High cadmium contamination at the gateway to Sundarban ecosystem driven by Kolkata metropolitan sewage in India

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Cr, Pb, Zn, Cd, Ni, Cu, Co and Fe content in the surface sediments was studied at three sites along a 20 km stretch on the Bidyadhari River (situated at the gateway to Indian Sundarban mangrove ecosystem) starting from the sewage outfall zone of the Kolkata metropolis (northern limit of Sundarban) during the dry (March) and wet (August) seasons in the year 2011. The concentration levels of the heavy metals (mg kg^{-1}) were in the following order: Fe (30399 ± 3679) > Cr (68.16 ± 39.28) > Zn (61.33 ± 10.47) > Ni (32.50 ± 4.79) > Cu (27.67 ± 6.59) > Pb (19.69 ± 5.20) > Co (15.23 ± 4.39) > Cd (1.70 ± 0.63). Computation of pollution load index comprising all the heavy metals was found higher during the wet season, mainly due to the enhanced surface run-off led by monsoonal rain. Concentrations of Cr and Co were found higher than the world average in a few cases (especially during wet season); however, computation of pollution indices like contamination factors, enrichment ratio and geo-accumulation index exhibited extremely high Cd contamination in all the sites and during both seasons.

Keywords: Cadmium, contamination, heavy metals, metropolitan sewage, pollution load index.

HEAVY metals like chromium, lead, zinc, cadmium, etc. are among the principal toxic substances which are of concern to the researchers worldwide during the last few decades[1,2]. Sediments lying adjacent to aquatic environments are potential sinks for various types of contaminants[3]. The contaminants of potential concern in sediments typically include trace elements (metals), organochlorine (OC) pesticides, polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs)[4]. Since the riverbed sediments are highly prone to heavy metal pollution, they serve as indicators of pollution load in the aquatic environment[5]. While the degree of heavy metal contamination is region-specific and varies to a great extent throughout the world depending upon both natural and anthropogenic sources, it is difficult to assess the magnitude of contamination simply based on the heavy metal concentration. In order to resolve this problem, several pollution indices have been derived based on the world shale average coined by Martin and Meybeck[6].

Pollution indices like contamination factor (CF), enrichment ratio (ER) and geo-accumulation index (I_{geo}) have been framed to determine the degree of contamination caused by a particular heavy metal, whereas pollution load index (PLI) refers to the extent of total heavy metal pollution from a holistic approach.

Kolkata is one of the most densely populated cities of the world, comprising a population of more than 13 million at present[7]. Rapid and unplanned urbanization, indiscriminate industrialization along with continued growth of the metropolis have made Kolkata one of the most polluted cities in the world[8]. There are several small and large-scale industries in and around the city which produce toxic substances. Some of these industries are chemical and chemical-based, leather and rexins, food and agro products, textile products, tanneries, rubber and plastic products and glass and ceramic products[9]. Most of the effluents released from these factories are untreated and find their way into the Bidyadhari River lock gate, which is the exit point of all the wastes produced due to multifarious industrial and anthropogenic activities[10]. The city sewage falls in the Bidyadhari River by means of different intricate canal systems, mainly by the storm weather flow (SWF) and dry weather flow (DWF) canals. This area is dominated with brick fields and aquaculture ponds, which contribute their waste to the river. Additionally, this region can be denoted as the northern limit of the Sundarban, the largest coastal wetland system of the world. The stretch of the Sundarban ecosystem is situated southwards from this point, marked by the Dampier Hodges line and appearance of the mangrove environment. As the river flows downstream, it bears all the waste towards the interior part of the Sundarban. Henceforth, this locale of the Sundarban can be affirmed as the ecotone between the mixing zone of the polluted sewage-fed riverine ecosystem and the mangrove dominated aquatic ecosystem. The present work was aimed to carry out a comprehensive assessment of heavy metal pollution in the surface sediments, within a 20 km stretch of the Bidyadhari River starting from sewage outfall zone. Another objective of the study was to estimate the seasonal variability of the extent of contamination (between the dry and wet seasons).

In order to conduct the present study three sampling sites were chosen, namely Ghusighata (situated 35 km southeast of Kolkata and 70 km upstream from the shoreline of the Bay of Bengal; 22°31′20.6″N, 88°41′14.8″E), Malancha (22°30′31.6″N, 88°46′27.6″E) and Kanmari Bazar (22°26′32.1″N, 88°48′13.1″E) located ~10 and ~20 km downstream respectively (hereafter referred to as S1, S2 and S3 respectively) (Figure 1). Three equidistant sites situated in the downstream region (one of the sites being the sewage outfall station) were deliberately chosen to monitor the spatial distribution and variability of pollution load throughout the stretch of the riverbed. Moreover, our intention was to observe the extent to which
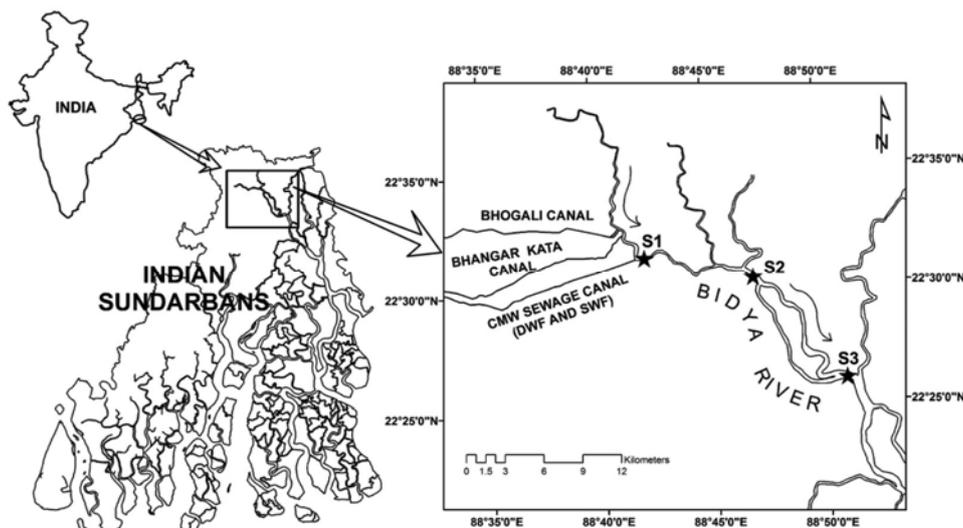
Figure 1. Map of the study area showing the sampling sites starting from Kolkata metropolitan sewage outfall zone to downstream areas along the Bidyadhari River.

gateway to the Indian Sundarban is getting polluted by the released pollution load from Kolkata through this riverine tract. Hence the sampling sites were selected so as to cover the stretch of the ecotone from an extremely polluted sewage outfall zone towards the comparatively less anthropogenically disturbed entrance of the Sundarban mangrove ecosystem. The sediment samples in each site were randomly collected and sampling was done during the dry season (March) and wet season (August) in 2011. Ten samples were collected from the top stratum of sediment in each site and each season during low tide. A composite sample was prepared by mixing four randomly collected sub-samples in case of preparing each set of samples.

Eight heavy metals, namely chromium (Cr), lead (Pb), zinc (Zn), cadmium (Cd), nickel (Ni), copper (Cu), cobalt (Co) and iron (Fe) were analysed from the surface sediments. Each sediment sample was air-dried at room temperature and sieved with <0.65 μm-nylon mesh[11]. The moisture content was removed by pre-treating the samples at $70 \pm 5°C$ until constant weight was achieved[12]. Then 5 g of the well-mixed homogeneous samples was taken in a 250 ml beaker and mixed with 50 ml deionized water and a 10 ml solution comprising aqua regia, hydrofluoric acid and perchloric acid. It was then allowed to digest over a hot plate at a temperature of $100 \pm 5°C$ (ref. 13) until the volume was reduced to around 20 ml. The remnant liquid was removed from heat and the digestion procedure was repeated. The final extract was cooled, diluted to 50 ml and filtered using Whatman No. 42 filter paper and the volume was made up to 100 ml with deionized water. A blank was prepared following the same method. The heavy metals of all samples were determined using an atomic absorption spectrometer (SPECTRAA-50, VARIAN, European Union). The elec-

trical conductivity (EC) and pH of the soil samples were measured on a 1 : 5 soil–water suspension[14]. The organic carbon of the soil samples was analysed using the Walkley–Black method[15].

All the reagents used were of analytical grade (MERCK, Germany) and double-distilled water was used throughout the analysis. Special care was taken to clean, rinse and wash the apparatus required for preparing the samples, standards and stock solutions. Quality control was assured by the use of reagent blanks, duplicates and standard reference material (NIST SRM 2709). The recovery rates for heavy metals in the standard reference material ranged between 87% and 104%. The precision of the analytical procedures, expressed as the relative standard deviation (rsd), ranged from 5% to 10% for different metals. The minimum detection limits and uncertainty in measurements of the heavy metals Cr, Pb, Zn, Cd, Ni, Cu, Co and Fe are 0.05, 0.05, 0.01, 0.01, 0.05, 0.025, 0.05 and 0.05 mg kg^{-1}, and ± 0.058, ± 0.06, ± 0.027, ± 0.007, ± 0.058, ± 0.028, ± 0.058 and ± 0.08% respectively.

Enrichment ratio (ER) was analysed according to the expression[16]

$$\mathrm{ER} = \frac{(C_n/\mathrm{Fe})_{\text{sample}}}{(C_n/\mathrm{Fe})_{\text{background}}},$$

where C_n is the concentration of metal n. The background value denotes the world surface rock average[6]. Fe is considered as the normalization element in order to compute ER, because even a lower proportion of its hydroxide and oxide in an aquatic ecosystem plays a decisive role in controlling the heavy metal distribution[17]. PLI of each site has been calculated using the formula

$$\text{PLI} = (\text{CF}_1 \times \text{CF}_2 \times \cdots \times \text{CF}_n)^{1/n},$$

where CF is the contamination factor of each metal studied[18]. The contamination factor is estimated according to the equation: CF = (Metal concentration in the sediments/Background value of the metal). PLI is calculated in order to assess the overall site-specific pollution impact. I_{geo} is evaluated according to the expression

$$I_{geo} = \log_2 [C_n/1.5B_n],$$

where C_n is the concentration of metal n, and B_n is its geochemical background value[19]. I_{geo} primarily determines the extent of accumulation of heavy metals in the sediments. The background value in the denominator is multiplied by a factor of 1.5 in order to account for the possible variation in background data due to the lithogenic effect. The I_{geo} scale is composed of seven grades (0–6), starting from unpolluted to extremely polluted.

Pearson correlation matrix was computed amongst the eight heavy metals studied ($n = 60$). Principal component analysis (PCA) amongst the heavy metals was performed separately for the dry and wet seasons. Two-factor factorial Analysis of variance (ANOVA) was implemented to determine whether there is any statistically significant difference in heavy metal concentration within the sites as well as seasons and/or both. All statistical analyses were performed using the software SPSS 16.0.

pH of all the sediments was found to be basic in nature, ranging from 7.93 to 8.87. EC varied over a wide range (222–1954 μS cm^{-1}) with a prominent increase towards the downstream due to the increasing tidal influence. The mean organic carbon content was found higher at S3 (1.09) compared to the source point S1 (0.57), which could be attributed to the fact that biological productivity is higher in downstream areas leading to higher accumulation of organic deposits in the estuarine sediments[20].

The mean heavy metal concentration of Cr, Pb, Zn, Cd, Ni, Cu, Co and Fe combining the data of two seasons was found to be 68.16 ± 39.28, 19.69 ± 5.20, 61.33 ± 10.47, 1.70 ± 0.63, 32.50 ± 4.79, 27.67 ± 6.59, 15.23 ± 4.39 and 30399 ± 3679 mg kg^{-1} respectively. The magnitude of different trace metals followed the hierarchy: Fe > Cr > Zn > Ni > Cu > Pb > Co > Cd.

The values of Fe concentrations were found much higher than the other heavy metals, which is in accordance with the fact that the world average for Fe in uncontaminated soils is also as high as 35,900 mg kg^{-1} (ref. 6). It ranged between 18,935.19 and 38,887.78 mg kg^{-1} with a higher mean level during the wet season. Concentrations of Cr were found to vary over a wide range, 20.75–139.30 mg kg^{-1}. The mean concentration of Cr also varied significantly in the two seasons studied. It was as low as 41.55 ± 34.41 mg kg^{-1} during the dry period whereas in the wet season it was found to be quite high (93.16 ± 24.43 mg kg^{-1}). Cr is known for its con-

tamination in tannery effluent and there are several small- and large-scale tanneries situated on the outskirts of the metropolis. The enhanced concentration of Cr in the sediments may be justified by virtue of the surface run-off from the entire catchment area by the rain. Zn was observed to be the least contaminating element in the study area. The mean metal concentration in all the stations was found less than half of the world surface rock average[3]. Zn concentration ranged from 40.92 to 99.49 mg kg^{-1}. No seasonal change was noted in the respective mean values, but comparatively higher levels were detected in the site S1 during the dry season. There was practically no difference in the mean value of Pb concentration in the two seasons studied (19.44 ± 4.33 mg kg^{-1} in the dry season and 19.92 ± 5.88 mg kg^{-1} in the wet season). It was found slightly higher than the average Pb concentration of 14 mg kg^{-1} in the Indian river sediments[21]. Throughout the two seasons, it varied from 12.16 to 50.00 mg kg^{-1}. Ni concentrations varied between 20.86 and 44.1 mg kg^{-1}. Like Pb and Zn, no significant seasonality in the Ni concentrations could be observed (31.71 ± 6.02 mg kg^{-1} in the dry season and 33.34 ± 3.16 mg kg^{-1} in the wet season). Concentration of Cu ranged from 14.30 to 56.6 mg kg^{-1}, while the mean concentration was relatively lower during the dry season (26.03 ± 6.83 mg kg^{-1}) compared to the wet season (29.21 ± 6.04 mg kg^{-1}). The world surface rock average for Co is 11 mg kg^{-1}. The concentration levels of Co in all the studied locations in both the seasons were found uniformly higher than the background value. Co concentration varied between 9 and 24.9 mg kg^{-1}, with a mean concentration of 16.33 ± 5.35 mg kg^{-1} in the wet season and 14.05 ± 2.67 mg kg^{-1} in the dry season. Among all the heavy metals studied, Cd pollution was of highest order. Cd is reported to have no use in any physiological process, but when present beyond a threshold it is known for its toxic and cumulative behaviour, which in turn poses a threat to natural systems[22]. During the wet season its mean concentration was 1.97 ± 0.68 mg kg^{-1}, which is almost 6.5 times more than the world surface rock average[6]. On comparing with the United States Environmental Protection Agency (USEPA) guidelines[23] for sediment quality, Cd concentration was found close to the toxic effect threshold (TET) of 3 mg kg^{-1} but never crossed it, while Cr in few cases was found beyond TET of 100 mg kg^{-1}.

ER (Table 1) and I_{geo} (Table 2) at each station were evaluated in order to assess the extent of metal contamination in the river sediments. The ER values showed depletion trend in case of Cr, Zn and Ni (<1). ER of Pb and Cu was found close to normal, while mild enrichment was detected in case of Co (>1). Cd exhibited alarming ER values (>8), indicating high contamination in the sediments. Its values were found to dissipate in the downstream sites, which signifies the presence of assimilated enrichment from the anthropogenic sources at the outfall zone (S1) and its dissolution downwards. Burning of

Table 1. Enrichment ratio of the eight heavy metals studied in the sites (S1–S3) during dry season (DS) and wet season (WS)

Site	Season	Cr	Pb	Zn	Cd	Ni	Cu	Co	Fe
S1	DS	0.988	1.280	0.650	7.493	0.815	0.990	1.621	1.000
	WS	1.400	1.061	0.551	9.879	0.749	1.028	1.229	1.000
S2	DS	0.363	1.206	0.584	5.455	0.806	1.012	1.574	1.000
	WS	0.969	0.984	0.497	4.126	0.761	1.130	2.092	1.000
S3	DS	0.313	1.149	0.545	4.951	0.795	1.029	1.574	1.000
	WS	0.907	1.328	0.566	8.544	0.785	0.925	1.652	1.000

Table 2. Geo-accumulation index (I_{geo}) of the eight heavy metals studied in the sites (S1–S3) during DS and WS

Site	Season	Cr	Pb	Zn	Cd	Ni	Cu	Co	Fe
S1	DS	−1.025	−0.651	−1.629	1.898	−1.303	−1.021	−0.311	−1.007
	WS	−0.896	−0.346	−1.577	2.339	−1.106	−0.868	−0.032	−0.756
S2	DS	−2.349	−0.617	−1.663	1.561	−1.199	−0.870	−0.232	−0.887
	WS	−0.364	−0.764	−1.709	2.455	−1.265	−0.809	−0.551	−0.849
S3	DS	−2.489	−0.610	−1.687	1.497	−1.143	−0.770	−0.156	−0.811
	WS	−0.722	−0.699	−1.686	1.369	−1.070	−0.500	0.389	−0.676

Table 3. Contamination factor of the eight heavy metals studied in the sites (S1–S3) during DS and WS

Site	Season	Cr	Pb	Zn	Cd	Ni	Cu	Co	Fe
S1	DS	0.737	0.955	0.485	5.591	0.608	0.739	1.209	0.746
	WS	1.166	0.884	0.459	8.227	0.624	0.856	1.024	0.833
S2	DS	0.294	0.978	0.474	4.425	0.654	0.821	1.277	0.811
	WS	0.806	1.180	0.503	7.590	0.697	0.822	1.467	0.888
S3	DS	0.267	0.983	0.466	4.233	0.679	0.880	1.346	0.855
	WS	0.909	0.924	0.466	3.873	0.714	1.061	1.964	0.939

Table 4. Pollution load index in the sites (S1–S3) during DS and WS

Site	DS	WS
S1	0.97	1.07
S2	0.87	1.13
S3	0.88	1.10

fossil fuels like coal and oil along with incineration of municipal waste are known to be a significant source of Cd to the environment[24]. Moreover, various kinds of smelting activities, tannery effluents and metal refining, especially the slag of metal refineries are potent sources of Cd[25,26]. All these types of activities are prevalent among the industrial set-up in and around the Kolkata metropolis, which might have lead to such high ER values for Cd. It is evident from Table 2 that except Cd, the I_{geo} values for all the heavy metals fall in class '0' indicating no pollution (negative I_{geo} values fall under the class '0') from these metals in any of the stations. The I_{geo} values for Cd were almost uniformly found in class '2' during the dry period, whereas in the wet season it was found in class '3' in S1 and S2, which further supports the high contamination of Cd revealed by the ER values.

Table 3 provides CF values of the heavy metals. Since CF is the ratio of original concentration and background value of a specific heavy metal, values higher than one indicate significant contamination. CF values for Cr, Pb and Cu were seldom found above one, whereas CF value for Co was always greater than one. On the contrary, Cd showed extremely higher values of CF in all the instances ranging between 3.873 and 8.227. The evaluated PLI did not show much fluctuation in the sampling stations (Table 4). On the whole, comparatively higher PLI values were noticed during the wet season in the entire study area than in the dry season. Higher PLI values imply higher pollution, which in this case can be attributed to more surface run-off in the wet season due to the effect of monsoonal rainfall. During the dry season, a general decreasing trend in PLI values was observed, specifying the effect of dilution and dispersion of metal content with increasing distance from S1. In the wet season no such trend was visible and highest polluting activity was observed in S2.

The correlation study revealed that most of the heavy metals are positively and highly correlated with each other (Table 5). This type of correlation ($r > 0.9$) may be due to their common source, mutual dependence and/or identical

Table 5. The Pearson correlation matrix between the eight heavy metals studied

	Cr	Pb	Zn	Cd	Ni	Cu	Co	Fe
Cr	1							
Pb	0.161	1						
	0.227							
Zn	0.453**	0.616**	1					
	0.000	0.000						
Cd	0.570**	0.379**	0.433**	1				
	0.000	0.003	0.001					
Ni	0.424**	0.548**	0.847**	0.193	1			
	0.001	0.000	0.000	0.146				
Cu	0.547**	0.301*	0.639**	0.047	0.857**	1		
	0.000	0.021	0.000	0.727	0.000			
Co	0.264*	0.179	0.400**	-0.341**	0.615**	0.670**	1	
	0.045	0.179	0.002	0.009	0.000	0.000		
Fe	0.466**	0.423**	0.691**	0.180	0.840**	0.773**	0.662**	1
	0.000	0.001	0.000	0.176	0.000	0.000	0.000	

$n = 60$, r value and the corresponding P value. **Correlation is significant at the 0.01 level. *Correlation is significant at the 0.05 level.
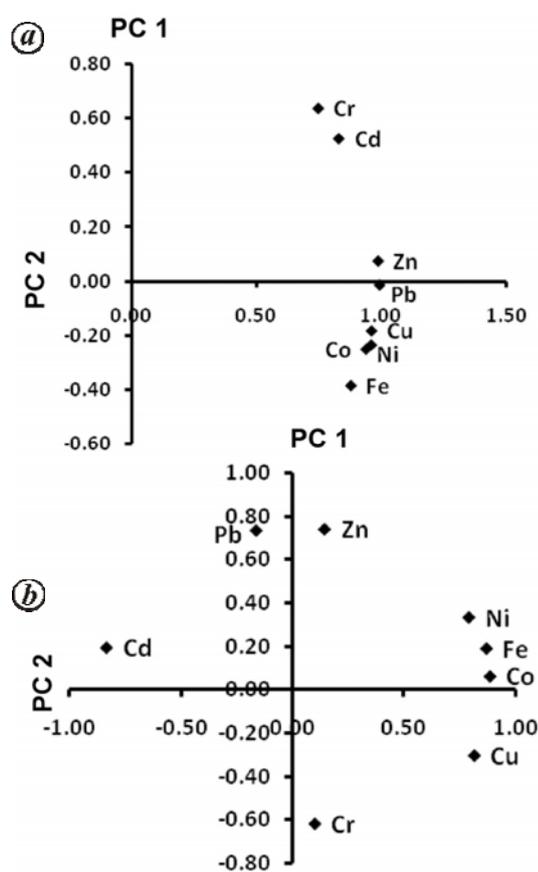
Figure 2. Scatter diagram obtained from principal component analysis during (*a*) dry season and (*b*) wet season.

behaviour during transport, as observed by Jain[27]. Cd was found to be an exception among them, which had correlated with only Cr, Pb and Zn. Fe and Zn were correlated with all other heavy metals (except Cd in case of Fe), which may be due to the fact that Fe in the form of oxyhydroxides has a potential capability to scavenge other heavy metals as they pass through the water en route to the sediments[28]. Most significant correlations were observed between Fe, Cu and Zn, indicating a strong association to geochemical matrix between Ni and these elements in the study area, as observed by Chatterjee *et al.*[29] in another study conducted in the Sundarban ecosystem.

Two PCA were performed with eight heavy metals separately for the dry and wet seasons (Figure 2). In the dry season, component 1 (PC 1) explains 83.67% of the variability and all the components are positively loaded (Figure 2 *a*). In case of the wet season, component 1 (PC 1) and component 2 (PC 2) explain 44.82% and 21.93% of the variability respectively (Figure 2 *b*). In PC 1, Ni, Cu, Co and Fe are highly positively loaded in contrast to negatively loaded variables like Cd. In PC 2 high positive loading was observed in case of Zn and Pb, in contrast to Cr. An association among Co, Cu and Ni was observed in both the scatter diagrams (Figure 2 *a* and *b*); it resulted independent of both season and sampling station. This suggests that these three heavy metals are poured permanently and randomly along the river, as has been observed by Topalián *et al.*[30] while working along the Reconquista River of Buenos Aires, Argentina.

Two-factor factorial ANOVA technique was applied to determine the relative effect of site and season on the concentration of heavy metals. Main effect of site was found to be significant at 1% level for Cr, Cd, Co and Fe and 5% significant level for Ni and Cu. On the contrary, no significant effect of site was found for Pb and Zn. Significant effect of season was found for Cr, Cd, Co and Fe at 1% level of significance. Interaction effect of both site and season was found only for Cd and Co at 1% level of significance.

To sum up, eight different heavy metals were studied in the surface sediments of Bidyadhari River (adjacent to the Kolkata metropolis) situated in the ecotone between the polluted sewage-fed riverine ecosystem and the

mangrove-dominated aquatic ecosystem of the Sundarban. Most of the heavy metals like Pb, Zn, Ni, Cu and Fe exhibited concentrations near the global average values. Cr and Co showed slightly higher concentrations compared to the global average in few instances; however, Cd was found to be present at substantially elevated levels compared to the global average. All the pollution indices computed (i.e. CF, ER and I_{geo}) showed extremely high Cd contamination in the surface sediments of Bidyadhari River, irrespective of site location and season. The presence of various smelters, tanneries and metal refineries along with anthropogenic activities like fossil-fuel combustion and incineration of municipal waste carried out in the industrial sectors of Kolkata metropolis might be responsible for such high Cd contamination. As a consequence, the Cd could get adsorbed and/or absorbed by lower forms of aquatic life and benthic organisms, which in turn might get bio-accumulated in fishes and subsequently lead to deleterious effects upon human consumption.

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