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Seasonal variation of Zn, Cu and Pb in the estuarine stretch of West Bengal

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Zn, Cu and Pb levels in the aquatic phase and underlying surface sediment from three stations (viz. Shankarpur, Canning and Bali Islands) of the coastal zone of West Bengal during different seasons in 2002 were recorded. The order of the heavy metal level in the ambient media of the selected stations is Zn> Cu> Pb. Highest concentrations of heavy metals were recorded in the surface water during monsoon, the period characterized by lowest salinity and pH of the ambient aquatic phase. During premonsoon season all the dissolved heavy metals exhibited minimum values. The biologically available heavy metals from surface sediment of the selected stations showed highest values during premonsoon and lowest during monsoon. Significant negative correlations between the concentrations of dissolved heavy metals and biologically available heavy metals from surface sediments elucidates a sharp exchange of selected metals between the aquatic phase and sediment in the study area.

[**Key words:** Heavy metal, coastal zone, correlation]

Introduction

Estuaries are important pathways for transport of dissolved and particulate materials from continents into the oceans. Out of 250×10^{14} g. year^{-1} of material that enter the ocean from the continents, 210×10^{14} g. year^{-1} is transported *via* rivers and estuaries[1]. The wastes of anthropogenic and industrial origin are of complex characters and have considerable percentage of heavy metals. The heavy metals in the brackish water phase generally deposit on the sediment bed or remain in dissolved state in the water column, depending on the nature of chemical species which are influenced by factors like aquatic salinity, pH etc. Under coastal regulation zone (CRZ) the coastal stretch of West Bengal has been demarcated into three categories-CRZ-I, CRZ-II and CRZ-III. A total of 4571.49 sq.km. area has been identified under CRZ of which CRZ-I alone constitutes 4164.84 sq.km., CRZ-II 14.25 sq.km. and CRZ-Ill 392.40 sq.km. The wastes containing appreciable concentration of heavy metals find their way into the coastal water and adjacent estuaries (particularly Hooghly-Matla estuarine complex). The present study consists of the seasonal variation of Zn, Cu and Pb in the aquatic phase and the surface sediments. It also consists the impact of relevant physico-chemical parameters viz. surface water temperature, pH and salinity on the metal levels of the ambient media as these variables have considerable effect on the process of compartmentation of heavy metals in the estuarine system[2, 3, 4, 5, 6].

Materials and Methods

The entire network of the present programme comprised of the monthly sampling of ambient water and the underlying sediment (upper 1 cm) from the intertidal zone of three stations viz. Shankarpur (Stn-I), Canning (Stn-II) and Bali Island (Stn-III) during 2002 during the high tide period. Stn-I is located in the Midnapore (E) district of the coastal West Bengal at the extreme lowermost stretch of the Hooghly estuary. The other two stations, Stn-II and Stn-III are situated in the MatIa estuary (Fig. 1).

Physico-chemical variables like surface water temperature, pH and salinity were analysed simultaneously from the respective stations with thermometer, pH meter and refractometer respectively after required calibration.

Surface water samples were collected using 10-1 Teflon-lined Go-Flo bottles, fitted with Teflon taps and deployed on a rosette or on Kevlar line, with additional surface sampling carried out by hand. Shortly after collection, samples were filtered through Nuclepore filters (0.4 µm pore diameter) and aliquots of the filters were acidified with sub-boiling distilled nitric acid to a pH of about 2 and stored in cleaned

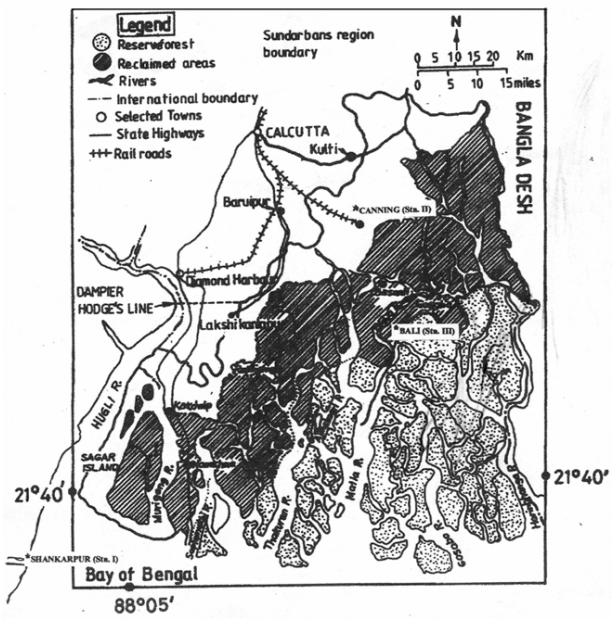

Fig. 1—Map showing the location of sampling stations

low-density polyethylene bottles. Dissolved heavy metals were separated and pre-concentrated from the seawater using dithiocarbamate complexation and subsequent extraction into Freon TF, followed by back extraction into HNO_3[7]. Extracts were analysed for Zn, Cu and Pb by Atomic Absorption Spectrophotometer (Perkin Elmer: Model 3030). The accuracy of the dissolved heavy metal determinations is indicated by good agreement between our values and reported for certified reference seawater materials (CASS 2) (Table la).

Sediment samples from surface (1 cm depth) were collected by scrapping using a pre-cleaned and acid washed plastic scale and immediately kept in pre-cleaned acid washed polythene bags which were sealed. The samples were washed with metal free double distilled water and dried in an oven at 105°C for 5-6 hours, freed from visible shells or shell fragments, ground to powder in a mortar and stored in acid washed polythene bags. Analyses of biologically available metals were done after re-drying the samples, from which 1 gm was taken and digested with 0.5 (N) HCI[8]. The resulting solutions were then stored in polythene containers for analysis. The solutions were finally aspirated in the flame Atomic Absorption Spectrophotometer (Perkin Elmer: Model 3030) for the determination of metal concentrations. No detectable trace metals were found in the reagent blank. Analysis of the NIES Sargasso sample was carried out to assure the quality of the data (Table 1b).

Table la—Analysis of reference material for near shore seawater (CASS 2)

Element	Certified value ($\mu g\ l^{-1}$)	Laboratory results ($\mu g\ l^{-1}$)
Zn	1.97 ± 0.12	2.01 ± 0.14
Cu	0.675 ± 0.039	0.786 ± 0.058
Pb	0.019 ± 0.006	0.029 ± 0.009

Table 1b—Analysis of reference material (NIES Sargasso sample) for sediments obtained from the National Institute of Environmental Studies, Japan

Element	Certified value ($\mu g\ l^{-1}$)	Laboratory results ($\mu g\ l^{-1}$)
Zn	28.6	26.2
Cu	14.9	13.7
Pb	2.4	2.9

The data generated on experimental analyses were subjected to statistical analysis to evaluate the inter-relationship between metal levels in ambient media and selected environmental variables through SYSTAT.

Results and Discussion

Monthly variations of surface water temperature, pH and salinity recorded during the present study are shown in Tables 2, 3 and 4. Similarly the concentrations of dissolved Zn, Cu and Pb and of biologically available Zn, Cu and Pb in the surface sediments have also been recorded (Tables 2, 3 and 4). Surface water temperature in all the sampling stations showed a more or less similar trend of variation with highest value during premonsoon (March-June) and lowest during postmonsoon (November-February). This parameter did not exhibit any significant relationship with the metal concentrations in water and sediment Zn in all the stations. However, with respect to sediment Cu significant positive correlations at 5% level were observed in all the stations and with respect to sediment Pb highly positive correlations were observed in Stn-I & II, but insignificant in case of Stn-III. The salinity and pH of the surface water showed highest values during premonsoon and lowest during monsoon, irrespective of stations and they exaggerated considerable influence on metal levels in surface water and sediments (as evidenced from the r-values).

Table 2— Monthly variation of physico-chemical variables and heavy metal levels at Stn-I

Month	Water temp (°C)	Salinity (%)	pH	Dissolved Zn (µg l^{-1})	Sediment Zn (mg kg^{-1})	Dissolved Cu (µg l^{-1})	Sediment Cu (mg kg^{-1})	Dissolved Pb (µg l^{-1})	Sediment Pb (mg kg^{-1})
January	27.8	21.44	8.32	406.85	37.58	100.66	9.19	16.61	4.60
February	29.9	22.69	8.31	394.66	40.87	98.44	11.45	15.00	5.43
March	30.0	23.37	8.33	355.30	48.79	96.30	13.35	12.07	7.35
April	32.9	26.99	8.33	345.09	52.55	94.75	12.71	18.14	9.81
May	33.4	26.86	8.34	326.90	45.53	90.15	15.29	9.45	8.83
June	34.3	30.04	8.34	309.67	63.05	87.78	18.45	5.60	9.48
July	34.2	21.45	8.32	347.61	38.72	105.58	11.16	13.27	6.63
August	33.7	19.98	8.32	441.70	36.21	207.45	9.22	18.10	5.90
September	33.3	18.22	8.30	497.85	27.54	213.36	9.70	31.14	4.95
October	32.5	19.88	8.31	453.30	29.09	206.30	13.48	19.27	7.79
November	30.9	20.59	8.31	437.40	33.88	200.93	10.93	17.66	6.34
December	27.1	20.99	8.32	411.22	38.83	107.50	9.09	16.00	3.39

Table 3—Monthly variation of physico-chemical variables and heavy metal levels at Stn-II

Month	Water temp. (°C)	Salinity (%)	pH	Dissolved Zn (µg l^{-1})	Sediment Zn (mg kg^{-1})	Dissolved Cu (µg l^{-1})	Sediment Cu (mg kg^{-1})	Dissolved Pb (µg l^{-1})	Sediment Pb (mg kg^{-1})
January	26.9	8.88	8.21	509.71	48.65	147.21	15.28	32.13	6.58
February	29.8	9.06	8.23	488.48	51.90	143.43	17.54	30.53	7.41
March	29.9	14.21	8.30	473.75	59.85	140.56	19.39	27.44	9.33
April	32.6	16.37	8.32	469.50	63.29	137.67	18.80	33.58	11.79
May	33.2	19.75	8.33	481.11	66.28	133.18	21.29	24.89	10.81
June	34.0	24.44	8.33	444.10	8.21	130.69	24.54	21.77	13.40
July	33.9	12.09	8.30	501.79	8.23	148.50	17.49	28.59	8.39
August	33.5	7.64	8.25	605.88	57.7	160.35	15.55	33.40	7.66
September	33.1	6.50	8.13	653.13	42.88	166.28	16.03	36.76	6.71
October	32.0	7.79	8.19	555.44	4.65	159.20	19.81	34.18	9.80
November	30.9	7.80	8.20	538.87	45.44	153.81	17.26	33.23	8.35
December	27.0	8.13	8.21	523.22	47.39	150.41	15.42	31.46	5.40

Table 4—Monthly variation of physico-chemical variables and heavy metal levels at Stn-III

Month	Water temp. (°C)	Salinity (%)	pH	Dissolved Zn (µg l^{-1})	Sediment Zn (mg kg^{-1})	Dissolved Cu (µg l^{-1})	Sediment Cu (mg kg^{-1})	Dissolved Pb (µg l^{-1})	Sediment Pb (mg kg^{-1})
January	27.5	22.97	8.31	241.82	21.76	76.14	6.21	10.21	2.91
February	29.9	23.66	8.32	220.49	25.01	73.18	8.47	8.70	2.74
March	30.0	24.11	8.32	203.70	32.96	70.20	10.32	5.71	4.66
April	32.8	23.57	8.33	201.46	36.40	69.32	9.73	7.80	6.79
May	33.3	27.16	8.33	196.25	29.39	62.44	12.22	7.29	5.81
June	34.1	28.05	8.34	190.06	42.22	46.58	15.47	6.11	8.40
July	34.0	21.54	8.32	233.77	22.57	78.70	8.15	10.26	3.49
August	33.6	17.65	8.31	299.51	20.06	89.94	6.21	12.54	2.66
September	33.1	14.34	8.29	341.90	15.17	96.81	6.69	14.47	2.04
October	32.1	19.85	8.30	287.60	12.94	87.27	10.47	12.80	4.80
November	30.8	21.43	8.31	270.88	17.73	83.43	7.92	11.67	3.35
December	27.0	22.88	8.31	255.15	19.68	80.59	6.08	9.31	4.40

The distribution of dissolved metals followed the order Zn > Cu > Pb in all the three sampling stations (Tables 2, 3 and 4) and exhibited a unique seasonal pattern with highest values during monsoon and lowest during pre-monsoon. This variation may be attributed to huge run-off from the adjacent land masses during the monsoon[9]. However, several studies have pointed out the phenomenon of chemical speciation to be a probable factor behind the high value during the monsoon season[10]. The phenomenon of chemical speciation is governed by a number of factors like distribution, mobility, biological availability of chemical elements (i.e. it's chemical or physical association), pH, redox potential and availability of reactive species such as complexing ligands (organic and inorganic), particle surface for adsorption and colloidal matter. In the present study, significant negative correlations between aquatic pH and dissolved heavy metals confirm the role of pH as one of the major factor influencing chemical speciation of the heavy metals in the present geographical regime. During monsoon, the dilution factor (df) of all the sampling stations in the coastal and estuarine zone of West Bengal increase manifold which results in the decrease of salinity and pH. The lowering of pH might facilitate the dissolution of the precipitated form of metals and increase the amount of metallic ions in solutions[2]. The increase of Cu concentrations with the decrease of salinity was also reported by several studies in identical regimes[11].

The distribution of biologically available heavy metals in sediment followed the order: Zn> Cu> Pb in all the three selected sampling stations and exhibited unique seasonal pattern with high values during premonsoon and lowest during monsoon. In the present study, the concentrations of biologically available heavy metals in sediment differed spatially as per the order Stn-II > Stn-I > Stn-III. Such spatial variations observed during the present study period could be due to the difference in the source of heavy metals, determined by a complex equilibrium governed by various physical, chemical and biological factors[12]. Stn-II, being the site of major anthropogenic activities like fish landing, tourism, fishing vessels and trawlers repairing etc., may generate considerable heavy metals in comparison to Stn-I, where, only fish landing takes place and Stn-III, which is the area near to the Sundarbans Tiger Reserve with minimum anthropogenic disturbances. The heavy metal distribution in the sediments of the world coastal

Table 5—Inter-relation between different physico-chemical variables, dissolved metal and sediment metal concentration at Stn-I

Combination	'r' value	'p' value
Temperature X Dissolved Zn	-0.18971	IS
Salinity X Dissolved Zn	-0.88570	<0.01
pH X Dissolved Zn	-0.89170	<0.01
Temperature X Sediment Zn	0.17652	IS
Salinity X Sediment Zn	0.94405	<0.01
pH X Sediment Zn	0.86146	<0.01
Dissolved Zn X Sediment Zn	-0.88264	<0.01
Temperature X Dissolved Cu	0.23359	IS
Salinity X Dissolved Cu	-0.71210	<0.01
pH X Dissolved Cu	-0.70283	<0.01
Temperature X Sediment Cu	0.47761	<0.05
Salinity X Sediment Cu	0.82508	<0.01
pH X Sediment Cu	0.67923	<0.01
Dissolved Cu X Sediment Cu	-0.38933	IS
Temperature X Dissolved Pb	-0.18960	IS
Salinity X Dissolved Pb	-0.79151	<0.01
pH X Dissolved Pb	-0.81145	<0.01
Temperature X Sediment Pb	0.65279	<0.01
Salinity X Sediment Pb	0.76157	<0.01
pH X Sediment Pb	0.64288	<0.01
Dissolved Pb X Sediment Pb	-0.47166	<0.05

* 'IS' means insignificant

Table 6-Inter-relation between different physico-chemical variables, dissolved metal and sediment metal concentration at Stn-II

Combination	'r'value	'p'value
Temperature X Dissolved Zn	0.11753	IS
Salinity X Dissolved Zn	-0.73828	<0.01
pH X Dissolved Zn	-0.78642	<0.01
Temperature X Sediment Zn	0.36789	IS
Salinity X Sediment Zn	0.88249	<0.01
pH X Sediment Zn	0.89163	<0.01
Dissolved Zn X Sediment Zn	-0.63641	<0.01
Temperature X Dissolved Cu	-0.03437	IS
Salinity X Dissolved Cu	-0.88051	<0.01
pH X Dissolved Cu	-0.85080	<0.01
Temperature X Sediment Cu	0.49332	<0.05
Salinity X Sediment Cu	0.86978	<0.01
pH X Sediment Cu	0.64204	<0.01
Dissolved Cu X Sediment Cu	-0.68895	<0.01
Temperature X Dissolved Pb	-0.19604	IS
Salinity X Dissolved Pb	-0.85035	<0.01
pH X Dissolved Pb	-0.77816	<0.01
Temperature X Sediment Pb	0.60845	<0.01
Salinity X Sediment Pb	0.85758	<0.01
pH X Sediment Pb	0.72281	<0.01
Dissolved Pb X Sediment Pb	-0.58124	<0.01

* 'IS' means insignificant

Table 7—Inter-relation between different physico-chemical variables, dissolved metal and sediment metal concentration at Stn-III

Combination	'r' value	'p' value
Temperature X Dissolved Zn	0.02325	IS
Salinity X Dissolved Zn	-0.94407	<0.01
pH X Dissolved Zn	-0.92463	<0.01
Temperature X Sediment Zn	0.24108	IS
Salinity X Sediment Zn	0.76560	<0.01
pH X Sediment Zn	0.91013	<0.01
Dissolved Zn X Sediment Zn	-0.84610	<0.01
Temperature X Dissolved Cu	-0.12876	IS
Salinity X Dissolved Cu	-0.93870	<0.01
pH X Dissolved Cu	-0.93068	<0.01
Temperature X Sediment Cu	0.47781	<0.05
Salinity X Sediment Cu	0.69108	<0.01
pH X Sediment Cu	0.70832	<0.01
Dissolved Cu X Sediment Cu	-0.81364	<0.01
Temperature X Dissolved Pb	0.08833	IS
Salinity X Dissolved Pb	-0.90683	<0.01
pH X Dissolved Pb	-0.87510	<0.01
Temperature X Sediment Pb	0.30320	IS
Salinity X Sediment Pb	0.73338	<0.01
pH X Sediment Pb	0.76816	<0.01
Dissolved Pb X Sediment Pb	-0.69346	<0.01

* 'IS' means insignificant

regions has been elaborately reported[13-22]. Sediment analysis offer certain advantages over water analysis for the control and detection of metal pollution in estuaries[23,24], although its metal concentrations can also fluctuate over time[25] and the rate of change is well below that of the water[6]. On the other hand, surface sediment often exchanges with suspended materials, thereby affecting the release of metals to the overlying water. Therefore, the top few centimeters of the sediments reflects the continuously changing present-day degree of contamination. Analysis of biologically available sediment is more preferred as it gives a direct impression on the recent anthropogenic pressure. In fact, the total metal concentration in the sediments represents the background heavy metal transported in the lattice structure of weathered crystal materials and has little relationship with the bioaccumulation in the tissue system. In the present study, the increase of metallic ions in the aquatic phase coincided with the decrease of respective metal concentrations in the underlying surface sediment. Estuarine bottom sediments accumulate metals and affect the near bottom water layer due to resuspension or dissolution processes[27].

The significant negative correlations between dissolved metals and biologically available metals from surface sediments in all the three stations (for Stn-I, $\mathbf{r}_{\text{dissolved Zn X sediment Zn}} = -0.88264$, $p<0.01$; $\mathbf{r}_{\text{dissolved Cu X sediment Cu}} = -0.38933$, p = insignificant; $\mathbf{r}_{\text{dissolved Pb X sediment Pb}} = -0.47166$, $p<0.05$, for Stn-II, $\mathbf{r}_{\text{dissolved Zn X sediment Zn}} = -0.63641$, $p<0.0l$; $\mathbf{r}_{\text{dissolved Cu X sediment Cu}} = -0.68895$, $p<0.01$; $\mathbf{r}_{\text{dissolved Pb X sediment Pb}} = -0.58124$, $p<0.01$ and for Stn-III, $r_{\text{dissolved Zn X sediment Zn}} = -0.84610$, $p<0.01$; $r_{\text{dissolved Cu X sediment Cu}} = -0.81364$, $p<0.01$; $r_{\text{dissolved Pb x sediment Pb}} = -0.69346$, $p<0.01$) stand as proof of interchange or exchange process. The significant negative correlations between salinity, pH and dissolved metals (Tables 5, 6 and 7) state the role of aquatic salinity and pH in the process of dissolution. This role is confirmed by the significant positive correlation values between salinity, pH and sediment metals (Tables 5, 6 and 7). Hence, in the present geographical locale, aquatic salinity and pH have a regulatory role in the exchange of heavy metals between the aquatic phase and underlying surface sediment through dissolution-precipitation phenomenon.

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