

## Aluminium in the northwestern Indian Ocean (Arabian Sea)

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### Abstract

The distribution of Al has been studied along two transects ( $\sim 15^\circ$  and  $18^\circ\text{N}$ ) in the Arabian Sea. The waters of the Arabian Sea along the Indian coast are relatively enriched in Al ( $\sim 55\text{--}74\text{ nM}$ ) over the offshore surface waters ( $\sim 37\text{--}52\text{ nM}$ ). A pronounced maximum in the surface mixed layer suggests the dissolution of Al from atmospherically derived particles to be the source of excess Al in the offshore waters of the Arabian Sea, compared to other oceanic regions. The characteristic Al decrease in the deeper waters of the Arabian Sea indicates its intense scavenging in the water column. A residence time of  $\sim 8.4\text{ yr}$  for Al in the Arabian Sea, with respect to the combined input of Al from fluvial and atmospheric sources, corroborates the high reactivity of Al in the oceans.

### 1. Introduction

Dissolved aluminium is highly reactive in aqueous solutions and, hence, is removed faster from seawater than many particle-reactive elements, whereas its relative abundance in coastal and offshore surficial waters could be an indicator of the influence of terrestrial material on the oceans. Inorganic processes have often been invoked to understand the marine geochemistry of Al (Hydes, 1979; Moore, 1981; Orians and Bruland, 1985); however, some evidence suggests that biological mechanisms could also influence the Al cycling to a significant degree (Stoffyn, 1979; Van Bennekom, 1981; Moran and Moore, 1988; Van Bennekom et al., 1989).

The vertical profiles of dissolved Al in the north Atlantic (Hydes, 1979, 1983; Measures et al., 1986) and the Pacific Oceans (Orians and Bruland, 1985, 1986) are found to exhibit surface enrichment, a mid-depth minimum and a gentle rise in concentrations near the bottom. In contrast, Al in the Mediterranean co-varies with silicate exhibiting nutrient type surface depletion and enrichment at depth (Caschetto and Wollast, 1979; Stoffyn and Mackenzie, 1982). However, the reasons for this contrasting behaviour and the factors controlling the Al distributions in seawater are not well known.

Since the Arabian Sea is surrounded by land on three sides, Al can be transported to the sea via various pathways, such as river runoff, deposition of atmospheric aerosols and coastal dumping of Al wastes. The northern Arabian Sea is more vulnerable to these processes than the southern part because of the closer proximity of the former to

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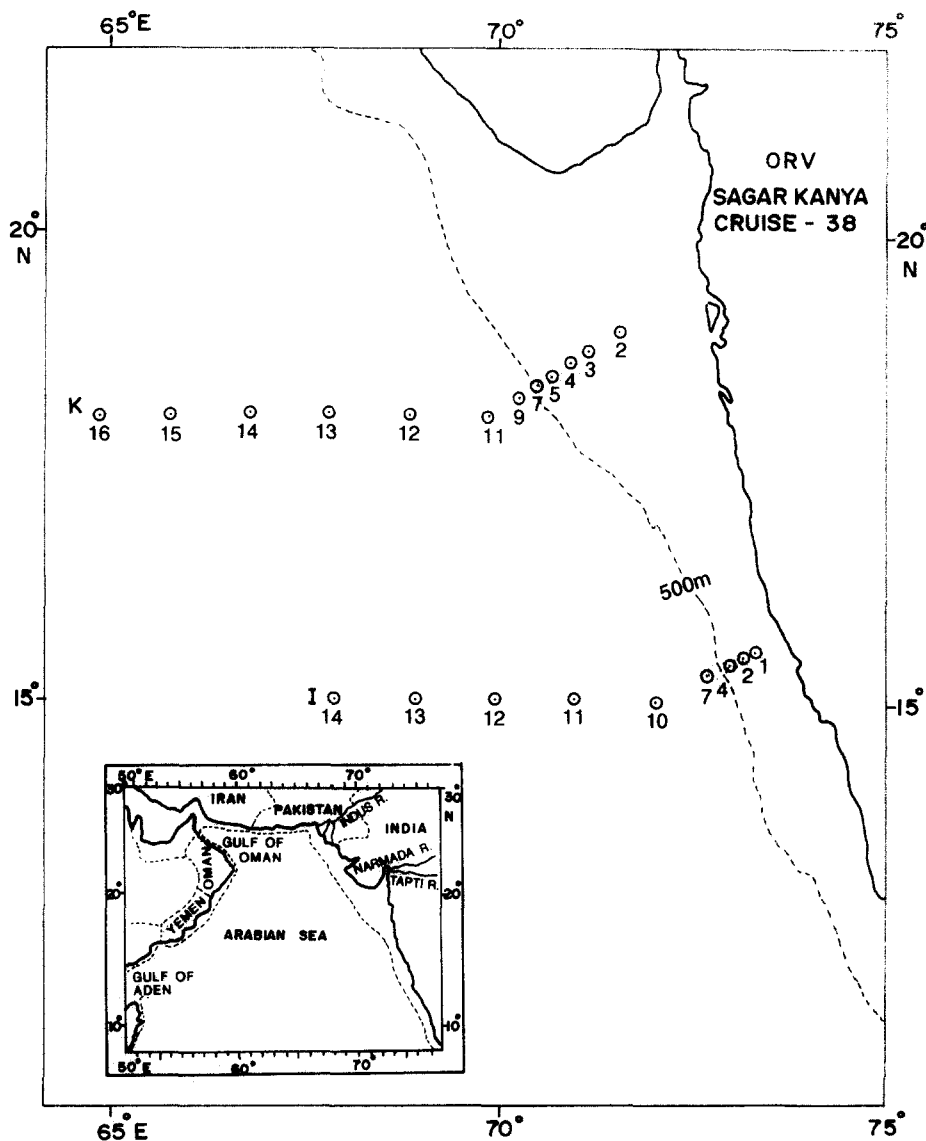


Fig. 1. Location of sampling stations in the Arabian Sea (dotted line represents the 500 m isobath).

land. Data from the eastern Arabian Sea reveal high and variable concentrations of dissolved Al in the surface microlayer and surface waters (Narvekar and Singbal, 1993). However, little information is available on the geochemistry of Al in the deeper layers of the northwestern Indian Ocean. Hence, the aim of this study is to obtain insight into the biogeochemical factors regulating the distribution and behaviour of Al in the Arabian Sea.

## 2. Materials and methods

The present study covered the central eastern part of the Arabian Sea: between 15 and 20°N and 64 and 74°E (Fig. 1). Two latitudinal transects, one off Marmugao (Goa) and the other off Bombay designated as I- and K-transects, respectively, were chosen. This study was carried out on board the ORV *Sagar Kanya* during her 38th cruise (January–February, 1988).

A PVC-coated wire was used for the collection of samples from standard depths in the Arabian Sea. Niskin samplers (51), with silicone rubber O-rings, were used along with plastic messengers. Reversing thermometers were used for temperature measurements. Data were collected for temperature, salinity, dissolved oxygen, phosphate, silicate and aluminium. Standard methods were adopted to estimate nutrients and other hydrographic parameters (UNESCO, 1981; Grasshoff et al., 1983).

The analytical technique employed in the determination of aluminium was the Lumogallion method of Hydes and Liss (1976). The samples were analysed using a standard addition technique in triplicate of 10 ml each. The samples were allowed to stand for 10–12 h at room temperature (25–30°C) in subdued light after the addition of the reagents. The fluorescence of the samples was measured using an excitation wavelength of 465 nm and an emission wavelength of 555 nm employing a Perkin-Elmer LS-3 Fluorescence Spectrometer.

With the use of analytical reagent grade chemicals, the reagent blank corresponded to approximately 37 nM Al. The estimated standard deviation of the reagent blank was  $\pm 3$  nM Al for 10 replicates. This yields a detection limit of about 3 nM Al (one standard deviation of the blank) for the method, which is low enough for a reliable analysis of ocean water. The precision of the analysis was usually within  $\pm 5\%$  at 74 nM Al. However, at low Al concentrations ( $\leq 37$  nM) a slightly larger deviation up to 8% was observed.

The analyses carried on unfiltered and filtered (through 0.4  $\mu\text{m}$  Nuclepore membrane filters) seawater samples yielded almost identical results. The differences (not more than 1.5 nM) were well within analytical uncertainties. Hence, the seawater samples were not filtered to avoid the possible risk of contamination. At a pH of  $\sim 5$  the dissolution of particulate aluminium is found to be negligible (Measures et al., 1984). However, aluminium estimated in an unfiltered seawater sample might include a minor fraction of adsorbed species and some in colloidal suspension together with Al in solution.

### 3. Results and discussion

#### 3.1. Hydrographic features of the Arabian Sea

The coastal waters off Goa (I-transect) were characterized by low salinity ( $\sim 35.4$ ), relatively low nutrient content ( $\text{PO}_4 < 0.2 \mu\text{M}$  and  $\text{Si} < 0.1 \mu\text{M}$ ) and high dissolved oxygen ( $\sim 200 \mu\text{M}$ ). Similar high  $\text{O}_2$  ( $\sim 200 \mu\text{M}$ ), low nutrients ( $\text{PO}_4 < 0.5 \mu\text{M}$  and  $\text{Si} < 0.1 \mu\text{M}$ ) but moderately high salinity ( $\sim 36$ ) were observed off the Bombay coast (K-transect). These represent the typical highly productive coastal waters of the Arabian Sea.

Nutrients were strongly depleted in the surface waters of the study area, suggesting intense biological activity in the mixed layers. Across the seasonal thermocline, at deep-sea stations, there was a sharp drop in the dissolved oxygen content which continued to decrease with depth resulting in a pronounced oxygen minimum ( $\sim 4.5 \mu\text{M O}_2$ ) in the water column. This was associated with an increased nutrient content with depth, indicative of intense oxidation of organic matter. Development of such a reducing environment in the Arabian Sea has been well-documented (Sen Gupta et al., 1976; Naqvi, 1987; Kumar, 1988).

##### 3.1.1. Distribution of Al

The coastal waters of the Arabian Sea are relatively enriched in Al ( $\sim 55$ – $74$  nM) over the offshore surface waters ( $\sim 37$ – $52$  nM). Similar high Al concentrations in the coastal waters of the Panama Basin (Measures et al., 1984), the European shelf (Kremling, 1985) and the German Bight and the French coast (Kremling and Hydes, 1988) associated with fluvial input have been reported. The highest levels of Al in the relatively low-salinity ( $\sim 35.4$ ) waters off the Goa coast (stations I-1 to I-4) compared to the low level of Al in high salinity ( $\sim 36$ ) waters off the Bombay coast (stations K-2 to K-7) conform with the fluvial sources, assuming that contributions from external sources like aerosols are of similar magnitude at both the locations. This attribution is made in view of draining of many small rivers into the former in contrast to the latter.

The concentration of Al ( $\sim 37$ – $52$  nM) in the open surface waters of the Arabian Sea is higher than in the Atlantic (16–33 nM, Hydes, 1983; 29–43 nM, Measures et al., 1984) and the Pacific (0.3–6 nM, Orians and Bruland, 1985, 1986). Our values, however, are consistently lower than those previously reported for the eastern Arabian Sea (23–657 nmol kg<sup>-1</sup> in the surface microlayer and 11–296 nmol kg<sup>-1</sup> in the surface waters; Narvekar and Singbal, 1993) which were obtained using the Pyrocatechol Violet spectrophotometric method.

Dust storms from the Thar desert of India and the arid zones of Arabian regions cross over the northern Arabian Sea (Kolla et al., 1981). The impact of atmospheric aerosols on the chemistry of Al in open surface seawater is known (Maring and Duce, 1987). Atmospheric Al concentrations over the northern Arabian Sea averaged 1227 ng m<sup>-3</sup> and are among the highest recorded for the marine environment (Chester et al., 1985, 1991). Hence, atmospherically transported land-derived material might contribute to such a high concentration of Al in the Arabian Sea.

The subsurface waters contain a reduced level of Al (<37 nM) which is quite comparable with other oceans (Hydes, 1979; Moore and Millward, 1984; Measures et al., 1986). These values are, however, much higher than those found in the northeast Atlantic ( $\sim 10$  nM, Hydes, 1983), the Pacific (<1 nM, Orians and Bruland, 1986), the southern Atlantic (3–6 nM, Measures and Edmond, 1990) and the Antarctic (1.5–2.5 nM, Van Bennekom et al., 1991). This inter-ocean variability may be related to geographical variations in atmospheric Al sources, magnitude of riverine Al fluxes and the very short residence time (<10 yr) of Al in the seas (see below).

### 3.1.2. Onshore–offshore variations of Al in surface layers

In view of the well-mixed nature of upper 30 m of surface water, the values of Al at 1, 10, 20 and 30-m have been averaged and plotted in Fig. 2a and b for the I- and K-legs, respectively. Al concentrations were high in low-salinity coastal waters and decreased with distance away from the shore. This trend resembles the Al distribution off the

German and French coasts (Kremling and Hydes, 1988) and is probably associated with continental runoff and terrigenous mineral dissolution. The study on the Mandovi estuary (Upadhyay, unpubl. data) reveals that only a small portion of the dissolved Al from the river water is removed in the estuary. Hence the excess Al concentrations in the near-shore waters along the I-transect are possibly due to strong fluvial sources, the most important of which is the Mandovi–Zuari estuarine system. Moreover, the remobilization of Al from pore waters or sediments can also act as a source for the coastal elevations.

A steady decrease of the surface Al level towards offshore along both the transects may be diffusion-controlled or, more probably, caused by the current system. The onshore–offshore gradients indicate that, apart from the atmospheric sources, fluvial input also seems to be significant in the Arabian Sea. The atmospheric dust influence could be seen near the extreme offshore stations. There is a general increase of Al level from station 10 to station 13 in the I-transect and from station 14 to station 16 in the K-transect. This is in good agreement with the trend observed for Al in the surface microlayer and surface waters in the same region (Narvekar and Singbal, 1993).

### 3.1.3. Vertical profiles of Al

Variation in Al levels is more prominent at station I-1 (Fig. 3a) than at station I-2 (Fig. 3b). In contrast, the vertical profiles at stations K-2 and K-5 (Figs. 3c and d) are almost invariant with depth. Station I-1 is very shallow and is located at the mouth of the Mandovi and Zuari estuarine system in Goa. Such variations observed at station I-1 could arise due to the influence of fluvial input and sediment disturbance (Mackin and Aller, 1984). The latter process could be seen from the near-bottom slight increase of Al at stations I-1, I-2, and K-5. This observation is, however, limited to one point only. During organic production, biological activity might remove some amount of dissolved Al (Measures et al., 1986; Moran and Moore, 1988) but due to strong fluvial and aeolian sources the effect of biological removal is obscured.

Vertical profiles of Al and other physiochemical

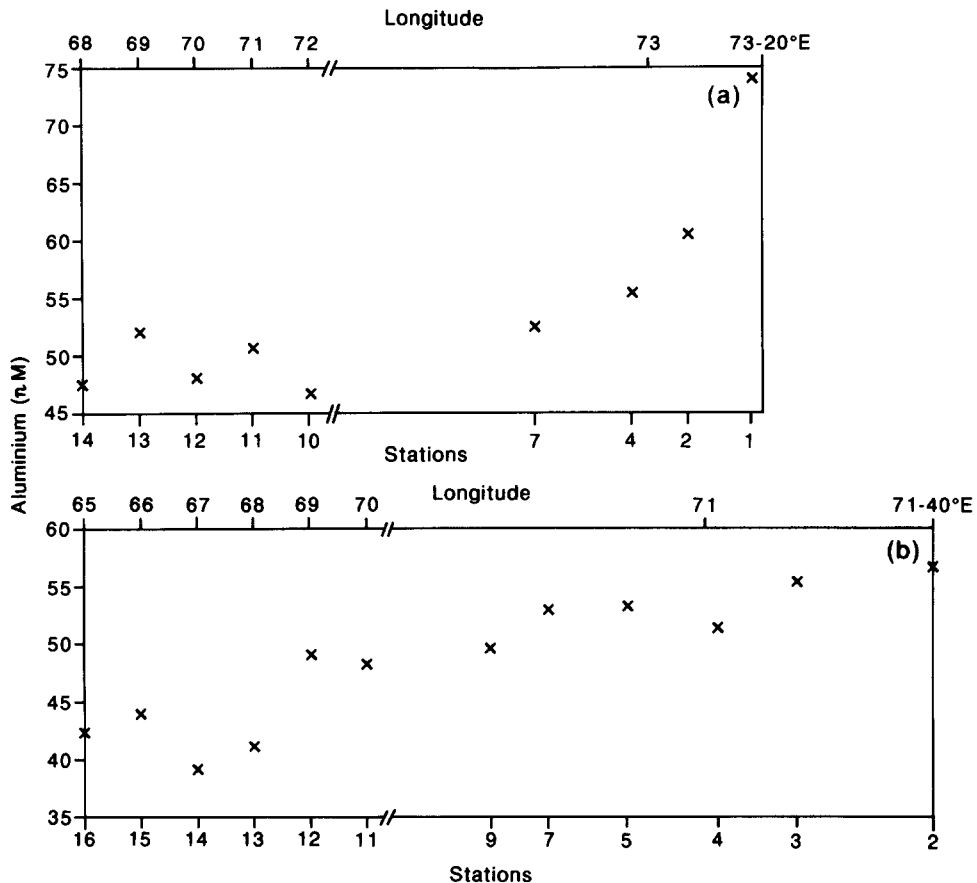


Fig. 2. Longitudinal distribution of Al in the surface mixed layer along (a) the I-transect (~15°N), and (b) the K-transect (~18°N).

parameters at some selected offshore stations are plotted in Figs. 4a–e. At almost all the deep stations, the Al maximum in the surface mixed layer is conspicuous. The pronounced surface maximum in Al in the Arabian Sea is compatible with the river supply coupled with atmospheric dust inputs. The relatively higher levels of Al in these waters suggests the dissolution of Al from atmospherically derived particles as the source of excess Al in the surface waters (Hydes, 1983; Maring and Duce, 1987) when compared to other oceanic regions (Moore, 1981; Moore and Millward, 1984). This is in conformity with the observations made by Narvekar and Singbal (1993).

As is the case for dissolved Si, rivers are also one of the major sources of Al in the Arabian Sea (Upadhyay, unpubl. data). Dissolved Si is strongly depleted in the surface mixed layers.

Though no such trend for Al could be found, some fraction of it may also be incorporated into planktonic cells which is perhaps replenished by aerosol dissolution.

Aluminium levels show a decrease in the sub-surface waters and mostly a depletion in the Al concentration in deeper layers of the water column is apparent. The mid-depth minimum is, however, much shallower than that found in the Atlantic (Hydes, 1979; Measures et al., 1986) and the Pacific (Orlans and Bruland, 1985, 1986). This is most likely an outcome of the high surface deposition rate of Al and its diffusion into the subsurface layers. In addition, the advection of Al-enriched coastal waters probably plays a significant role.

Aluminium concentrations in some profiles do not vary significantly with depth below the

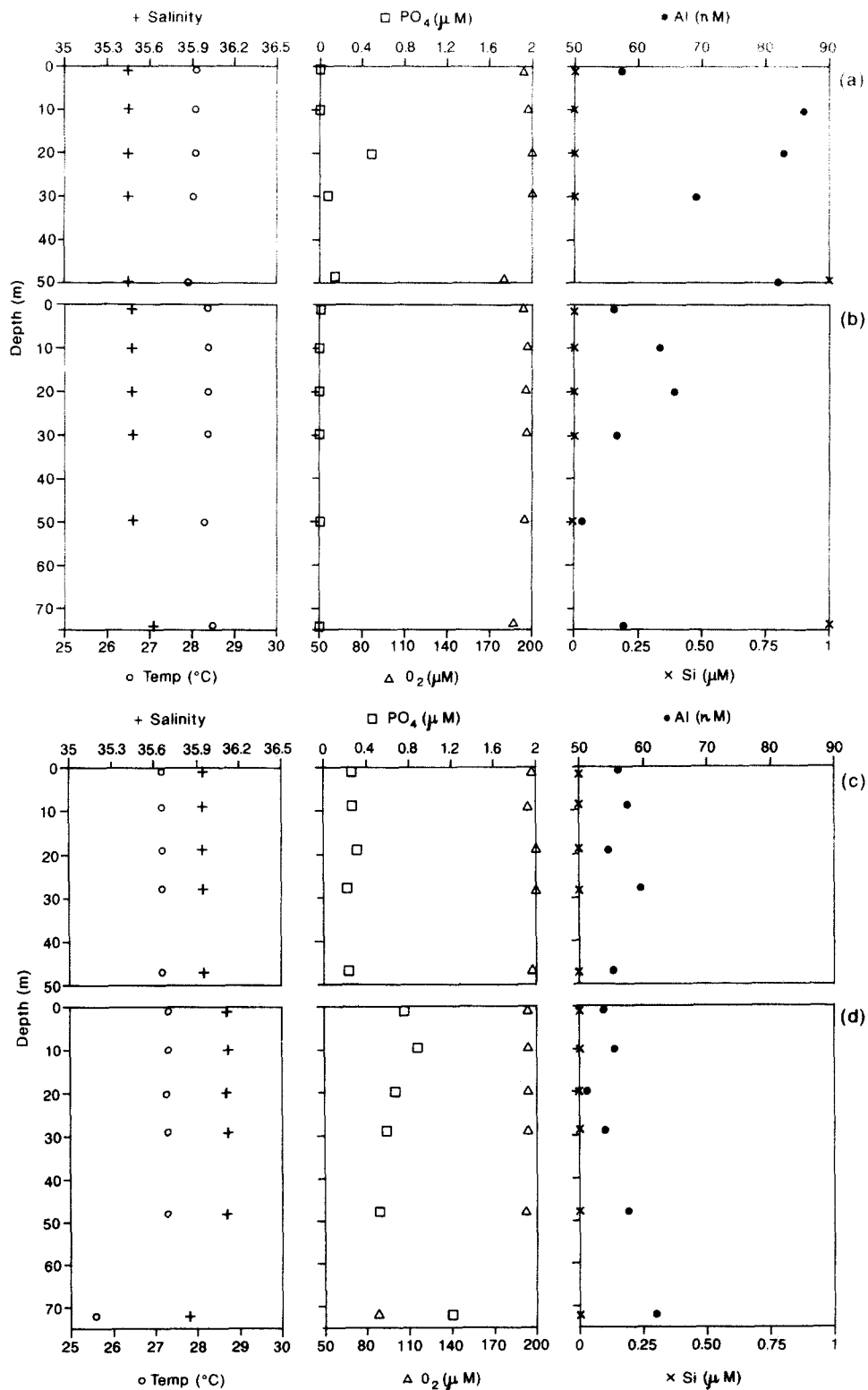


Fig. 3. Vertical profiles of Al and hydrographic data at shallow coastal stations (a) I-1, (b) I-2, (c) K-2, and (d) K-5.

thermocline. Near-bottom samples could not be obtained except at station K-9 (Fig. 4c). Therefore, with only one observation, it is difficult to ascertain the nature of any diffusive flux from the bottom sediments or the probable shape of the bottom profile. At offshore positions the large difference of deep water values ( $\sim 20$ – $\sim 30$  nM; Fig. 4d and e) with similar water mass characteristics is perplexing and there is no obvious explanation for this. A decline in Al levels at depth, in general, suggests that dissolved Al is scavenged in the water column.

The vertical distribution of Al at deep sea stations exhibits an inverse relationship with nutrients. More appropriately, Al distribution does not bear any simple correlation with nutrient elements. Below the upper thermocline, Si concentrations continuously increase with depth whereas Al is either invariant or decreases with depth. Although the influence of regeneration/dissolution of Al may be seen at certain places, scavenging seems to dominate the remineralization and/or regeneration processes and, thus, regulate Al distribution in the water column. This implies that inorganic processes rather than biological activities largely control the Al distribution in the Arabian Sea.

The aqueous chemistry of Al is very similar to that of Th.  $^{232}\text{Th}$  is highly particle-reactive (Huh and Bacon, 1985). Particle-reactive metals are scavenged more efficiently by biogenic particles with  $-\text{OH}$  and  $-\text{COOH}$  groups due to surface complexation (Balistrieri et al., 1981). The estimated oceanic residence time of Al is short ( $< 200$  yr) indicating that dissolved Al, probably in the forms of  $\text{Al}(\text{OH})_3$  and  $\text{Al}(\text{OH})_4^-$ , is extremely particle-reactive in the ocean (Orians and Bruland, 1985).

The Arabian Sea is a highly productive zone due to the continuous supply of nutrients by rivers and the monsoonal upwelling. Apart from surficial biological production, the advective diffusion from the coastal waters supplies a stream of biogenic particles to the deep Arabian Sea, a site of intense nitrate reduction (Naqvi, 1987). The rain of the biogenic particles through the water column induces particle–particle and particle–dissolved species interactions. As suggested by Hydes

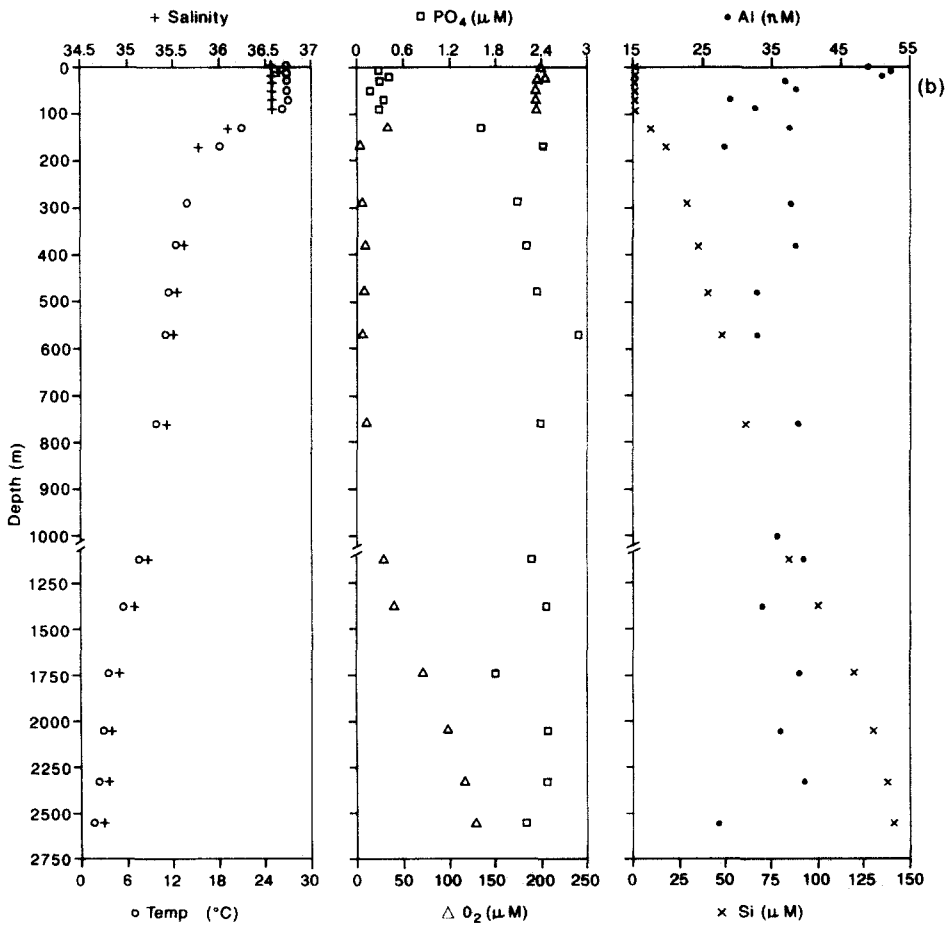
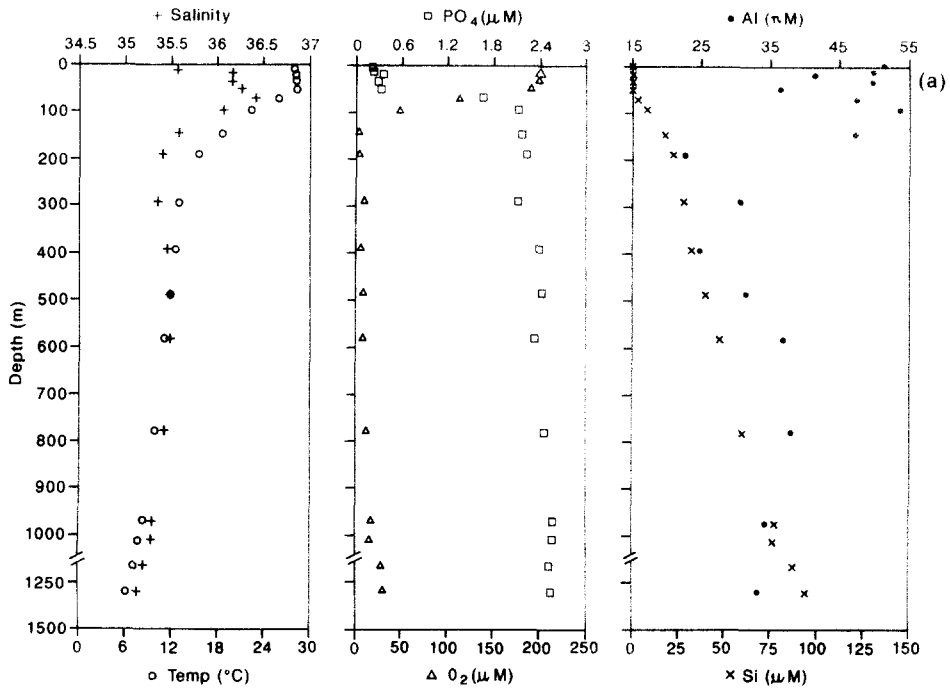
(1979) and later supported by Orians and Bruland (1986), these biogenic particles can provide active surface adsorption sites for dissolved Al, thus removing it from solution. Hence, depletion of Al in subsurface and deep waters is probably a result of scavenging by biogenic particles.

Dead siliceous shells have been found to contain a significant fraction of Al (Martin and Knauer, 1973). Aluminosilicate phases have also been reported from dead diatom frustules (Hurd, 1973). In addition, authigenic minerals and aluminosilicate clays constitute a significant fraction of deep sea sediments (Elderfield, 1976; Hein et al., 1979). In the light of these observations, it is evident that an irreversible adsorption mechanism of Al onto hard silica shells of degraded diatoms and radiolaria (Hydes, 1979) and also the formation of authigenic aluminosilicate minerals might exert a control on the oceanic mass balance of Al.

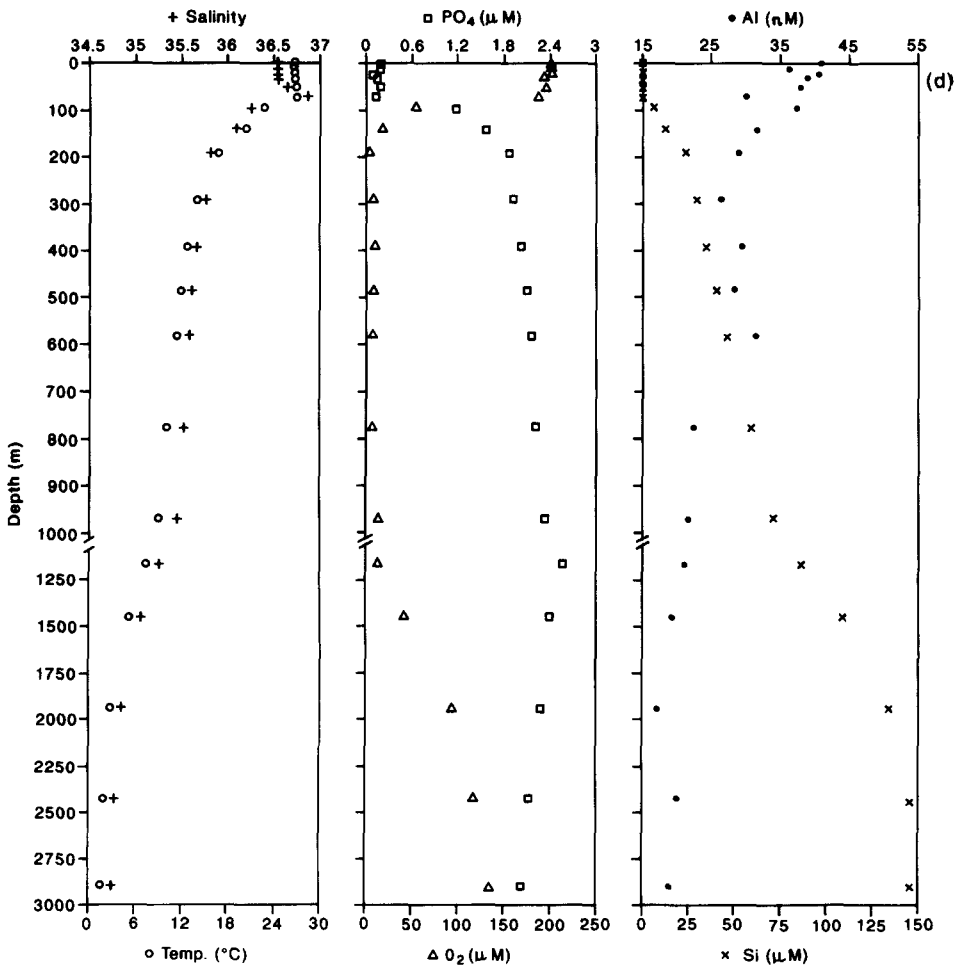
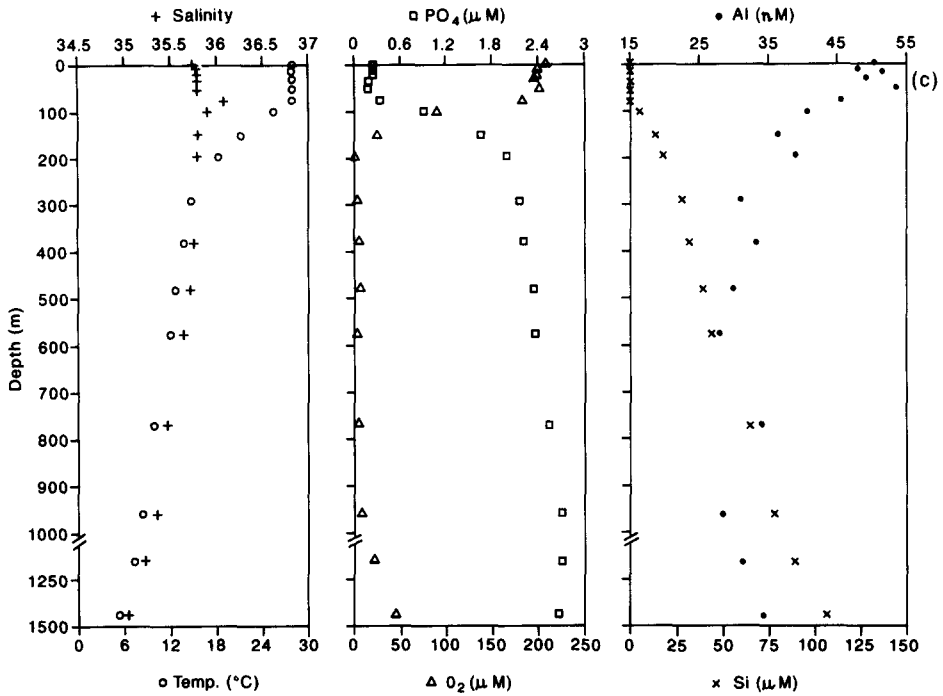
#### 3.1.4. Budget and residence time of Al in the Arabian Sea

The net flux of water from the three major rivers (Indus, Narmada and Tapti) flowing into the Arabian Sea (Fig. 1) is reported to be  $0.266 \times 10^{12} \text{ m}^3 \text{ yr}^{-1}$  (Subramanian, 1979). The influx from minor rivers is not included in the present computations due to lack of data and their probably negligible contribution. Using the lowest value of  $5 \mu\text{g/l}$  in an average riverine concentration range of  $5$ – $10 \mu\text{g/l}$  Al [the lowest estimate based on the results of the Mandovi river (Upadhyay, unpubl. data); Al data from the major rivers being not available], which is a range in the lower side of the average Al concentration range of  $10$ – $80 \mu\text{g/l}$  for the world rivers (Stoffyn and Mackenzie, 1982); Al input to the Arabian Sea is calculated to be in the range  $\sim 1.33$ – $2.66 \times 10^9 \text{ g yr}^{-1}$ .

An area of  $2.55 \times 10^6 \text{ km}^2$  between  $10$ – $25^\circ\text{N}$  and  $52$ – $77^\circ\text{E}$  (Fig. 1) with an average depth of  $2.8 \text{ km}$  is considered here for budget calculations. It is assumed that Al in other parts of the Arabian Sea does not deviate drastically from the presently observed concentrations. With an average seawater Al concentration of  $0.8 \mu\text{g/l}$  ( $30 \text{ nM}$ ) and the volume of the Arabian Sea being  $7.14 \times 10^6 \text{ km}^3$ , the total Al content in the Arabian Sea north







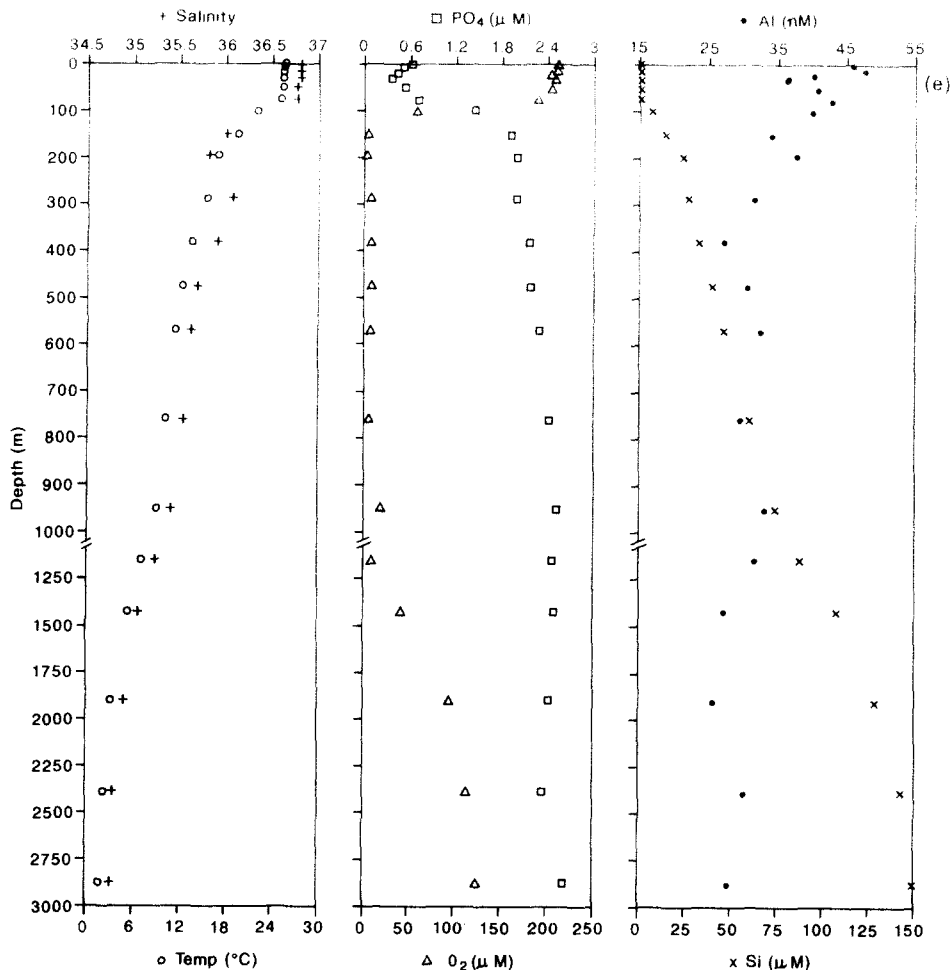


Fig. 4. Vertical profiles of Al and hydrographic data at deep offshore stations (a) I-12, (b) I-14, (c) K-9, (d) K-14, and (e) K-16.

of  $10^{\circ}\text{N}$  is  $5.71 \times 10^{12}$  g. This estimate implies that the residence time of Al in the Arabian Sea (within the geographical region considered) with respect to stream supply is 2100–4300 yr. However, with the inclusion of input of Al from other rivers which could not be considered for the reasons cited above, a shorter residence time could be expected. This value is in good agreement with the residence time range of 470–3400 yr estimated for the world ocean with respect to input from world rivers (Stoffyn and Mackenzie, 1982).

The above is, however, an overestimate since the addition from atmospheric dust has not been considered in the computation. This is, however, likely to be highly significant in the light of observations

by Maring and Duce (1987) and especially of Chester et al. (1985, 1991) who reported high aerosol Al loads over the northern Arabian Sea.

The average of annual dust ovaccumulation, based on a record of Holocene sediments in the Arabian Sea during the last 8000 yr, amounted to about  $100 \times 10^6 \text{ t yr}^{-1}$ ; this compares well with a transcoastal dust flux of  $115\text{--}215 \times 10^6 \text{ t yr}^{-1}$  derived for 1979 from satellite images (Sirocko and Sarnthein, 1989). The average Al concentration in atmospheric aerosols over the northern Arabian Sea is 8.5% (Chester et al., 1985). Hence, total Al transport to the Arabian Sea through the atmosphere amounts to  $8.5 \times 10^{12} \text{ g yr}^{-1}$ . If 8% of Al in the atmospheric

aerosol is dissolved in seawater (Maring and Duce, 1987), Al input to the Arabian Sea through the atmosphere is calculated to be  $68 \times 10^{10} \text{ g yr}^{-1}$ . This estimate yields a residence time of  $\sim 8.4$  yr for Al in the Arabian Sea with respect to combined input of Al from fluvial and atmospheric sources. This is consistent with the short oceanic residence time of Al (<200 yr) found in most oceanic regions (Brewer, 1975; Bruland, 1983; Orians and Bruland, 1985, 1986). The above figures are just first approximations. The short residence time indicates the high reactivity of Al during biogeochemical interactions in the Arabian Sea.

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