fat-soluble and hence can be concentrated by marine organisms. To determine their concentration in water large volume of water samples have to be extracted. We tried upto 5 litres without success and hence concentrated our studies only on zooplankton samples. 12 samples were collected for this; but on closer examination, only 5 of them were found to be free from floating tar particles and paint chips. We report here their chlorinated hydrocarbon concentrations, as t-DDT (total DDT), determined by Gas-Liquid Chromatography. The results obtained from the analyses are presented in Table 2. As can be seen from the table, a high concentration of t-DDT,

Table 2. DDT and Metabolites in ppm Fresh Weight

Station No.	O-p DDT (a)	p-p' DDT (b)	O-p DDE (c)	p-p' DDE (d)	Others (PCB & BHC)	Extract-able lipid (%)	t-DDT (a+b+c+d)
293	2.00	0.90	ND	0.31	P	6.00	3.21
297	1.28	ND	0.15	ND	P	3.70	1.43
299	0.94	ND	ND (Trace)	0.33	P	2.30	1.27
304	ND	ND	ND	0.05	ND	1.60	0.05
309	ND	ND	ND	ND	ND	2.00	—

ND=Not Detectable

P=Presence indicated

depending on the fat content, was recorded. The presence of BHC (Benzene hexachloride) and PCB (Polychlorinated biphenyls) was also noticed. No attempt was, however, made to quantify these peaks

(figure 3) as the components were not separated from one another. Although the chromatograms obtained were not complex the interference from certain other chlorinated hydrocarbons, specially PCB, cannot be ruled out and more so because the PCB peaks are found to interfere with those of DDT, and this may be the reason for the higher values.

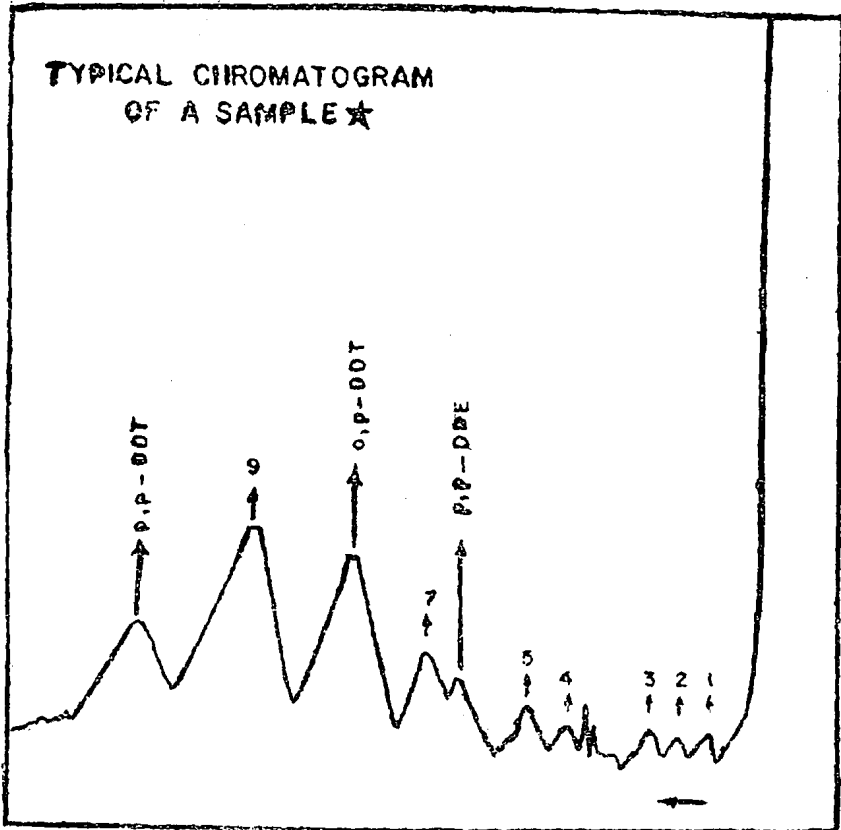


Fig. 3. Typical gas-chromatogram of a plankton sample for chlorinated hydrocarbons.

* = Numbers showing unidentified peaks.

Peaks 1, 2 & 3 — may be BHC.

Peaks 4, 5, 7 & 9 — may be PCB.

Since the chlorinated hydrocarbons are lipophilic compounds, it becomes rather difficult to interpret the data, because of the varying lipid contents of the organisms themselves. This is even more difficult in plankton, which have a variation in their lipid content with seasons, together with a seasonal change in the quantity of inert material related to species composition (Williams and Holden, 1973). However, from the present study; it becomes evident that the plankton between Bombay and

Goa, do contain high levels of t-DDT. This should be a cause of concern as they constitute the major source of food for animals, higher in the food chain.

HEAVY METALS

The sea route is now-a-days used more frequently for business and other purposes and consequently considerable variations in the trace metal contents can be expected to occur. Metal-ions present in atmosphere and soil also enter the marine system through precipitation and runoff. Most of the heavy metals are present in sea water at concentrations in parts per billion (PPB) range. They exhibit an enormous range of chemical reactions and are closely related to the biogeochemical cycle in the marine environment. The transition group of metals, especially those with half-filled d-shells, have a tendency to attach to donor atoms to undergo bioaccumulation. Hence; they need more attention to study possible hazards to marine biota.

Metals in certain concentrations are desirable for the growth of marine organisms, but their over-accumulation is equally hazardous. A case on point is mercury, which is considered as the most toxic of all heavy metals. Its over-accumulation by organisms is hazardous to the organisms themselves as well as to those who consume them directly or indirectly through their food. The toxic character of this metal roused worldwide attention since the tragic incident of the 'Minamata disease' in Japan. Information about concentrations of heavy metals in marine environment of India is few and far between. With rapid industrialization in our country; the risk of discharge of heavy metals, both toxic and non-toxic, to the marine system is gradually increasing. This attempt of ours may, probably, be considered as the first of its kind to collect baseline information.

We applied the standard-addition atomic absorption spectrophotometry (AAS) to analyse water samples for Cu, Fe, Zn, Ni, Co and Mn. Their concentrations in plankton samples and in particulate matter were determined by first digesting the material before running them into AAS. Mercury was determined by the cold-vapour AAS.

The concentrations of the metals in water, particulate matter and zooplankton samples are presented in Tables 3 and 4. No particular trend of depthwise variations could be noted for almost all the metals in their dissolved state. Average concentration of *copper* was found to be $4.7 \pm 0.4 \mu\text{g/l}$ at 10 m and the values at most of the stations were around the average. The highest value observed was $7.7 \mu\text{g/l}$ at station 317 about 40 km off the coast of Goa. Higher values of copper at depths than at the surface at shallow stations indicate higher accumulation of the metal in sediments. It has been observed (Sen Gupta, 1972) that some of the organisms in the sea can concentrate copper. At shallow stations quite frequently higher copper values near the bottom were observed indicating

that the fraction of this metal, concentrated at the surface, has been released by the process of decomposition. In deeper stations, lower values were obtained for dissolved copper at mid-depths while the particulate fractions were considerable. Deeper down, however, there was an increase in the dissolved fraction. Solubility of copper complexes is very much dependant on the redox condition in water (Latimer, 1952). In the Arabian Sea, oxygen concentrations have been observed to be very low between 100 and 800 m (Sen Gupta *et al.*, 1976). This indicates that conditions at mid-depths were not sufficient to dissolve the copper accumulated at the surface. Average concentration of iron at 10 m was $26.5 \pm 4.8 \mu\text{g/l}$. The highest value observed was $66.9 \mu\text{g/l}$ at station 320 about 70 nm off the coast. Off Bombay, the values were of intermediate concentrations. Observed concentrations of iron give a measure of truly soluble complexes of Fe^{3+} ion and colloidal ferric hydroxide. Ferric ion present as $\text{Fe}(\text{OH})^{2+}$ cannot exceed concentration of the order of $1 \mu\text{g/l}$ (Stumm and Morgan, 1970). Hence, most of the iron concentration in our data can be attributed to soluble complexes and colloidal Fe^{3+} compounds. Richness of the deep water in both the particulate and soluble fractions of iron probably indicates that the metal settled as insoluble iron hydroxides. Table 3 combined with Table 4 also indicate that the metal is not concentrated to any appreciable extent by the organisms at the surface, which is in agreement with earlier observations from the Indian seas that iron is not a limiting factor for plankton production in the sea (Subrahmanyam and Sen Gupta, 1965). Thus, it can be concluded that in the Arabian Sea, iron is present mostly in suspended and particulate forms.

Average concentration of zinc at 10 m is $20.8 \pm 3.0 \mu\text{g/l}$. Concentrations at stations off-shore are lower than the average while nearshore they are higher. Highest concentration of zinc observed was $42.4 \mu\text{g/l}$ at station 320, where the highest concentration of iron was also noted. This is obviously due to coastal input. Zinc concentrations in pre-treated sewage near almost all the big cities show, on long term measurements, regularly occurring high concentrations interspersed with normal values (Sen Gupta, 1972). Concentrations of zinc in the near-shore waters and sewage of Bombay are fairly high, 3.4 - 347 and 0.2 - 221 $\mu\text{g/l}$ for water and sewage respectively (N.B. Bhosle and Analia Braganca - personal communication). We attribute the addition from land as the cause for high zinc concentrations in some of our samples.

Zinc is concentrated by almost all the organisms in the sea in varying quantities. Combining our values for particulate and suspended fractions with those for zooplankton (Table 4) we find that the latter makes up for a significant fraction at selected localities.

Our attempts to measure cobalt and nickel showed very low or non-detectable concentrations in water and in particulate matter. The

Table 3. Concentration of Metals

Station No.	Total depth(m)	Sampling depth in (m)	Cu		Fe		Zn		Co		Ni		Mn	
			Dissolved $\mu\text{g l}^{-1}$	Particulate $\mu\text{g g}^{-1}$	Dissolved $\mu\text{g l}^{-1}$	Particulate $\mu\text{g g}^{-1}$	Dissolved $\mu\text{g l}^{-1}$	Particulate $\mu\text{g g}^{-1}$	Dissolved $\mu\text{g l}^{-1}$	Particulate $\mu\text{g g}^{-1}$	Dissolved $\mu\text{g l}^{-1}$	Particulate $\mu\text{g g}^{-1}$	Dissolved $\mu\text{g l}^{-1}$	Particulate $\mu\text{g g}^{-1}$
292	1700	10	6.8	65.0	—	260	—	5734	1.0	ND	4.4	ND	—	148
		500	2.8	36.2	—	2420	—	1804	0.5	ND	4.4	ND	—	58
294	1700	10	—	174	—	2720	—	3329	—	ND	—	—	—	43
		100	—	427	—	2393	—	1692	—	ND	—	—	—	26
296	350	500	5.0	127	11.0	746	5.7	944	2.0	ND	4.0	ND	—	22
		1000	7.9	201	13.3	1846	6.3	1114	0.8	ND	2.1	ND	—	40
		10	6.6	53	10.0	522	5.5	258	1.0	ND	3.9	ND	—	528
297	180	100	6.7	135	18.2	935	7.5	1619	3.3	ND	4.0	ND	—	600
		280	4.6	139	14.1	350	5.8	723	4.9	ND	1.0	ND	—	15
		10	5.0	131	11.7	1619	25.4	4450	3.3	ND	1.5	ND	—	69
301	49	100	3.5	196	10.4	2186	31.5	2505	4.1	ND	2.5	ND	—	41
		150	6.0	91	13.1	4591	16.0	5366	1.6	ND	2.0	ND	—	161
303	22	10	4.8	126	25.9	1880	29.4	4526	ND	ND	2.0	ND	—	74
		45	7.3	234	12.8	1430	16.1	1591	ND	ND	1.5	ND	—	117
307	86	10	2.5	33	10.4	22187	18.3	1464	ND	ND	1.5	ND	—	675
		20	6.3	59	16.8	25207	14.2	1089	ND	ND	2.5	ND	—	882
307	86	10	3.1	170	16.2	4236	8.0	5264	ND	ND	1.0	ND	—	19
		75	1.7	147	8.0	6819	7.6	11787	0.8	ND	3.0	ND	—	213

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315	900	10	3.8	99	19.7	3423	16.1	10845	0.8	ND	5.0	ND	14
		500	5.2	183	14.1	515	23.3	13014	6.6	ND	4.5	ND	84
		800	3.1	414	25.0	5397	16.5	13759	3.3	ND	11.5	ND	34
	240	10	7.7	301	8.4	1913	37.6	4495	2.8	ND	4.0	ND	19
		200	4.5	71	11.9	1388	8.8	3976	2.4	ND	2.0	ND	12
		10	5.0	355	66.9	2605	42.4	7342	ND	ND	3.5	ND	92
320	70	65	4.5	39	34.4	8407	27.2	3506	3.3	ND	4.5	ND	86
		10	4.4	524	35.7	3405	16.9	7690	3.3	ND	ND	ND	42
324	87	80	2.5	232	11.6	7488	29.0	305	2.4	ND	2.0	ND	134
		10	4.4	250	14.3	70500	16.0	204750	ND	ND	2.5	ND	3333
334	44	40	5.8	1417	21.9	69583	23.1	162583	ND	ND	1.5	ND	917
		10	4.2	400	49.5	32500	15.1	88800	0.8	ND	3.0	ND	3200
338	1700	100	7.7	700	19.2	13900	19.6	68900	2.4	ND	2.0	ND	800
		500	4.2	450	23.0	25750	31.2	69650	ND	ND	4.5	ND	450
		1000	3.4	500	20.0	48000	18.0	154700	0.8	ND	4.0	ND	500
344	1400	10	3.3	176	29.8	14176	19.5	83235	0.8	ND	1.0	ND	823
		100	5.8	111	07.2	24333	37.9	88389	0.8	ND	4.0	ND	778

ND=Not Detectable

Table 4. Concentration of Metals in Zooplankton* and Particulate Matter at 10m (All Values in mg/g DRY Weight).

Station No.	<u>Cu</u>		<u>Fe</u>		<u>Zn</u>		<u>Co</u>		<u>Ni</u>	
	Plank- ton	Parti- culate	Plank- ton	Parti- culate	Plank- ton	Parti- culate	Plank- ton	Parti- culate	Plank- ton	Parti- culate
315	72	99	1010	3423	1382	10845	ND	ND	ND	ND
320	111	355	762	2605	942	7342	23	ND	17	ND
334	10	250	1923	70500	9274	204750	ND	ND	ND	ND
338	79	400	2984	32500	22494	88800	34	ND	ND	ND
Concent- ration factor	2060		6487		31370		2989			

* Kureishy and George (1977).

depthwise variation of the two metals are erratic, but an almost approximate trend of decrease could be noted. It has been suggested (Sen Gupta, 1972) that organisms might concentrate nickel at the surface which can be released at depths. Our data in Table 3 give an indication that these metals are not present in particulate forms, whereas they can be selectively absorbed by the organisms as a local phenomenon. According to the so called "Irving-Williams order", both nickel and cobalt complexes are among the most stable of the transition metal cations (Goldberg, 1965). In water solution, especially in the presence of carbonate ions, the chemistry of cobalt, nickel and their compounds are very largely those of the bivalent forms Co^{2+} , Ni^{2+} (Latimer, 1952). From published data, it has been concluded (Goldberg, 1965) that the concentration factor for cobalt and nickel, calculated according to Liebig's "Minimum Law", is among the lowest in the transition group metals. We could not calculate the concentration factor for nickel for want of representative data, but that for cobalt is one of the lowest among the transition group metals (Table 4). Probably the small amount of nickel that could be concentrated by organisms in the surface layer is released in the deep water and being able to form some highly stable bivalent compound, remains almost unchanged in the water column. Their absence in particulate form even at depths probably indicate that these metals settle to the bottom.

Our mercury values include both the dissolved and a part of the particulate mercury that could be dissolved after acidification with 9N H_2SO_4 . We, therefore, express our values as total mercury concentration in the samples. Concentrations are presented in Table 5. The lowest concentration observed was 13 ng/l and the highest concentration was 407 ng/l. We have presented all the surface values in Figure 4.

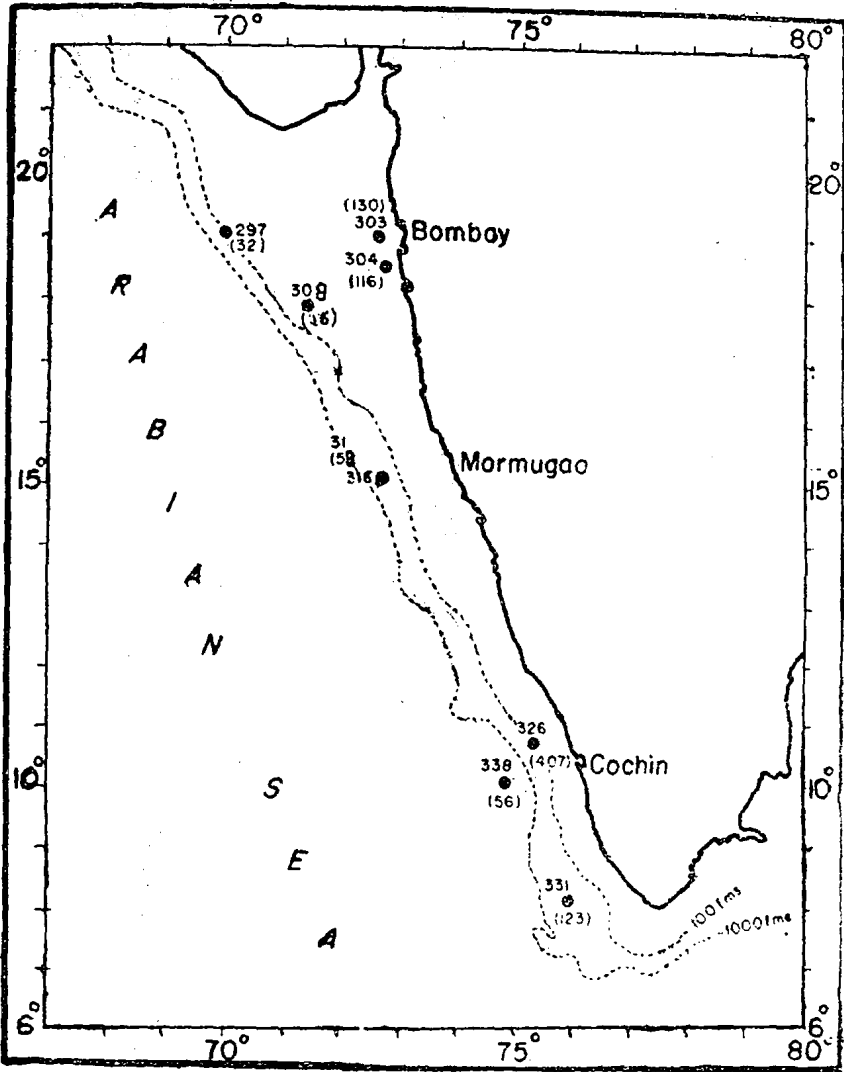


Fig. 4. Concentration of total mercury (in brackets) in the surface waters of the observation stations in ng/l.

Excluding the high value, we could obtain a surface average of 77 ng/l which compares well with the accepted global range of 10-50 ng/l, if we consider that our values include a part of particulate mercury also. In general, deeper mercury concentrations are lower than those at the surface layers. The decrease appears to be quite appreciable in the depth range between the thermal discontinuity layer and 500 m which is the region of low oxygen concentration in the Arabian Sea. This supports the view that mercury has strong association with suspended inorganic and organic

Table 5. Total Mercury Concentrations in Arabian Sea Waters

<i>Station No.</i>	<i>Depth (m)</i>	<i>Concentration ng/l⁻¹</i>
297	1	32
	100	52
	150	13
303	1	130
	10	71
	20	84
304	1	116
	10	142
	15	32
308	1	26
	25	32
	50	136
	75	84
315	1	58
	100	52
	500	26
	800	39
316	1	—
	10	187
	65	90
326	1	407*
	50	181
331	1	123
	500	90
	1200	181
338	1	58
	500	187
	1000	110

Surface average 77 ng/l⁻¹.

* Excluded from calculation. Reason in the text.

matter in the marine environment. However, values at surface decrease with increasing distance from the shore. The sole high value of 407 ng/l at station 326, may be due to the effect of leaching of native mercury from the crustal rocks. It is believed that the rocks between Calicut and Karwar on the west coast of India contain native mercury (Mr. H.N. Siddiquie - Personal communication). Methylation of mercury takes place at or near the surface of the ocean. Some of the ions are absorbed by

particulate matter, which settles down to the bottom in due course getting incorporated in the sediments. When the sediment surface is subjected to microbial degradation mercury ions, among others, can be expected to be leached out to the overlying water. These can be the reasons for higher mercury concentrations at depths at stations 331 and 338.

The spatial variation at the surface might be due to qualitative and quantitative differences in the composition of land drainage, atmospheric flux or the effect of upwelled water. Such factors have been observed to affect the concentrations of mercury in the surface waters of the oceans (Chester *et al* , 1973).

Finally, we make the reservation that this report presents only a preliminary assessment of the pollution situation in the Arabian Sea. The work is by no means complete. We have already completed three more cruises for this purpose in the Arabian Sea and the Bay of Bengal and propose to undertake several more. In this way, we expect to be able to present a total picture about the health of the seas around India.

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