DISTRIBUTION OF MERCURY IN A TROPICAL ESTUARY (INDIA) SITUATED NEAR A CHLORO-ALKALI PLANT

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ABSTRACT: The distribution of mercury in water, sediment and some biological samples of the Rushikulya estuary, east coast of India were assessed during Jan-Dec. 1989. Both the dissolved plus acid leachable mercury contents in water and the sediment mercury discerned conspicuous spatial and seasonal fluctuations. Adsorption on to the suspended particulates was found to be the most likely mechanism for removal of mercury from the water column. Exchange of mercury from sediments to water was observed at high salinities (20-30x10⁻³). The residual mercury contents in the biological samples revealed that bio-accumulation by bottom-dwelling organisms are higher than the pelagic components.

KEY WORDS: Mercury contamination - water - sediment - biotic samples.

INTRODUCTION

Although mercury, occurs as one of the rare elements of the earth's crust, its wide range of industrial applications over the past 50 years have contaminated almost everything everywhere. Entry of mercury into aquatic ecosystems takes place in many ways. However, industrial outfalls account for half of the total Hg input into the environment (Goldberg, 1970). Grant (1969) contended that 80 different types of industries use Hg either as raw material and/or as catalysts. But the entry of Hg into the environment from the Chloro-alkali plants is the highest (Jernelove and Wallin, 1973; Zingde and Desai, 1981; Powell, 1983; Campbell *et al.*, 1986; Shaw *et al.*, 1988; Mueller *et al.*, 1989). Thus, the environments around a chloro-alkali plant often heavily contaminated with mercury.

The mercury contents of oceanic water ranges from 0.001-0.004 μ g l⁻¹ (Olafsson, 1983) but can increase by an order of magnitude in coastal waters and estuaries which are in receipt of effluents from Hg emitting industries (Baker, 1977). Of late, many areas other than the Minamata Bay have been marked as "hotspots" on account of the enhanced level of Hg in their water and sediments. The present study area is one such "hotspot" along the east coast of India (Shaw *et al.*, 1985).

In India, awareness about Hg pollution came into existence only when Choudhury (1980) pointed out the possible adverse implications of Hg loss from 38 chloro-alkali plants to the tune of 180 t yr⁻¹. Observations of enhanced levels of Hg in water, sediment and commercial biota of the Arabian Sea (Singbal *et al.*, 1978 and Sanzigiri *et al.*, 1979). Bay of Bengal (Qasim *et al.*, 1988; Kureishy *et al.*, 1983), Thane creek (Zingde and Desai, 1981) and Rushikulya estuary (Shaw *et al.*, 1988) have strongly substantiated Choudhury's contention. Additional confirmation has been provided by more recent literatures on this subject (Sasmal *et al.*, 1987; Patel and Chandy, 1988; Daniel, 1990; Krishnakumar and Pillai, 1990). The Pollution Prevention and Control Boards of various states and the Central Governments have expressed their concern over the Hg pollution issue of the Thane creek and Rushikulya estuary and have suggested periodical monitoring and environmental impact assessments to ensure

their sustainable uses. The present paper describes and discusses the Hg distribution in Rushikulya estuary assessed during 1989.

MATERIALS AND METHODS

The study area, Rushikulya, is one of the minor peninsular rivers that joins the Bay of Bengal near Ganjam (Orissa), east coast of India. On an average, it debouches about 1700x10³ km³ of silt borne freshwater per year (personal communication). Most part of its catchment basin (9000 km²) is occupied by farm lands. Before merging with the sea, the river forms a shallow (maximum depth 3.5m) tidal estuary (Fig.1) which is influenced by semi-diurnal tides. The water quality and biotic communities of the estuary undergo well pronounced diurnal, spatial and seasonal variations due to tidal change and monsoon cycle (Gouda and Panigrahy, 1989; 1992). The estuary is widely known for its rich fisheries and as a suitable ground for prawn seed collection. A chloro-alkali plant producing caustic soda by the age old Hg-cell method, discharges its Hg-rich alkaline effluents directly into the estuary (Fig.1). The loss of Hg from this plant has been estimated at 1030 kg yr⁻¹ (Sahu and Panda 1988) of which most parts of it enters into the estuary. In fact, increased levels of Hg in water, sediments, flora, and fauna of this region have been reported by Shaw et al., (1985, 1988), and Sahu and Panda (1988). Most of these observations have been made on the basis of samplings by periodic surveys and does not speak much about the behaviour or mechanisms of dispersion. In recognition of this, the present work was undertaken as a part of our multidimensional reserach programme relating to the ecology of the estuary.



Fig. 1. Map of Rushikulya estuary showing sampling stations 1-5 and E.

SAMPLING AND ANALYSIS

The study was made during Jan-Dec. 1989. Water and sediment samples were collected at fortnight interval from five stations (1-5) in the estuary and one station (E) in the effluent channel (Fig.1). Surface water samples were collected by immersing 250 ml BOD bottles (corning) to a depth of about 0.35 m, while bottom water samples were collected with a Mayer's shallow water sampler. Each sample (250 ml) was treated with 1 ml of conc. HNO₃ immediately after the collection. Sediment samples were collected using Peterson's Grab sampler and were transferred into polyethylene bags. Biological samples viz., fish, prawn, crab and bivalve molluscs, common in this estuary, were collected from the local fishermen. The test organisms were brought into the laboratory in ice-flasks.

Mercury levels in water, sediment, and animal tissues were determined using a Mercury Analyser MA 5800D, in conjunction with a Cold Vapour Atomic Absorption Spectrophotometer (ECIL India), after suitable digestion. A 100 ml of water sample was digested over a waterbath, after being treated with 5 ml of conc. H₂SO₄, 3 ml of HNO3, 15 ml of 5% (w/v) KMnO4 and 8 ml of 5% (w/v) K2S2O8. Residual oxidants were neutralised by addng 12% (w/v) hydroxylamine hydrochloride solution at room temperature and the aliquot was then used for Hg determination. Three analyses of the same sample gave a standard deviation of 0.981 μ g l⁻¹ at Hg levels of 0.266 μ g l⁻¹ and 1.524 μ g l⁻¹ at Hg levels of 13.30 μ g l⁻¹. For sediment Hg determination, 1 g of air dried sediment was first digested with 5 ml of aqua regia and 5 ml of double distilled water. Then it was redigested with same chemical reagents used for water samples. Replicate digestion and analysis of the same sample yielded a standard deviation of 0.008 μ g g⁻¹ at Hg level of 0.051 and 1.284 μ g g⁻¹ at Hg level of 4.963 $\mu g g^{-1}$. Test organisms were dissected in normal saline, soaked with blotting paper and 1 g of the body tissue was digested with 15 ml of acid mixture (H₂SO₄:HNO₃=2:1). After complete dissolution of the tissues, the aliquote was treated with 20 ml of 5% (w/v) KMnO4 and 10 ml of 5% (w/v) of K2S2O8 and redigested at 80°C. Excess oxidants were neutralised as above, the aliquot was made to a specific volume (100 ml) and then used for Hg determination. Three analyses were made for each tissue and mean was taken as the total residual Hg present in the respective tissues.

RESULTS AND DISCUSSION

HYDROGRAPHY

Our findings on hydrographic features *viz.*, temperature, salinity, dissolved oxygen, pH and transparency have already been published elsewhere (Gouda and Panigrahy, 1991, 1993). Each of these hydrographic events showed conspicuous spatio-temporal variations. Salinity conditions ranged from 0.1×10^{-3} to 35.7×10^{-3} at the surface and $0.3^{-3} \times 10^{-3}$ to 35.7×10^{-3} at the bottom. The estuary remained alkaline (pH=7.7 - 9.1) throughout the study period. Like any other tropical estuary the temperature gradient was between 23.5 and 32.3° C. Monsono cycle and tidal fluctuations emerged as the key factors governing the hydrography of this estuary.

MERCURY IN WATER

Since water samples were treated with conc. HNO3 immediately after their collection, the Hg contents represent both soluble plus acid leachable fractions. In the effluent channel water, the concentration of total Hg ranged from 17.3 - 672.4 µg 1^{-1} yielding an annual mean value of 192.4 \pm 281.1 µg 1^{-1} . Higher values were observed during February-April. The lower values of the rainy season could be ascribed to dilution by the ingress of surface run off into the open effluent channel. The Central Board for Prevention and Control of Water Pollution in India has fixed 0.01 mg l^{-1} (10 µg l^{-1}) of Hg in effluent waters of the chloro-alkali plants as the maximum permissible limit. Thus Hg contents in the effluent water draining into the estuary are much above this limit. But the present values are less than those reported by Shaw et al., (1988) for the same location. During their investigation, from March 1984 - April 1985, they encountered a concentration gradient of 20 - 1548.7 μ g l⁻¹. According to Mishra (1984), the plant discharges 50000 gallons $(2275 \times 10^3 \text{ l})$ of effluent water per hour into the estuary. Hence the Hg input through effluent water is estimated at 386 kg yr⁻¹, which accounts for about 38% of total Hg loss (1030 kg) from the plant.

Mercury contents in estuarine water showed conspicuous seasonal as well as spatial variations (Table-I). Higher values were observed at Station 4 which is situated close to the effluent fallout point. The surface and bottom water concentrations have ranged from 0.27 - 13.30 μ g l⁻¹ yielding an annual mean value of 6.86 μ g l⁻¹ ± 4.94 and from 0.66 - 9.57 μ g l⁻¹ giving an annual mean value of 5.60 ± 3.63 μ g l⁻¹ respectively. Both, the down stream as well as upstream decrease was encountered from this junction zone. But undetactable levels were never encountered. The concentration gradient in surface water at station 1 was between 0.13 - 4.92 (annual average 1.23 \pm 1.48) µg l⁻¹, while at station 5 it was between 0.12 - 6.78 (annual average 2.66 ± 22.24) µg l⁻¹. In general, higher values were observed during Jan-April, when the effluent water concentrations too were high and the estuary was under high salinity regime. A comparative account of Hg levels in Arabian Sea, Bay of Bengal and some selected estuarine habitats are given in Table-II. The Rushikulya estuary concentrations greatly surpasses the reported concentrations elsewhere eventhough some of them are also contaminated with Hg rich effluents from chloro-alkali plants. The most reasonable explanation is that our results include both dissolved and acid leachable Hg. Rushikulya can be considered as more contaminated than the Thane creek because there the Hg (dissolved plus acid leachable) levels hardly exceeds 0.32 μ g l⁻¹ (Zingde and Desai, 1981).

Distribution of Hg in an estuarine environment is govered by several factors : adsorption on to suspended particulates, chemical precipitations and coagulations, biological uptake etc. However, adsorption onto suspended particulates has been put forward as the dominating mechanisms leading to effective sink of Hg to the bottom. According to Frenet-Robin and Ottmann (1978) release of Hg into the water back from sediments is favoured by increase in salinity. The exchange mechanism is also favoured greatly by resuspension of bed sediments resulting in short term increase of Hg in water column (Linderberg *et al.*, 1975). Both of these contentions sees to be true in Rushikulya estuary owing to the fact that higher Hg concentrations were

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Stns/	Station 1			Stn 2			Stn 3			Stn 4			Stn 5		Station E	
Months	Wa	nter	Sediment	Wa	ter S	ediment	Wate	r Se	diment	Water	Sedir	nent	Water Se	diment	Water S	Sediment
	S	B		S	В		S	B		S	В		S		S	
Jan.	0.66	4.79	0.58	1.86	4.12	3.85	5.45	4.52	2.04	5.45	5.19	3.64	4 4.52	1.00	49.21	206.23
Feb.	0.53	0.53	3 1.67	0.66	4.29	1.00	5.18	4.19	0.99	13.30	9.31	4.9	5 4.12	0.59	651.7 1	860.85
Mar.	1.06	3.72	2 1.87	0.39	0.66	0.85	0.58	0.40	2.08	10.39	8.86	2.20) 1.99	0.94	651.70	2020.45
Apr.	4.92	3.72	2 1.87	3.72	3.72	1.87	1.73	6.92	1.87	9.31	9.57	1.44	4 6.78	0.85	672.40	1260.34
May	3.40	4.79	0.98	3.06	4.92	1.05	4.52	4.65	1.00	5.19	5.19	1.5	7 4.79	1.00	51.87	648.57
Jun.	1.86	4.79	1.00	2.09	1.73	0.98	3.26	1.86	1.44	6.52	5.32	1.8	7 1.76	1.57	31.11	282.24
Jul.	0.66	1.33	3 0.98	1.86	1.86	0.97	4.79	2.39	2.17	13.31	9.31	1.0	5 1.68	0.19	17.29	172.96
Aug.	0.93	3.72	2 0.32	0.66	0.66	0.44	1.33	2.18	0.22	13.30	9.31	0.08	3 4.78	0.05	17.29	207.25
Sep.	0.27	0.53	3 0.25	0.27	0.13	0.77	0.27	0.40	0.25	1.99	0.93	0.8°	7 0.93	0.52	37.24	312.29
Oct.	0.27	0.53	3 0.25	0.66	0.27	0.40	1.57	2.79	0.09	1.28	2.66	0.8	6 0.12	0.49	19.97	148.46
Nov.	0.13	0.93	3 0.25	0.13	0.13	0.75	0.13	0.40	0.13	0.27	0.66	0.7	0 0.13	0.58	60.51	429.36
Dec.	0.13	0.13	3 0.61	0.67	0.39	0.46	1.00	0.24	0.49	2.13	0.93	0.59	9 0.27	0.52	49.21	422.23
Mean	1.24	2.40	5 0.89	1.34	1.91	1:12	2.48	2.59	1.07	6.87	5.60	1.60	5 2.66	0.69	192.46	664.27
S.D.	1.48	1.94	4 0.62	1.17	1.84	0.94	2.03	2.13	0.83	4.94	3.63	1.39	9 2.24	0.41	281.49	669.89

Table I. Mercury contents in water ($\mu g l^{-1}$ and sediments ($\mu g g^{-1}$) dry wt. of the Rushikulya estuary during Jan-Dec.1989 (S=surface; B=bottom).



Fig.2. Graph showing relationship between water mercury and sediment mercury.

Area	Range of concentrations	Sources
Oceanic waters,	0.001-0.004 μg l ⁻¹	Olafsson (1983)
Arabian Sea	$13-407 \text{ ng l}^{-1}$ (0.013-0.407 µg l ⁻¹)	Singbal <i>et al.</i> , (1978)
Bay of Bengal	0.5 ppb $(0.5 \ \mu g \ l^{-1})$	Qasim et al. (1988)
Minamata Sea (water)*	60 ng l^{-1}	Kumagai & Nishimura (1978)
Mersey estuary, U.K.	$5-185 \text{ mg l}^{-1}$	Campbell et al., (1986)
Tagus estuary, Portugal	11.5-67 ng l ⁻¹ (0.0115-0.067 μg l ⁻¹)	Figueres et al., (1985)
Loire estuary, France	8.5-131 ng l^{-1} (0.085-0.131 µg l^{-1})	Figueres et al. (1985)
Thane Creek (Bombay	79-320 ng l ⁻¹	Zingde & Desai (1981)
harbour), India	(0.079-0.320 μg l ⁻¹)	
Ennor estuary	BDL-5600 ng l^{-1} (5.6 µg l^{-1})	Daniel (1990)
Adyar Backwater	BDL-14,200 ng l^{-1} (14.2 µg l^{-1})	Daniel (1990)
Rushikulya estuary	007-0.480 mg l ⁻¹	Shaw et al. (1988)
(Orissa), India.	$(7-480 \ \mu g \ l^{-1})$	
Rushikulya estuary (Orissa), India.	0.123-13.300 μg l ⁻¹	Present study

 Table II. A comparative account of Mercury distribution in open ocean and some estuaries.

* Dissolved inorganic mercury only.

reported during periods of high salinity, when the surficial sediments were in suspension, as a consequence of tidal incursion. The correlationships between water and sediment Hg contents (Fig.2) endorses this contention.

Correlation and regression analysis made between the salinity and Hg depicted highly variable pictures (Table III and Fig.3). Significant positive correlations (P \leq 0.5) have been obtained at stations 1 and 2 as well as between salinities 20-30x10⁻³. Campbell *et al.*, (1986) opined that removal of Hg from suspended matter in the Mersey estuary was favoured greatly at salinities of about 25x10⁻³. Thus, our observations agree with their findings The negative correlations between salinity and Hg at salinities 0- 20x10⁻³ and an opposite situation at salinity 20x10⁻³ also suggest that release of Hg back to water from sediments content occurs with increase of salinity above 20x10⁻³

SEDIMENT MERCURY

The total Hg contents in bed sediments of the effluent channel and estuary are given in Table 1. In effluent channel sediments, it ranged from 148-2020 μ g g⁻¹ yielding an annual mean value of 669±669 μ g g⁻¹. Higher values were obtained during

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Station	Parameter	r	R
1.	Surface salinity vs surface Hg	0.560*	Y= -0.277 - 0.067x
	Bottom salinity vs bottom Hg	0.702**	Y = -4.024 + 0.225x
2.	Surface salinity vs surface Hg	0.542	N.S.
	Bottom salinty vs bottom Hg	0.669*	Y= -3.241 + 0.189x
3.	Surface salinity vs surface Hg	0.133	N.S.
	Bottom salinity vs bottom Hg	0.367	N.S.
4.	Surface salinity vs surface Hg	0.004	N.S.
	Bottom salinity vs bottom Hg	0.166	N.S.
5.	Surface salinity vs surface Hg	0.373	N.S.

Table III. Correlation coefficient (r) and corresponding regression equations (RE) indicating the relationships between salinity and total mercury contents.

* Significant at 5% level; ** Significant at 1% level



Fig.3. Relationships between salinity and mercury contents of different salinity subranges (A, sal.0-10x10⁻³, B, sal.10-20x10⁻³, C, sal.20-30x10⁻³ and D, sal.30-36x10⁻³).

Area	Range of concentrations	Sources
Rushikulya estuary Orissa.	0.75 - 8.55 ppm (μg g ⁻¹)	Sasmal et al. (1987)
Rushikulya estuary Orissa.	$0.03 - 58.26 \text{ mg kg}^{-1} (\mu \text{g g}^{-1})$	Shaw et al. (1989)
Rushikulya estuary Orissa.	$0.78 - 181.69 \text{ mg kg}^{-1} (\mu \text{g g}^{-1})$	Shaw et al. (1988)
Adyar Backwater Madras	BDL-110 ng g ⁻¹	Daniel (1990)
	$(BDL-1\mu g g^{-1})$	
Minamat Bay, Japan	28 - 713 ppm (μg g ⁻¹)	Fujuki (1973)
Hooghly estuary	1.2 - 1.75 ppm (μg g ⁻¹)	Sasmal et al. (1987)
Thane creek, Bombay	0.17 - 8.21 ppm (μg g ⁻¹)	Zingde & Desai (1981)
Thane creek, Bombay	0.18 - 2.40 μg g ⁻¹	Patel & Chandy (1988)
Tamar estuary (U.K.)	0.83 μg g ^{-1*}	Bryan & Langston (1992)
Mersey estuary (U.K.)	3.01 μg g ⁻¹	Bryan & Langston (1992)
Wyre estuary (U.K.)	1.52 μg g ⁻¹	Bryan & Langston (1992)
Calcasieus Lake, Louisiana	<0.05mg g ⁻¹ (µg g ⁰¹)	Muller et al. (1989)
Tagus estuary, Portugal	0.044 - 42.5 μg g ⁻¹	Figueres et al. (1985)
Plym estuary	18 - 2610 ng g ⁻¹	Milliward & Herbet (1981)
	$(0.18 - 2.61 \mu g g^{-1})$	
Rushikulya estuary Orissa.	0.051 - 4.963 μg g ⁻¹	Present study

 Table IV: A comparative account of mercury distribution in sediment of some estuarine environment.

* Average concentrations.

January-May. The effluent discharged by the plant contains considerable quantities of suspended solids and therefore enhanced levels of Hg in the bed sediments are very much to be expected. Sahu and Panda (1988) in a December 1986 cruise reported 2464 ppm (μ g g⁻¹) of Hg in the same effluent channel.

Sediment Hg contents in the estuary showed well marked spatial as well as sesonal fluctuation. Station 4 is found to be more heavily contaminated $(0.59 - 4.96 \ \mu g \ g^{-1})$, and from there both upstream and seaward decrease was noticed. At station 5 the Hg levels ranged from $0.05 - 1.56 \ \mu g \ g^{-1}$, while at station 1 (mouth) it varied between 0.25 and 1.87 $\ \mu g \ g^{-1}$. Earlier Sasmal *et al.*, (1987)., Shaw *et al.*, (1988, 1989) and Sahu and Panda (1988) found a similar type of Hg distribution in this estuary. Gradual seaward and upstream decrease of Hg have also been reported in other estuaries polluted by Chloro-alkali plant at mid-reaches (Zingde and Desai, 1981; Figueres *et al.*, 1985; Campbell *et al.*, 1986). Comparision of results (Table IV) further suggests that the Rushikulya estuary is more highly contaminated than most other areas receiving effluent discharges from chloro-alkali plants.

MERCURY IN BIOTIC SAMPLES

Residual Hg contents in body tissues of 7 species of fish and 3 species of shell-fish were estimated in order to trace the major routes of mercury transfer through biotic

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Species	No. of specimens examined	Mean length (cm)	Mercury concentration (mg kg ⁻¹ wet wt.)		
Fish					
Gobius giuris Hamilton	5	10.72 <u>+</u> 0.57	1.27 <u>+</u> 0.02		
Liza macrolepis Smith	5	12.30 ± 0.57	0.23 <u>+</u> 0.02		
Mugil cephalus Linnaeus	5	10.70 ± 0.57	0.62 <u>+</u> 0.03		
0	5	13.00 <u>+</u> 0.50	0.48 <u>+</u> 0.02		
	3	18.00 <u>+</u> 0.50	0.27 <u>+</u> 0.01		
Mystus gulio Hamilton	5	11.30 ± 0.27	1.32 ± 0.03		
Sillago sihama Forsskal	5	08.70 ± 0.57	0.98 ± 0.02		
Tachysurus arius Hamilton	6	22.50 ± 1.30	0.17 ± 0.01		
Therapon jarbua Forsskal	8	08.20 ± 0.57	2.14 <u>+</u> 0.03		
Shell Fish					
Donax sp.	. 8	-	1.16 ± 0.05		
Metapenaeus monoceros	8	6.95 ± 0.39	0.52 ± 0.03		
Penaeus monodon	8	7.81 ± 0.53	0.32 ± 0.02		
Scylla serrata Forsskal	5	-	2.02 ± 0.04		

Table	V:	Residual	mercury	contents ir	1 tissues	of	some	fish	and	shell-fish	fauna
	of the Rushikulya estuary.										

components and to ascertain the accumulation efficiency of various organisms in this estuary. Among the fish fauna, *Therapon jarbua* accumulated highest amount (2.14 $\pm 0.03 \text{ mg kg}^{-1}$) of Hg, while least accumulation was observed with *Tachysurus arius* (0.17 $\pm 0.01 \text{ mg kg}^{-1}$). In general, all the species of shellfish and the bottom dwelling fishes showed higher accumulations than the pelagic components (Table V). But on the whole the biota of the Rushikulya estuary seems to have been more contaminated than those of the Bombay harbour area reported by Somayajulu and Ramaswamy (1972) and Tejam and Halder (1975); in Thane creek by Zingde and Desai (1981) and Patel and Chandy (1988) and in the Darwin National Reserve of Russia (Haines *et al.*, 1992), but were less polluted than those of the Calcasieu estuary, Louisiana (Mueller *et al.*, 1989).

Earlier observations have shown that Hg assimilation in fish and shell fish occurred through adsorption via the gills and body surface (Norstrom *et al.*, 1976). Metabolic activities (Norstrom *et al.*, 1976; Smith and Armstrong, 1975), food and feeding behaviour also often significantly govern the mercury bioaccumulation. Quoting Levitan *et al.*, (1974), Mitra (1986) stated that herbivorous species. The low Hg contents in the body tissues of *Mugil cephalus* and *Liza macrolepis* of the present study endorses this contention. Furthermore, all the bottom dwelling organisms showed higher levels of Hg contaminations than did the pelagic ones as had been reported earlier (Mueller *et al.*, 1989; Bryan and Langston, 1992). Hence, mercury transfer through biotic components mainly takes place here through sedimentory food

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web. However, it was not true with respect of *Tachysurus arius* which was a casual migrant into the estuary from the sea. Amogn the pelagic species mercury accumulation was very high in *Therapon jarbua*. Shaw *et al.*, (1985) opined that smaller fishes could assimilate more Hg because of their high metabolic rates. The enhanced level of mercury accumulation in *Therapon jarbua* species could be ascribed to this.

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