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ORIGINAL ARTICLE

Distribution and ecological risks of heavy metals in Lake Hussain Sagar, India

Arulpoomalai Ayyanar[1] · **Shashidhar Thatikonda**[1,2]

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Abstract Lake contamination by anthropogenic activities has become a serious threat to the aquatic ecosystem due to the presence of a high concentration of toxic heavy metals. In this study, a contaminated Lake Hussain Sagar in India was assessed for toxic heavy metal pollutants at sites associated with industrial discharges and idol immersion activities. The observed high concentration of As, Cd, Pb, Zn, Cu, and Ni in the surface water was due to industrial effluent discharge. About 1.5 times the high concentration of the same elements were observed in surface sediments (0–10 cm) by both industrial discharges and idol immersion activities, compared to deep sediments (0–40 cm) associated only with industrial discharge. The depth-wise variation of heavy metal concentration in surface sediments is due to sorption and settling behavior of suspended solids; whereas in deep sediment, it is due to constant loading of effluents, over a period of time. The sorption capacity of surface sediments is in increasing order as $Pb > Cd \geq Ni \geq Cr \geq Zn \geq Cu$, and the desorption revealed that Pb and Cu were retained to a greater extent due to high clay and organic content. By fractionation study, 20–50% of Zn and 50–80% of Cd were associated with exchangeable and carbonate fraction of sediments. By risk assessment code, Zn and Cd are classified under high risk to a very high-risk category, and Cr, Pb, Ni, and Cu are classified under the medium risk category. The enrichment factor value of sediments for Cd (20.42–119.48), Zn (2.19–4.85), Cu (2.02–3.19), and Pb (2.85–7.72) signifies the significant pollution by anthropogenic activities. Therefore, this study evaluates the intensity and distribution of heavy metals in the lake environment for remediation and restoration of the lake ecosystem.

Keywords Trace metals · Lake sediment · Geochemistry · Speciation · Industrial effluents · Idol immersion

1 Introduction

In India, heavy metal pollution of the aquatic system has become a common and rapidly increasing problem due to anthropogenic and industrial activities. Some of these heavy metals such as Pb, Cd, Cr, Ni, Hg, Zn, Cu, and As are classified under toxic hazardous substance by the national recommended water quality criteria (US EPA 2016), which poses serious damage to the ecosystem when they are above maximum allowable limits (Huang et al. 2017; Wang et al. 2003). These heavy metals tend to bioaccumulate in the food chain that gradually affects the aquatic and public health (Chandra Sekhar et al. 2004; Malik et al. 2010).

"Hussain Sagar" an urban lake, situated in the center of Hyderabad city, India is a major focus as it had become a sink for industrial effluents, domestic sewerage, and idol immersion (Jain et al. 2010). The industrial effluents are the major source of heavy metal pollutants that contribute to lake contamination, specifically, Zn, Pb, and Cr from metallurgical, paint manufacturing, smelting, and tanning process, Cu from textile industries, anti-fouling agents, copper-based fungicides and pesticides, and Cd from industrial processes, anticorrosive agents, pigments, fabrication processes of

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✉ Shashidhar Thatikonda
shashidhar@iith.ac.in

[1] Department of Civil Engineering, Indian Institute of Technology Hyderabad, Kandi, Telangana 502285, India

[2] Present Address: Department of Civil Engineering, Indian Institute of Technology Hyderabad, Kandi, Telangana 502285, India

nickel–cadmium batteries (Huang et al. 2017). In addition, the idol immersion activities as a result of religious ceremonies contribute to the pollution load to a larger extent (Reddy and Kumar 2001). Most of the idols and religious disposals composed of fine clay, plaster of paris, ornaments, jute, straw, paints, varnishes, and cosmetic items (Vyas et al. 2008; Dixit and Tiwari 2007). Decomposition of these materials causes increased dissolved, suspended, and settleable solids to interact with heavy metals and settle at the surface of sediments and pose a threat to the ecosystem (Giripunje et al. 2014; Amiard et al. 2007). The heavy metal mobility depends on pH, redox potential, organic matter, buffering capacity (Villen-Guzman et al. 2015), particle size (Jain et al. 2004), clay minerals and iron, and manganese oxide content (Covelo et al. 2007).

In this study, the heavy metal concentration of Hussain Sagar lake water with the depth-wise profile of the lake sediment was studied to assess the intensification of heavy metal pollution. The fractionation and risk associated with heavy metals were evaluated to highlight the significance of the area of interest for an immediate cleanup action plan. Furthermore, the adsorption and desorption capacity of heavy metals to sediments were studied to understand physical remediation potential of the stated highly polluted lake environment.

2 Methodology

2.1 Site description

Hyderabad is the fifth largest city in India, and it covers an area of 650 km^2 with a population of nearly 10.86 million. Hussain Sagar lake is an urban lake situated (15°N and 81°E) between the twin cities of Hyderabad and Secunderabad. The lake was excavated and constructed between the period of 1550–1580. The lake water was used as a drinking water source for the city up to 1930. Due to unplanned urbanization and rapid industrial development surrounding the lake, it had become a sink for untreated domestic waste and other industrial effluents. Four inlet streams contribute untreated waste to the lake, namely: Picket Nala, Kukatpally Nala, Banjara Nala, and Bowenpally Nala. Among all inlet streams, Kukatpally Nala majorly contributes untreated effluents from industries (pharmaceuticals, biochemicals, synthetic chemicals, detergents, batteries, electroplating, rubber, alloys, etc.) located in Jeedimetla, Balanagar, and Sananthnagar.

2.2 Sampling locations

Sampling points (Fig. 1 and Table 1) were chosen to understand the spatial distribution of heavy metal concentration in lake sediments. Sampling points at Site 1 and 2 depicts the area where common religious practices take place by the local population. The physical appearance of this site seems to be disagreeable due to disposals of decoratives, plastics, garments, flowers, coconut coir, piths, jute, and other religious worn-outs. Sampling point at Site 3 depicts the area where an inlet stream of Kukatpllay Nala discharges industrial effluents from pharmaceuticals, drugs, synthetic chemicals, rubber, and battery manufacturing units combined with urban runoff. Sampling points at Site 4 and 5, depicts the area where idol immersion activities take in a larger extent during the religious festival such as Ganesh Chaturthi and Durga Pooja.

2.3 Sample collection and preservation

All sampling was done during the dry season, post-monsoon period (November 2017). Surface water and sediment samples were collected using a deck boat. Surface water samples were collected by clean acid-washed polypropylene bottles. The Eijkelkamp sediment core sampler (Type Beeker) was used to collect the sediment samples. The sampler contained an assembly of stainless steel cutting head, a transparent acrylic tube (diameter of 7.5 cm and height of 1 m) operated by a piston that facilitates the collection of the undisturbed core without any loss. Two types of sediment samples were collected namely surface sediment and undisturbed core sediment sample. The surface sediment samples contain three grab samples collected from the top portion (up to 10 cm) from an area of 10 m^2 from each site. The collected samples were brought to the lab with a clean polyethylene zip-lock bag. The undisturbed core samples contain 60 cm of core sediment, closed the ends with plastic caps and brought to the lab and further sectioned to 5 cm slices. The sediment samples were dried at room temperature and sieved with 300 μm sieve. All the samples stored in the refrigerator at 4 °C for further analysis.

3 Materials and methods

All chemicals used in this study were of analytical grade bought from Merck (India). Double deionized water with a conductivity of 0.055 $\mu S\ cm^{-1}$ was used throughout the study. All glassware was soaked in 10% HNO_3 for 24 h and rinsed several times with double deionized water prior to use.

3.1 Soil classification, pH and organic matter

The sediment particle size analysis was done by standard procedure ASTM (D422). The pH of sediment samples was

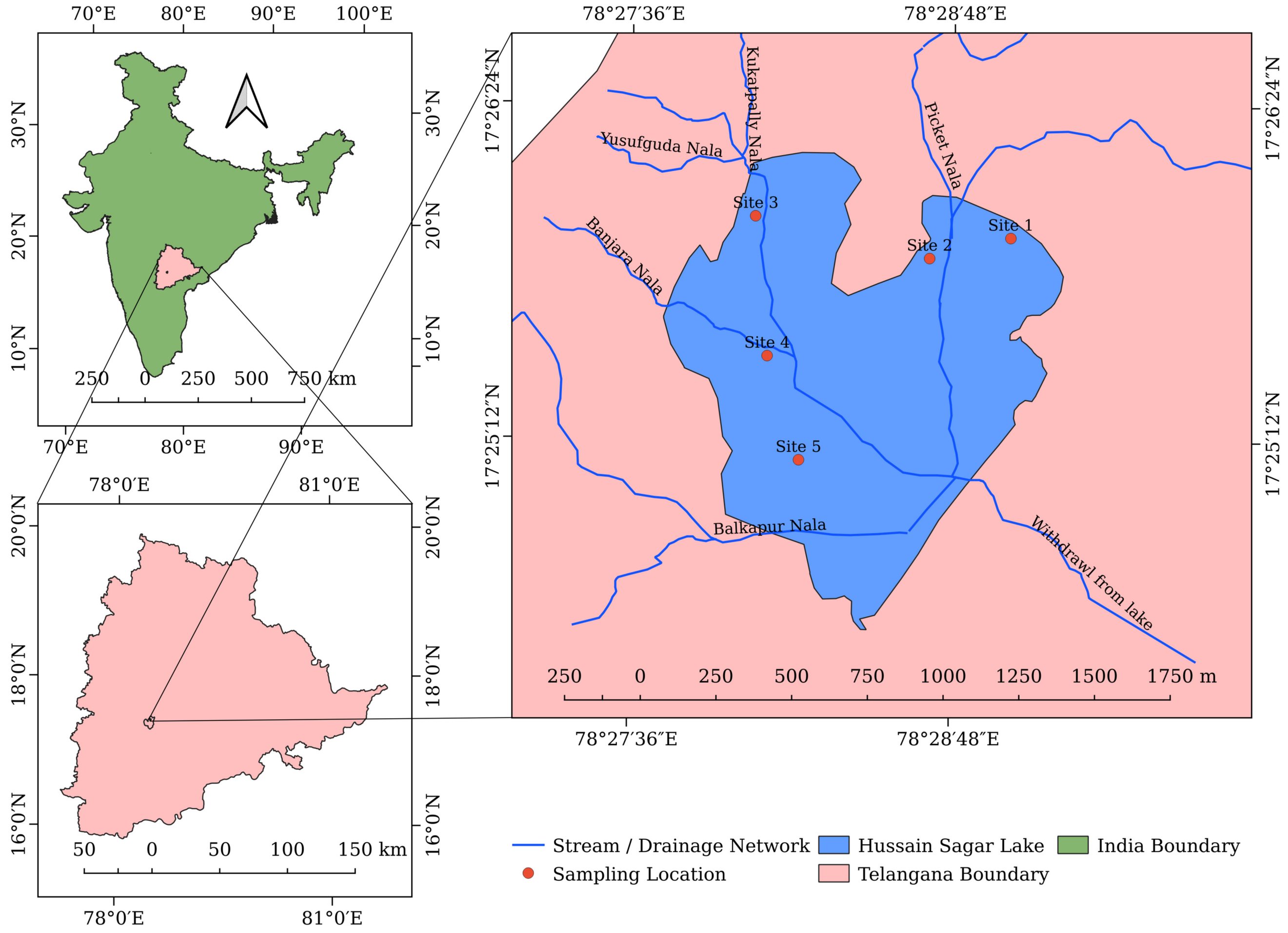

Fig. 1 Sampling locations of Hussain Sagar lake

Table 1 Locations of sampling points

Site no.	Latitude	Longitude
Site 1	17°25′55.5362″N	78°29′0.9165″E
Site 2	17°25′51.1086″N	78°28′42.7572″E
Site 3	17°25′59.8939″N	78°28′3.7221″E
Site 4	17°25′29.9064″N	78°28′6.6182″E
Site 5	17°25′7.6067″N	78°28′13.8812″E

measured by taking 10 g of sediment in a clean polyethylene centrifuge tube with 50 ml of deionized water and shaken vigorously for 5 min. The samples were centrifuged at 10,000 rpm for 10 min (Remi model no.: C-24 plus). Then the supernatant was taken for pH measurement. The water samples pH directly measured by inserting the pH electrode (EPA 9045C). The pH meter used here was a Hanna 3151A with an Ag/Agcl electrode model no.: HI 1131 B. Sediment Organic Matter (OM) was measured by loss of ignition (LOI) method by heating the sediment at 550 °C for 4 h.

3.2 Cation exchange capacity

The Cation Exchange Capacity (CEC) was measured by sum of all exchangeable cations such as Ca, Mg, K, Na, Al, Fe, and Mn (Carter and Edward 2007); 0.5 g of air-dried sediment sample was added with 30 ml of 0.1 M $BaCl_2$ shaken with vortex shaker for 2 h. Then the sediment samples were centrifuged at 10,000 rpm for 10 min. The supernatant was measured for Ca, Mg, K, Na, Al, Fe, and Mn with ICP-MS (Bruker, Aurora M90). The CEC is expressed as (cmol kg^{-1}).

3.3 Buffering capacity

The pH buffering capacity of the soil was determined by using the titration technique (Aitken and Moody 1994). 5 g of sediments were taken into the polyethylene tubes and mixed with up to 25 ml of double deionized water (1:5 ratio). Then the sediment mixture was titrated with 0.1 M HCl to the change of unit pH with continuous mixing. The pH buffering capacity was noted as mmol kg^{-1} pH^{-1}.

3.4 Microwave digestion of sediments

The sediment digestion was followed by EPA (3051A). For microwave digestion, 1 g of 300 µm sieved sediment was added with an acid combination of 1 ml of HF, 1 ml of HNO_3, 3 ml of HCl, and 1 ml of $HClO_4$ in a closed Teflon vial. The vial was heated using microwave digester (MARS 6, CEM) to 150 °C with a ramp of 10 °C per min and held for 20 min. Then, the digested samples were transferred into clean polyethylene tubes and diluted with double deionized water up to 50 ml.

3.5 Heavy metal analysis

The digested samples were centrifuged at 10,000 rpm for 10 min and the supernatant was taken for heavy metal measurement by ICP-MS. The ICP-MS was calibrated with multi-element standard (ESI, scientific, USA) with four-point calibration. All the analyses were done by duplications with five replicates, and average values were reported here. For heavy metals in water samples 2% HNO_3 was added and centrifuged at 10,000 rpm for 10 min to remove the suspended matter and the supernatant was directly introduced to ICP-MS.

3.6 Precision and quality assurance

The Method Detection Limit (MDL) for ICP-MS for heavy metals concentration in liquid samples were determined by the procedure EPA (200.8). The MDL was found by 7 replicates of blank (1% HNO_3) and its standard deviation was multiplied with 3.14. The MDL for various heavy metals were as Cd0.2, Cr0.11, Ni1.3, As0.16, Zn3, Pb10, and Cu1.5 $\mu g \cdot L^{-1}$. Average values of five replicates of standards and samples were taken for determination and its Relative Standard Deviation (RSD) were less than 5%. Quantification of heavy metals was done based on the calibration. After calibration, the instrument was checked with the standards, and the precision was more than 95%. Recoveries and interference effects of heavy metals from the sediments were done by standard addition method according to the procedure by EPA (6020A). During analysis, after each 5 samples, blanks were introduced and checked for the calibration. If the calibration deviated more than 5% recalibration was done and the analysis was continued.

3.7 Metal fractionation study

The metal fractionation was studied by the procedure developed by Tessier et al. (1979). The detailed procedure is given in Table 2.

3.8 Enrichment factor (EF)

The degree of anthropogenic input in sediment can be estimated through enrichment factor (EF) by five categories (Sutherland 2000): (1) EF $<$ 2 Depletion to minimal enrichment suggestive of no or minimal pollution. (2) EF 2–5 Moderate enrichment, suggestive of moderate pollution. (3) EF 5–20 Significant enrichment, suggestive of a significant pollution signal. (4) EF 20–40 very highly enriched, indicating a very strong pollution signal. (5) EF $>$ 40 Extremely enriched, indicating an extreme pollution signal. The EF is calculated as below.

$$EF = \frac{\left(\frac{C_i}{C_{ref}}\right)_{sample}}{\left(\frac{C_i}{C_{ref}}\right)_{backgroud}}$$

where C_i is the concentration of the element of interest, and C_{ref} is the concentration of normalizing element or immobile element (Al/Li/Fe), which has minimum variability in heterogeneous sediment. The selection of normalizing element was based on the field conditions as given in the literature (Brady et al. 2015). In this study, Fe was chosen as normalizing element, and average Fe concentration (27,000 mg kg^{-1}) from the site was taken as a background value.

Table 2 Metal Fractionation study proposed by Tessier et al. (1979)

Step	Fraction	Procedure
1	Exchangeable	1 g of sediment was added with 8 mL of 1 M $MgCl_2$ at pH 7 and agitated continuously for 1 h
2	Bound to carbonates	Residue from step 1 leached with 8 mL of 1 M NaOAc with pH 5 and agitated continuously for 1 h
3	Bound to Fe–Mn oxides	Residue from step 2 extracted with 0.04 M of $NH_2OH \cdot HCl$ in 25% v/v HoAc. The sediment mixture was heated up to 100 °C with occasional agitation for 1 h
4	Bound to organic matter	Residue from step 3 was added with 3 mL of 0.02 M and 5 mL of 30% adjusted the pH to 2 with HNO_3 and the mixture was heated to 85 °C for 2 h with occasional agitation. 3 mL of 30% H_2O_2 added and adjusted pH to 2 with H_2O_2, then the mixture was heated again to 85 °C for 3 h with occasional agitation
5	Residual	Residue from step 4 was digested with a combination of HF-$HClO_4$

3.9 Risk assessment code (RAC)

The risk assessment code (RAC) index proposed by Perin et al. (1985) was used for assessing the adverse effects of toxic heavy metals to aquatic life. The risk assessment code guidelines are given in Table 3.

3.10 Sorption and desorption studies

One gram of acid-washed, well-dried, sieved sediment samples (Site 4) was used to perform sorption experiments. 20 ml multi-metal solution consisting of Pb, Cd, Cu, Ni, Zn, and Cr with varying concentration in the range between 5 and 200 $\mathrm{mg \cdot L^{-1}}$ kept in a 50 ml centrifuge tube was tested against sorbent (1 g of sediment) for all sorption studies. These batch reactions were carried out at pH 7.5 (pH of natural lake environment) over a period of 24 h in a rotary shaker at 160 rpm to achieve sorption equilibrium. After achieving appropriate equilibrium time, the samples were centrifuged at 10,000 rpm for 10 min and separated supernatant was measured for heavy metal concentration. Here, the adsorption capacity is calculated as,

$$q_e = \left(\frac{C_0 - C_e}{M}\right)V$$

where q_e is adsorbed capacity (mg $\mathrm{g^{-1}}$), C_0 is initial concentration of sorbate (mg $\mathrm{L^{-1}}$), C_e is equilibrium concentration of sorbate (mg $\mathrm{L^{-1}}$), M is mass of the adsorbent (g), and V is the volume of sorbate solution. The desorption study was carried out using the sorbed heavy metal sediment samples from the above experiments. 20 mL of heavy metal-free, sodium acetate buffer was used to desorb the sorbed heavy metals from sediment samples. The experimental time and conditions were followed as same as sorption experiments. The retained amount in the sorbent is measured as,

$$q_d = q_e - \left(\frac{C_{de}}{M}V\right)$$

where q_d is retained amount (mg $\mathrm{g^{-1}}$), C_{de} is equilibrium concentration after desorption (mg $\mathrm{L^{-1}}$), M is mass of the residue (g), and V is the volume of sodium acetate solution (L).

Table 3 Risk assessment code (RAC) criteria from Perin et al. (1985)

Risk assessment code (RAC)	Percentage of metal bound in exchangeable and carbonate fraction (%)
No risk	< 1
Low risk	1–10
Medium risk	11–30
High risk	31–50
Very high risk	> 50

4 Results and discussion

4.1 Heavy metals in surface water

Table 4 shows the concentration of heavy metals in different sampling sites. The pH of water samples was found as neutral to slightly alkaline (Table 5). Site 3 showed the highest concentration of heavy metals among all sites, it is due to the untreated industrial discharge entering the lake via Kukatpally Nala. The heavy metals As, Cd, Pb, Zn, Cu, Ni, Mn, and Al were found above permissible limits. Among these Mn, Zn, and Pb were found as 10.73, 3.255, and 8.24 times higher than surface water quality (ISI-IS-2296-1982). Also, Cd was found as 18.055 and 7.66 times higher than criteria continuous concentration (CCC) and criteria maximum concentration (CMC) by US EPA (2016), which causes severe threat to the aquatic life. The increased concentration of Cd is due to the discharge of the effluent from manufacturing industries such as batteries and electroplating. In Site 2, a higher concentration of Cr was found compared with other sites. This was due to the disposal of pigments and cosmetic items as a part of a religious ceremony. Lower concentration of heavy metals observed in Site 4 and Site 5, due to heavy metal adsorbed by suspended particles, were settled in the bottom sediments. All heavy metal concentrations in surface water from Site 1, 2, 4, and 5 were found below the permissible limits.

4.2 Heavy metals in sediments

The sediments found from Site 1 and 2 were characterized as sandy loam, Site 3 as clay loam, and Site 4 and 5 as clay. The pH of all collected sediment samples was found between neutral and slightly alkaline (Table 5). The organic matter (OM) in all sediments were found to be between 4.19 and 11.64%. In Site 4 and 5, OM was found as 10.822 and 11.64% respectively, which is higher than OM in Site 3 (6.75%). Activities involving idol immersion at Site 4 and 5, can be major the reason for high OM as the idol components such as clay, straws, jute, and bamboo contributes to microbial degradation and the formation of humus. Table 6 and Fig. 2 shows the concentration of heavy metals in sediments at various sites. In Site 3, 4, and 5, Cr was found to be more than the Average Shale Background (ASB) (Turekian and Wedepohl 1961) and Effective Range Low (ERL) (Long et al. 1995), which was

Table 4 Heavy metal concentration in Hussain Sagar lake surface water

	Metal concentrations (μg L$^{-1)}$				
	Site 1	Site 2	Site 3	Site 4	Site 5
Al	599.87 ± 11.03	898.15 ± 24.96	**104,787.7 ± 1037.73**	5.12 ± 0.14	51.85 ± 2.31
Cr	9.25 ± 0.18	**26.09 ± 0.1**	5.42 ± 0.16	8.68 ± 18.054	4.50 ± 0.06
Mn	133.34 ± 1.46	305.08 ± 0.42	**5365.19 ± 104.06**	147.03 ± 1.74	139.36 ± 0.85
Co	68.83 ± 1.43	114.30 ± 1.54	**183.49 ± 4.55**	BDL	0.05 ± 0.001
Ni	31.64 ± 0.35	98.96 ± 0.11	**479.91 ± 9.43**	4.42 ± 0.12	4.39 ± 0.08
Cu	165.12 ± 2.13	444.31 ± 2.13	**808.68 ± 11.95**	3.57 ± 0.63	7.46 ± 0.11
Zn	68.55 ± 0.61	154.84 ± 1.06	**4883.63 ± 104.98**	3.57 ± 0.4	7.46 ± 0.19
As	1.06 ± 1.378	1.82 ± 0.02	**26.24 ± 0.67**	4.16 ± 0.061	2.69 ± 0.11
Cd	0.72 ± 1.288	6.33 ± 0.11	**13.79 ± 0.57**	0.38 ± 0.563	0.33 ± 0.01
Pb	6.36 ± 0.1	16.74 ± 0.28	**824.36 ± 43.09**	0.44 ± 0.02	3.70 ± 0.15

Bold values indicates highest concentration of heavy metals found in the surface water

BDL Below Detection Limit

Table 5 Physico-chemical parameters of Hussain Sagar lake Surface water and Sediments

	Parameters	Site				
		Site 1	Site 2	Site 3	Site 4	Site 5
Surface Water	pH	7.24	7.76	7.33	7.48	7.23
	EC (μS cm^{-1})	750	1123	1777	2457	1704
Sediment	pH	7.85	7.76	7.73	7.92	7.23
	OM (%)	4.2	5.95	6.75	10.82	11.64
	CEC (cmol kg^{-1})	6.26	8.33	9.9	12.12	8.94
	BC (mmol kg^{-1} pH^{-1})	11.8	15	42	56	78
	Sand (%)	93	78	39	32	25
	Clay (%)	6	19	59	67	73
	Silt (%)	1	3	2	1	2
	Surface area (m^2 g^{-1})	7.91	8.43	39.15	47.19	51.1

Table 6 Heavy Metal concentration in Hussain Sagar lake Sediments

	Metal concentrations (μg g$^{-1)}$				
	Site 1	Site 2	Site 3	Site 4	Site 5
Al	54,302.03 ± 1379.27	54,670.96 ± 1366.75	70,090.06 ± 890.14	51,601.79 ± 1243.58	**73,859.44 ± 2252.69**
Cr	71.93 ± 0.84	67.24 ± 0.15	**105.83 ± 1.76**	105.16 ± 1.04	102.34 ± 0.53
Mn	274.54 ± 2.43	260.35 ± 2.68	262.06 ± 1.46	**690.02 ± 10.07**	444.96 ± 4.36
Co	8.31 ± 0.44	8.73 ± 0.10	10.54 ± 0.12	12.68 ± 0.06	**15.05 ± 0.19**
Ni	26.59 ± 0.45	37.76 ± 0.38	45.73 ± 0.57	**70.56 ± 0.88**	54.66 ± 0.9
Cu	96.00 ± 0.57	70.98 ± 0.79	94.01 ± 0.89	**137.49 ± 1.26**	52.55 ± 0.88
Zn	369.58 ± 1.80	175.61 ± 1.94	284.21 ± 1.87	**441.94 ± 4.93**	96.72 ± 1.63
As	8.36 ± 0.04	8.77 ± 0.097	13.89 ± 0.091	**20.76 ± 0.23**	9.36 ± 0.10
Cd	5.35 ± 0.36	BDL	19.38 ± 0.14	**34.36 ± 0.24**	BDL
Pb	**134.82 ± 3.96**	62.93 ± 0.811	58.95 ± 1.12	75.92 ± 0.98	50.74 ± 0.47
Fe	23,568.39 ± 992.21	22,741.18 ± 523.04	27,956.78 ± 1101.49	25,881.76 ± 872.18	**34,714.62 ± 968.52**

Bold Values indicates highest concentration of heavy metals found in the sediments

BDL Below Detection Limit

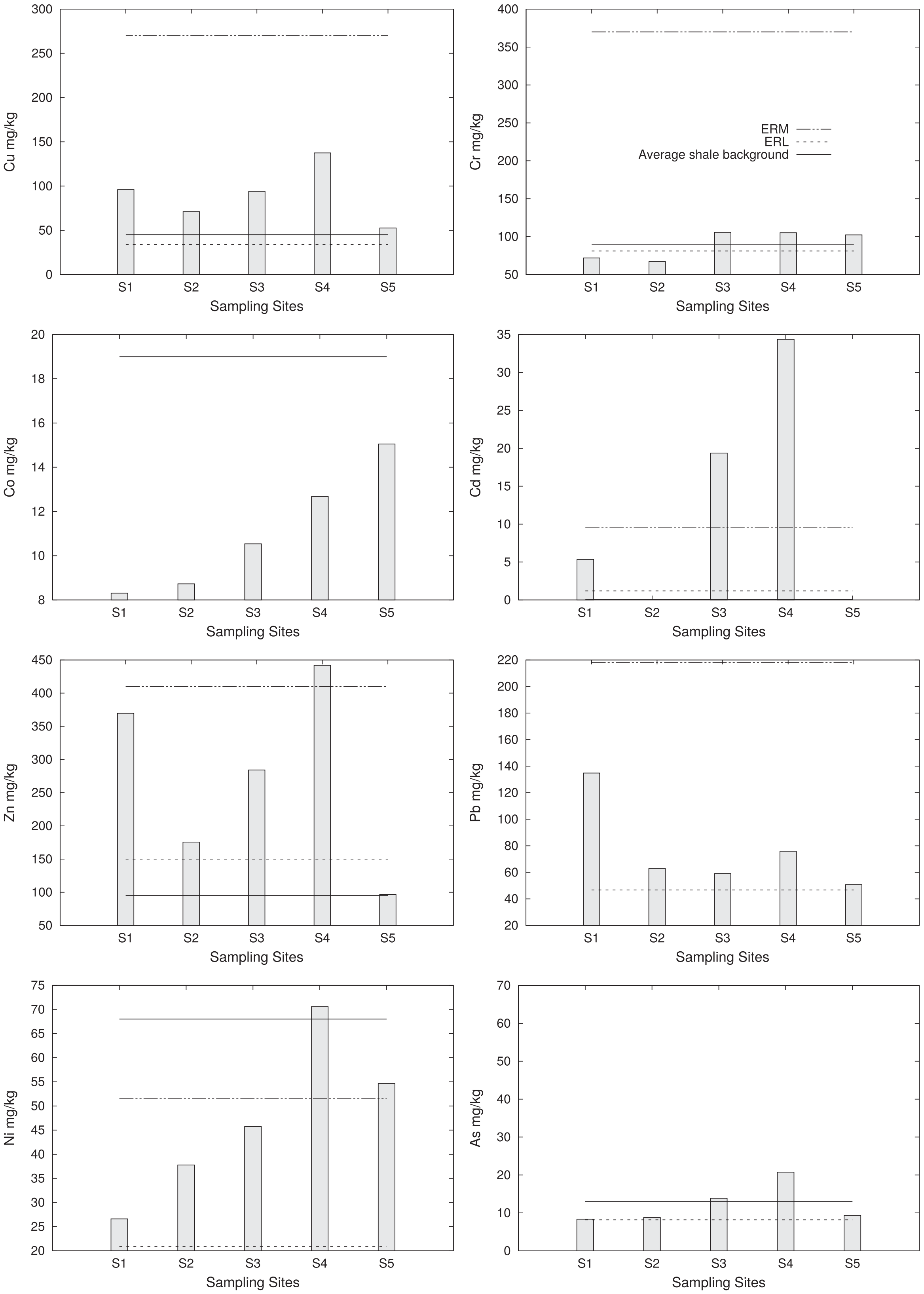

Fig. 2 Concentration of heavy metals in different sites

Table 7 Comparision of Heavy metal concentration in sediments of different lakes and rivers in India and China

Sl. no.	Name of the Lakes	Metal Concentrations (μg g^{-1})						References
		Cd	Cu	Pb	Ni	Zn	Cr	
1	Kolleru lake, India	0.2	205	3.4	0.39	380	40	Chandra Sekhar et al. (2004)
2	Veeranam lake, India	0.81	94.12	30.06	63.61	180.08	88.20	Suresh et al. (2012)
3	Nansi lake, China	0.228	37.0	28.7	39.3	87.5	85.5	Wang et al. (2014)
4	Gomti river, India	2.42	5	40.33	15.17	41.67	8.15	Singh et al. (2005)
5	Yamuna river, India	9.5	22.2	60	–	59.2	–	Jain (2004)
6	Yamuna (Delhi), India	4.50	275	76	159	561	394	Singh (2001)
7	Yamuna (Agra), India	32.50	339	168	101	554	263	Singh (2001)
8	Bortala river, China	0.17	30.09	31.98	22.32	99.19	51.55	Zhang et al. (2016)
9	Mahanadi basin, India	9.6	36	131	55	137	72	Sundaray et al. (2011)
10	Dianchi Lake, China	29.5	201.6	131	–	595.8	–	Gu et al. (2017)
11	Poyang Lake, China	1.08	26.89	37.98	20.46	–	–	Mingbiao et al. (2008)
12	Hussain Sagar lake, India	13.6	132	50.8	164	214	32	Jain et al. (2010)
13	Hussain Sagar lake, India	19.89	90.108	79.885	47.042	273.14	90	Present study

Table 8 Sediment Quality Guidelines (SQGs) based on heavy metal concentration

	Metal concentrations (μg g^{-1})									
	Al	Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb
ERL	–	81		–	20.9	34	150	8.2	1.2	46.7
ERM	–	370		–	51.6	270	410	70	9.6	218
Average Shales Background	80,000	90	850	19	68	45	95	13	0.1	20

ERL (Effect Range Low) guideline values indicate concentrations below which adverse effects on biota are rarely observed (Long et al. 1995)

ERM (Effect Range Median) guideline values indicate concentrations above which adverse effects on biota are frequently observed (Long et al. 1995)

Average Shale Background value from Turekian and Wedepohl (1961)

above 100 μg g^{-1}. Possible sources of Cr include tanning and dying industries' effluents discharge and deterioration of paints from immersed idols. Cu and Zn concentration in all sites were more than the ASB and ERL. Except for Site 5, where the concentration of Zn was found below ERL. In Site 4, Cu (137.49 μg g^{-1}) and Zn (441.94 μg g^{-1}) were found to be higher when compared to other sites, which is more than the ASB and ERL. This is due to the presence of clay fraction (67%) and organic matter (10.822%) which causes binding of certain heavy metals as described in Sect. 4.3. In Site 3 and 4, Arsenic (As) was found to be more than ASB and ERL, with the highest concentration of 20.76 μg g^{-1} noted in Site 4. Cd was found to be 19.38 μg g^{-1} in Site 3 due to the discharge of industrial effluents discharge from batteries, ferrous, and non-ferrous metal manufacturing units. Site 4 shows further higher Cd concentration as 34.36 μg g^{-1}. It is due to the idol immersion activities which increases the suspended particles in water. These suspended particles pose higher surface area which traps the heavy metals and settle in the bottom of sediments. The Cd found in Site 3 and Site 4 are above Effective Range Median (ERM) and higher than ASB comes under severe risk to aquatic life, also evident studies shown in (Jain et al. 2010). The comparison of average heavy metal concentration in other contaminated lakes and rivers in India and China is shown in Table 7. This comparison shows that a higher level of Cd, Zn, Cu, and Pb were found to be significant level of contamination by anthropogenic activities. An earlier study conducted on Hussain Sagar lake sediments by Jain et al. (2010) showed that the heavy metal concentration of Cd, Zn, Pb, and Cr increased from 13.6, 214, 50.8, and 32 mg kg^{-1} to 19.89, 273.14, 79.885, and 90 mg kg^{-1} in the present study (Tables 8 and 9).

4.3 Metal fractionation study

Figure 3 shows the fractionation studies of sediments from different sites. The fractionation study reveals the approximate proportion of heavy metals bound to various fraction such as exchangeable, bound to carbonates, bound to iron, and manganese oxides, bound to organic matter,

Table 9 Water quality criteria

	Metal concentrations (μg $L^{-1)}$									
	Al	Cr	Mn	Co	Ni	Cu	Zn	As	Cd	Pb
Surface water quality standard as per ISI-IS-2296-1982 (Class A)	–	50	500	–	–	1500	1500	50	10	100
Criterion continuous concentration (CCC)	87	–	–	–	52	–	120	150	0.72	2.5
Criteria maximum concentration $(CMC)^2$	750	–	–	–	470	–	120	340	1.8	65

CCC is an estimate of the highest concentration of a material in surface water to which an aquatic community can be exposed indefinitely without resulting in an unacceptable effect

CMC is an estimate of the highest concentration of a material in surface water to which an aquatic community can be exposed briefly without resulting in an unacceptable effect

and residual. These fractions contribute to the mobility of heavy metals in sediments with existing and changing environmental conditions. It was revealed that 60% As and 80% Co were found to be associated with "residual" fraction. The residual fraction is the primary and secondary minerals contained heavy metals in the crystalline lattice, which contribute to the bulk of this fraction (Gleyzes et al. 2002). These heavy metals are strongly bound to sediments and not readily released to the environment under the normally existing conditions in nature (Gao et al. 2012). 50% Cd, 30% Zn, and 10% Cr were found to be associated with "Carbonates". It was also found that certain concentrations of these heavy metals were associated with "Exchangeable" (25% Cd) and "Fe–Mn oxides" (30% Zn and 20% Cr). The stability of these heavy metals associated with carbonates is dependent on pH. Thus, changes in pH can influence the release of elements from sediments. Besides, the heavy metals in exchangeable fractions were weakly bonded or adsorbed onto the sediment particles are influenced by changes in the ionic strength, that facilitates the release of metals from sediments. However, Cr and Zn associated with Fe–Mn oxides that remain stable due to oxides, which acts as scavengers for metal cations, may become unstable at anoxic conditions (low Eh) and readily released to surface water. Among all heavy metals bound to various fractions, Cu has an affinity to bind organic matters in sediments (Gupta et al. 2013; Yu et al. 2001; Jain et al. 2010) which was shown in this study as 80% Cu was found associated with "Organic" fraction. In contrast to Fe–Mn bound heavy metals, the organic bound heavy metals are stable under anoxic conditions but on OM degradation at oxidizing conditions, the heavy metals can be readily released and bioavailable.

4.4 Risk assessment code (RAC) and metal enrichment factor

The risk assessment code explains the eco-toxicity to benthic and aquatic fauna by heavy metals availability with a comparison of bond strength of heavy metals to sediments. The bond strength of heavy metals to the sediments is explained by metal fractionation study (Tessier et al. 1979). It states that heavy metals bound to exchangeable and carbonate fractions are weakly bonded; and heavy metals bound to Fe–Mn oxides, organic and residual fractions are strongly bonded to the sediments. The RAC with the percentage of heavy metals associated with exchangeable and carbonate bound fractions shown in Table 3.

The fractionation of heavy metals in sediments for all sites was shown in Fig. 3. It was found that 50–80% of Cd were associated with exchangeable and carbonate bound fractions. According to RAC code, Cd falls under the very high-risk category. Similar observation was also found in the Hussain Sagar sediments. Major portions of Cd were found in exchangeable or carbonate bound fraction (Jain et al. 2010). The loosely bound Cd poses a severe threat to the micro-organisms (Doelman and Haanstra 1984), aquatic species (Das and Banerjee 1980; Jain 2004), vertebrates, and invertebrates (Russell et al. 1981). Also, Cd poses harmful effects humans including cancer, Itai-itai disease, and other health issues related to bone, liver, lungs, and kidney as a result of long term exposure (Satarug et al. 2010). In addition to Cd, 20–50% of Zn is associated with exchangeable and carbonate bound fraction and classified as high-risk category. Zn can cause acute toxicity to the aquatic species and affect the morphology of fishes (Taylor and Chapman 2011). 3–15% of Cr, 10–24% of Pb, 3–22% of Ni, and 2–16% of Cu associated with exchangeable and carbonate bound fraction and falls under low to medium risk category. The risk of heavy metals are in increasing order of Cd > Zn > Cr > Pb > Ni > Cu > As > Co in all sites.

The degree of contamination due to anthropogenic sources can be estimated by Enrichment Factor (EF). The EF values classified under different categories based on the Average Shale Background are shown in Table 10. The higher EF value (≥ 2) of Pb, Zn, Cu, and Cd implies the pollution level from moderate to extreme pollution (Zhang and Liu 2002). The highest EF of Pb was found to be 7.72 in Site 1, indicating contamination including vehicular emission deposits from nearby roadways. Similar studies

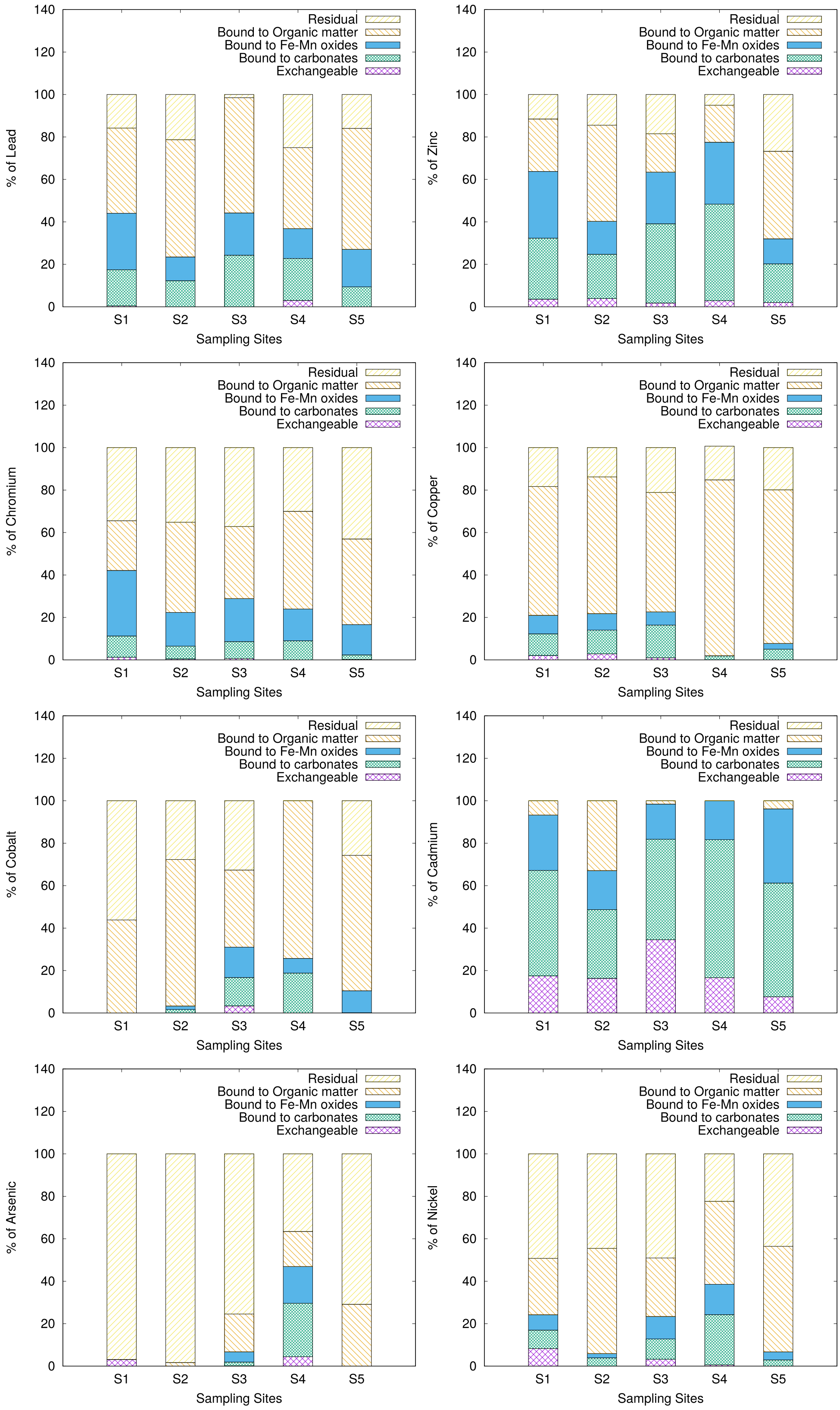

Fig. 3 Fractionation studies of Hussain Sagar sediments

found a significant amount of Pb contamination in lake sediments due to vehicular sources (Yang and Rose 2005; Gupta et al. 2013; Njenga et al. 2009). For Cd, EF in Site 1, 3, and 4 were 20, 60, and 120, respectively, that falls under the extremely polluted category. It is also noted that in the previous study, the reported EF was only 45.07 (Jain et al. 2010) suggesting that Cd level increased to a greater extent due to industrial discharge from the battery, electroplating, pigments, ferrous, and non-ferrous metal manufacturing units. The EF of Zn (0.8–4.85) and Cu (0.91–3.19) showed moderate contamination level. Sources of Zn is from industrial activities such as galvanizing, plating, and urban runoff due to wear and tear of tires (Zn used as vulcanization agent in vehicle tires) and brake lining (Singh et al. 2015; Sutherland 2000). The main contributor of Cu to the lake is from metal plating, antifoulants, brake lining wear, and usage of algicide and fungicide (Sutherland 2000; Duodu et al. 2016).

4.5 Depth wise profile study

Figure 4a, b show the Depth wise profile of heavy metal contamination along the vertical direction of sediment. In Site 3, high concentration of As (25.78 µg g^{-1}), Cr (170 µg g^{-1}), Cd (25.45 µg g^{-1}), Cu (133 µg g^{-1}), Pb (160 µg g^{-1}), Ni (60 µg g^{-1}), and Zn (450 µg g^{-1}) were found in the deep sediments up to 40 cm. This is due to the constant loading of effluents from electroplating, battery manufacturing, chemical, and pharmaceutical industries entering the Site 3 via Kukatpally Nala. In contrast to Site 3 and Site 4 shows the highest concentration of Ni, Cd, Zn, Cu, and Pb as 71.05, 73, 570, 165.66, and 128.8 µg g^{-1} was found in the top layer of sediment up to 10 cm. This could be due to idol immersion activity that allows the suspended particles and other particulate matter to form a fluffy layer to localize the heavy metal concentrations only on the top layer of sediment. Also, some heavy metals were adsorbed to the suspended particles, organic matter, and plaster of paris from idols to settle down at the top layer of the sediments. In certain conditions, the site heavy metal characteristics show the distribution of toxicant is localized in the top surface with fluffy nature when compared to other sites. In order to choose the active clean-up technology, the sites were closely evaluated to understand the nature of sediments to implement any physical treatment process.

Table 10 Enrichment factors (EF) for Hussain Sagar sediments normalised with respect to the Fe content

Metals	Sites				
	Site 1	Site 2	Site 3	Site 4	Site 5
Ni	0.45*	0.66*	0.65*	2[†]	2[†]
Cu	2.44[†]	1.87*	2.02[†]	3.19[†]	0.91*
Zn	4.46[†]	2.19[†]	2.89[†]	4.85[†]	0.79*
As	0.74*	0.80*	1.03*	1.67*	0.56*
Cd	20.42[‡]	0.00*	62.40**	119.48**	0.00*
Pb	7.72[§]	3.74[†]	2.85[†]	3.96[†]	1.97*
Cr	0.10*	0.09*	0.12*	0.13*	0.09*
Co	0.50*	0.55*	0.54*	0.70*	0.62*

*EF < 2; no or minimal pollution

[†] EF 2–5; moderate pollution

[§] EF 5–20; significant pollution

[‡] EF 20–40; very strong pollution

**EF > 40; extreme pollution

4.6 Adsorption and desorption

Figure 5 shows the adsorbed and retained amount of heavy metals against its initial concentration during adsorption and desorption, respectively. The sorption of heavy metals in sediments (Site 4) are increasing order as Pb > Cd ≥ Ni ≥ Cr ≥ Zn ≥ Cu and the retention of heavy metals are as Pb > Ni > Cu ≥ Zn > Cd > Cr. The maximum adsorption capacity (Q_o) and bonding energy (b) were calculated based on the Langmuir equation. The Freundlich parameters KF and n indicates the adsorption capacity and intensity. Values of n are greater than 1 shows that adsorption intensity is good or favorable over the entire range of concentration (Eloussaief and Benzina 2010). Table 11 shows the Langmuir and Freundlich parameters Qo, b, K_F, and n. During the adsorption stage, almost all sorption of heavy metals are well fitted with Langmuir and Freundlich isotherm and adsorption intensity found greater than 1, which is favorable. During the desorption stage, Pb and Cu only fitted with the Langmuir and Freundlich isotherm, other heavy metals such as Cr, Cd, Ni, and Zn were found to have a lesser affinity to the sediments. This can be due to the high amount of clay fraction such as vermiculite, kaolinite (Covelo et al. 2007), and high surface area (Jain and Ram 1997) that retained Pb up to 1.53 mg g^{-1}. Cu was retained up to 0.25 mg g^{-1} due to high organic content (10.82%), and Cu has the tendency to bind with an organic fraction as described in Sect. 4.3 and Covelo et al. (2007). Thus, competitive adsorption and desorption studies imply the sorption phenomena of sediment found in the lake to understand the treatability of localized heavy metals.

5 Conclusions

In this study, the heavy metal concentration in Lake Hussain Sagar is evaluated for ecological risk. The observed concentration of heavy metals such as As, Cd, Pb, Zn, Cu,

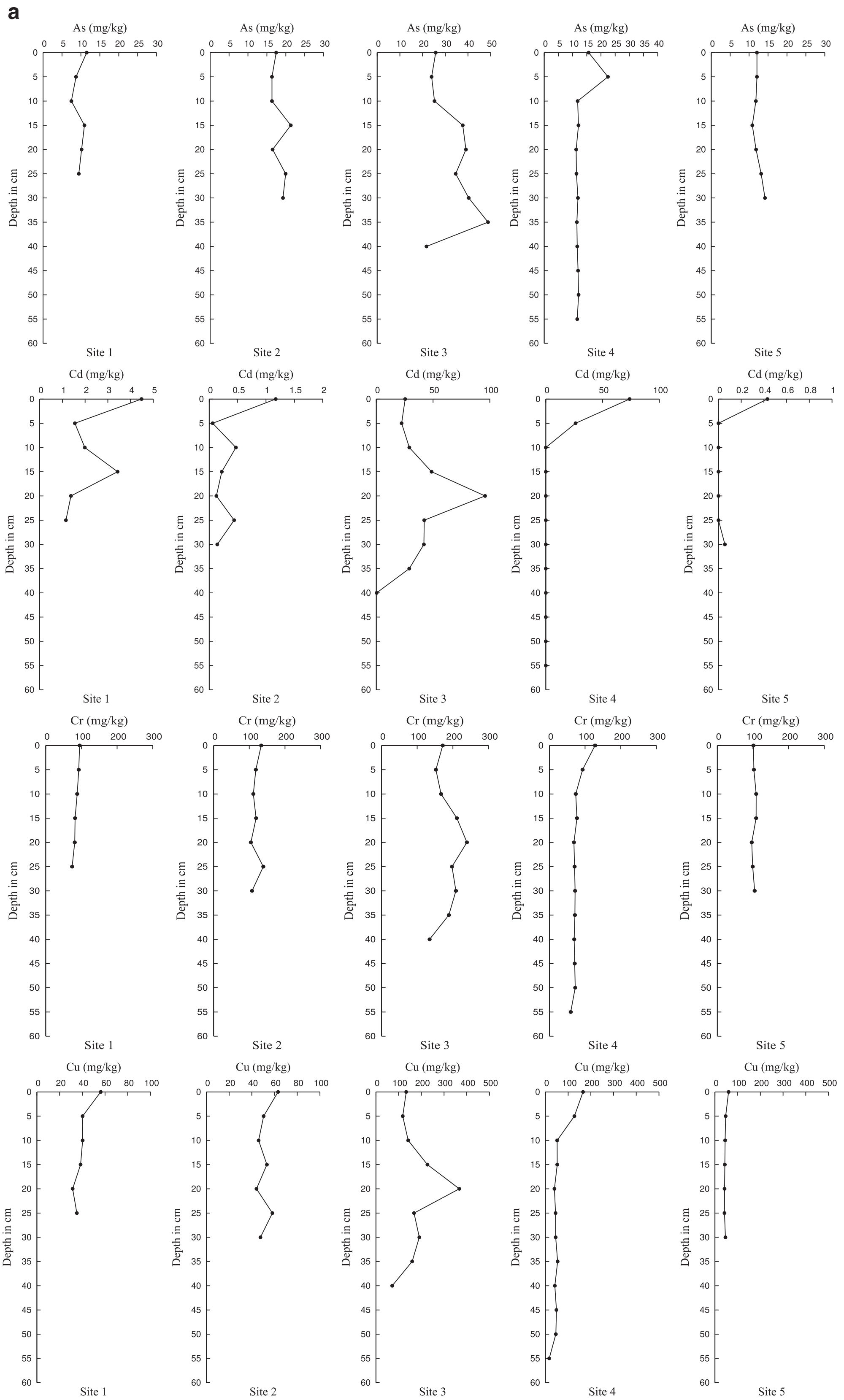

Fig. 4 Depth-wise distribution of heavy metals in Hussain Sagar lake

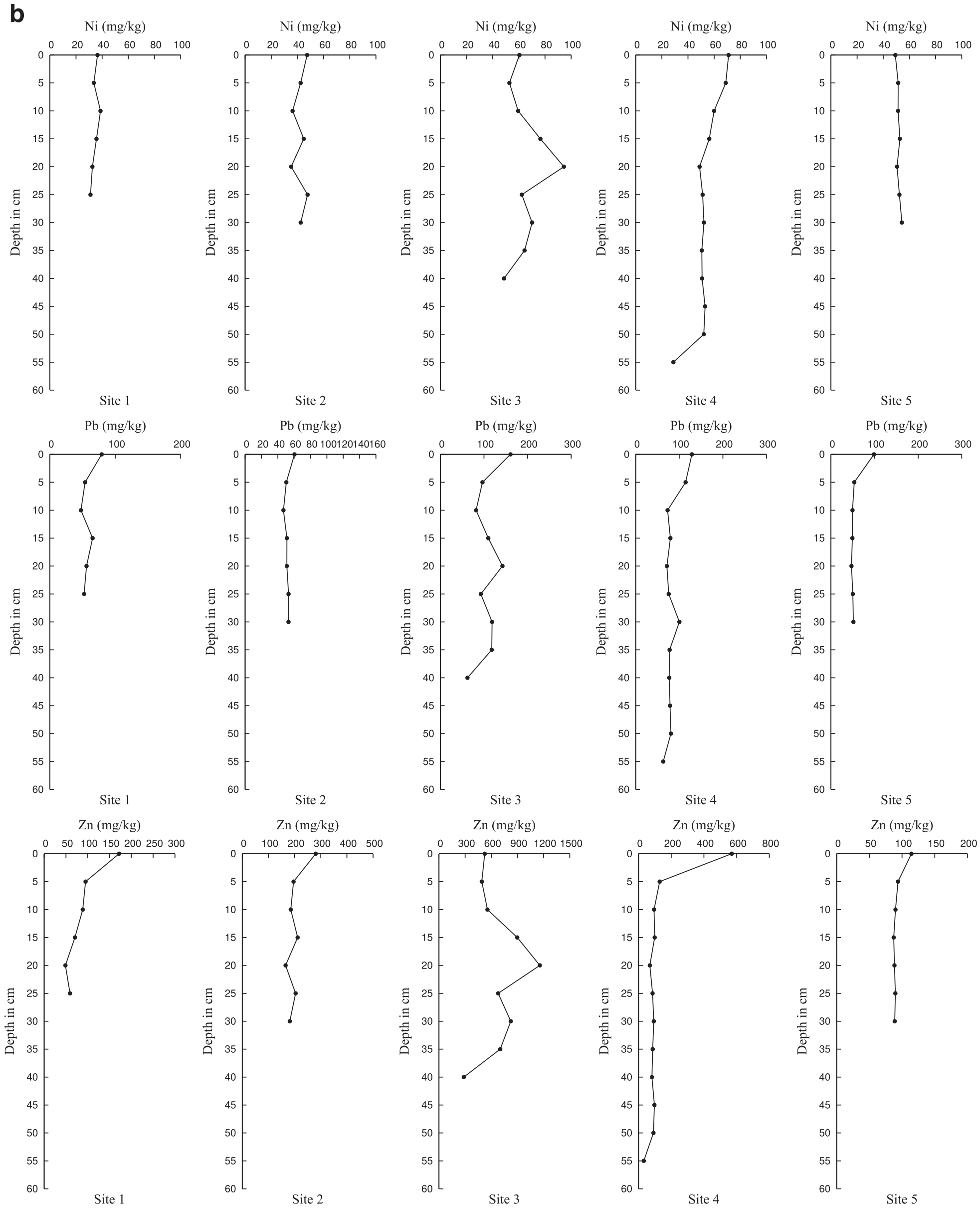

Fig. 4 continued

and Ni in surface water at site 3 is found to be higher, showing the significant contamination by industrial discharge compared to site 1 and 2. The possible industrial sources of these heavy metals are Cd, Ni, and Zn from batteries and electroplating effluent, Pb from paint and battery manufacturing effluent, and Cu from metal plating effluent. It was revealed that the constant incoming flow of industrial discharge in site 3 caused the distribution of these heavy metals in deep sediments (0–40 cm). Conversely, the heavy metals concentration in surface water at site 4 and 5 are found to be less but more significant concentrations were observed in surface sediments (0–10 cm). The sources of Cr, Ni, Zn, and Pb are from tanning and dyeing industrial effluent, urban runoff, vehicular deposits, and idol components such as paints and decoratives. The higher heavy metal concentrations in surface sediments at site 4 is majorly due to the interactions with increased suspended solids (i.e., Plaster of paris, fine clay, jute, and straw) caused by idol immersion. The sorption and desorption study revealed that Pb and Cu retained to a greater extent due to high clay and organic content. It was found that 25–50% of Zn and 50–80% of Cd are associated with exchangeable and carbonate fraction and classified under high risk to a very high-risk category

Fig. 5 Plots of amount of heavy metals adsorbed and retained during sorption and desorption experiments

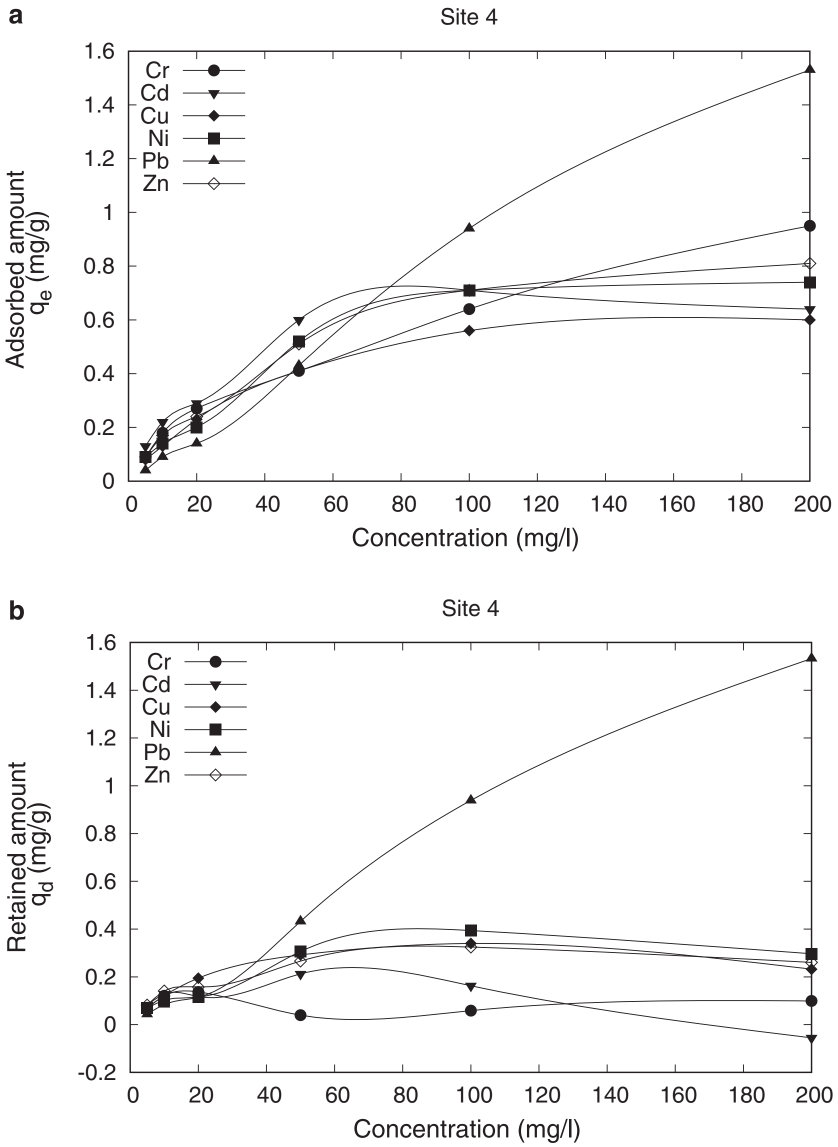

Table 11 Langmuir and Freundlich correlations for sorption and desorption study

Heavy metals	Langmuir			Fruendlich		
	R^2	Q_o (mg g^{-1})	b (L mg^{-1})	R^2	K_F (L mg^{-1} g^{-1})	n (g L^{-1})
Sorption						
Cr	0.9317	0.4935	0.0508	0.9843	0.1299	2.8770
Cd	0.9305	0.3316	0.0503	0.9729	0.1162	2.4606
Cu	0.7315	0.3100	0.1285	0.9945	0.0941	2.9820
Pb	0.9810	2.8425	0.0055	0.9793	0.0156	1.0277
Ni	0.7315	0.310	0.1285	0.9157	0.1205	2.4813
Zn	0.8048	0.4584	0.0693	0.9606	0.1272	2.4813
Desorption						
Pb	0.9683	1.3385	0.0117	0.9793	0.0157	1.0277
Cu	0.9489	0.2579	0.5790	0.7756	0.0997	3.9154

by the risk assessment code. The potential risk of these heavy metals are in increasing order as Cd > Zn > Cr > Pb > Ni > Cu > As > Co. Out of all heavy metals, the EF value of sediments for Cd, Zn, Cu, and Pb were above 2 that signifies the pollution level by anthropogenic activities. Thus, our study shows the extent of contamination by evaluating the distribution of heavy metals for appropriate remediation and preventive measures to restore the lake quality and its ecosystem.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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