

Risk Assessment and Mitigation Measures on the Heavy Metal Polluted Water and Sediment of the Kolleru Lake in Andhra Pradesh, India

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ABSTRACT: The Kolleru Lake is a famous Ramsar wetland of international significance. In this study heavy metal contents in water and sediment samples are reported. It is found that certain potentially toxic metal ions like chromium (4.5–80 µg/L), copper (1–20 µg/L), manganese (1–313 µg/L) and zinc (1.2–57 µg/L) are present in variable quantities in the lake water. When normalized with respect to concentration of each element in clean surface waters, the normalized ratio is found to be highly heterogeneous (chromium=4.5–80, copper=0.3–3.3, manganese=0.07–20.8, zinc=negligible to 2.8). At several places, the normalized ratio is greater than 1, indicating anthropogenic input. The concentration of iron (4–20 µg/L) in water, however, is less compared to the clean surface waters. Chemical analyses and quality assessment of Kolleru Lake sediments have been carried out through estimation of four pollution indices, which include enrichment factor (*EF*), geoaccumulation index (*I_{geo}*), contamination factor (*CF*) and pollution load index (*PLI*). Evaluation of these contamination indices with respect to average sediment composition of Taylor & McLennan (2001) confirmed that the Kolleru Lake sediment is polluted with a number of heavy metals that include cobalt (*EF*=2, *I_{geo}*=0.64, *CF*=2.4), chromium (*EF*=1.5, *I_{geo}*=0.18, *CF*=1.7), copper (*EF*=1.6, *I_{geo}*=0.29, *CF*=1.9), manganese (*EF*=1.3, *I_{geo}*=0, *CF*=1.4), vanadium (*EF*=1.5, *I_{geo}*=0.19, *CF*=1.7) and zinc (*EF*=1.5, *I_{geo}*=0, *CF*=1.5). The level of contamination, however, is minor to moderate and is in good agreement with the heavy metal chemistry of the lake water. Based on these results some measures for environmental rehabilitation of the lake and its surroundings have been proposed.

Keywords: Kolleru Lake; Ramsar wetland; toxic element; pollution indices; ecosystem.

INTRODUCTION

Lakes provide livelihood to many people across the globe. In developing countries, more than 60 million people rely on lakes for their income (FAO, 2015). However, pollution of Lake Ecosystem and its surroundings due to anthropogenic activities has become a major global issue in many parts of the world. The Anzali

wetland in Iran is registered under the Ramsar International Convention. This wetland is located along the southwestern part of the Caspian Sea coast and receives wastewater discharges from domestic, agricultural, and industrial sources (Hargalani et al., 2014). The total concentration of metals as well as their chemical partitioning and bioavailability in the sediments were measured to calculate the anthropogenic contribution of metals in

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the sediment (Hargalani et al., 2014). The Bamdezh wetland in Iran is another habitat, which represents a good food source for aquatic ecosystem, besides attracting migratory birds. Geochemical studies conducted on the sediments of this wetland indicate that the level of pollution due to anthropogenic activities vary significantly in different parts of the wetland, which range from unpolluted through moderately polluted to strongly polluted (Vaezi et al., 2015a). In a recent study Koff et al. (2016) have demonstrated the use of paleolimnology as a potential tool to ascertain vital information from sediments, which in turn, allows a historical reconstruction. The extent of the effect of anthropogenic pollutants or other environmental stressors can be pinpointed so that adequate insight for eutrophication assessment can be gained. Preservation, restoration and management of water bodies can therefore be planned in a better way (Koff et al., 2016). In another significant study, Vaezi et al. (2015b) analyzed sediment samples from the Petrochemical Special Economic Zone of Mahshahr Bay, Persian Gulf. They recorded high concentration of trace metals in the sediments, whose origin was entirely geogenic. Therefore they cautioned that the assessment of pollution hazard on aquatic bodies in such geological set up should not be conducted through conventional approaches.

In India, the dependency on lakes for livelihood is also seen in different parts as evidenced from the explosion of agricultural and livestock farming, fishery industries and tourism around many lakes of the country (Zutshi & Khan, 1988; Gupta et al., 2001; Dahdouh-Guebas et al., 2006; Iwasaki & Shaw, 2009). All these Indian lakes are subjected to human-induced adverse impacts, which are undesirable. The Kolleru Lake is one of the largest natural freshwater lakes in India located in the southern state of Andhra

Pradesh. During August 2002 the lake has been designated as a wetland of international importance (see Wikipedia for List of Ramsar sites in India) under the International Ramsar Convention. Wetlands are of considerable economic importance, as they nourish diverse biota and are natural habitat for a large variety of resident and migratory birds. Naturally, the Kolleru Lake was also a “paradise of birds” and used to be a place of tourist attraction. However, during the past two and half decades the lake habitat has degraded significantly owing to various anthropogenic activities around its vicinity that include intensive agricultural, industrial and farm activities. For example, Amaraneni & Pillala (2000) made an assessment of the impact of aquaculture on the water quality of Kolleru Lake. They collected water samples from selected fish ponds in every 2 months during production year and analysed them for different physicochemical parameters. The results indicate an increase in mineral and heavy metal concentration in the waters. They also carried out further studies on fish samples (Amaraneni & Pillala, 2001). The following maximum concentrations of pesticide residues in Kolleru Lake fish on wet weight basis were reported: alpha-BHC=123.8 µg/g, gamma-BHC=98.7 µg/g, malathion=2.5 µg/g, chlorpyrifos=88.6 µg/g, isodrin=0.91 µg/g, endosulfan=76.5 µg/g, dieldrin=1.98 µg/g, and p,p'-DDT=157.4 µg/g. The complete data set were helpful to evaluate the baseline value and the extent of pesticide pollution in the lake's ecosystem (Amaraneni & Pillala, 2001). Chandra Sekhar et al. (2003) reported concentrations of Zn, Cd, Cu, Pb, Cr, Ni and Co in sediment samples and three different species of fishes, viz., *C. striata*, *C. catla* and *O. Mossambicus* from the Kolleru Lake. The metal concentrations in sediments were found to be greater than the background values of metals compared to the Indian sediments. This indicates

anthropogenic origin of metals in the Kolleru Lake sediments. The concentrations of Zn, Cu and Pb in different species of fishes were also found to be very high in the following order: gills > liver > muscle. Based on these results they concluded that the Kolleru Lake fishes are contaminated with metals. Hence it is advisable not to use them for human consumption (Chandra Shekar et al., 2003). Subsequent study was carried out by Mastan (2014) on the heavy metal concentrations of cadmium, zinc, copper, chromium, lead and mercury in various tissues of *Labeo rohita* and *Channa striatus* and in the water samples. These samples were collected during 2009-2010. The results indicated that the concentrations of heavy metals in different organs of fishes vary. While *L. rohita* followed the order liver > kidney > gills > muscles, in the case of *C. striatus*, the order was liver > kidney > gills > muscle. Based on the obtained results, Mastan (2014) concluded that the concentrations of heavy metals are within the maximum permissible levels for drinking water and fish. Rao et al. (2004) processed the IRS-ID LISS-III multi-band digital image data of the Kolleru Lake region using various image enhancement techniques. Their study revealed that more than 50% of the original lake was lost due to high sedimentation rate, and these elevated portions of lake areas were subsequently occupied by local residents for aquaculture and agriculture. The rest of the lake also did not show any stretch of clear water, and it is either covered by weed or dried out by reclamation. In a recent study on characterization of water chemistry of the Kolleru Lake in conjunction with its sediment ^{13}C and ^{18}O compositions, Das Sharma & Sujatha (2016) documented hazardous effect of saline water intrusion into the lake. This poses a serious threat to the lake ecosystem. Three major contributors for seawater intrusion into the

lake have been identified. These are: (i) prevalence of lake's dry condition originating from excessive groundwater exploitation for agriculture and other purposes surrounding the lake areas, (ii) reduced inflow and outflow of freshwater into the lake due to piling up of huge sediments and residues that drain into the lake, and (iii) proximity of the lake (~35 km inland) to the Bay of Bengal (Das Sharma & Sujatha, 2016). We also documented highly heterogeneous distribution of certain potentially toxic metal ions like chromium, copper, manganese and zinc in the lake waters. Indiscriminate disposal of domestic and industrial effluents in and around the lake perhaps are responsible for the presence of these toxic heavy metals (Das Sharma & Sujatha, 2016). Based on these results, several remedial measures were proposed by us to revive the lake ecosystem. One of the measures was to restore free flow of water in the lake. Therefore it was recommended to clean up the lake bed at a regular interval so that a deep lake bed is ensured. This, in turn, would facilitate recharge of large volumes of freshwater into the lake during the rainy seasons. In this context it is important to note that the lake and its surroundings receive an average annual rainfall in excess of 1000 mm of which ~60% is received during south-west monsoon period lasting from mid June to mid October (ISRO, 2011).

In view of the obtained results from our previous study and above-mentioned recommendation to clean up the lake bed at a regular interval (Das Sharma & Sujatha, 2016), it becomes mandatory to study the heavy metal concentrations in surface sediments of the Kolleru Lake. This is because, if the sediments of the Kolleru Lake are contaminated by heavy metals due to anthropogenic activities, and if these sediments are dumped in the lake surroundings (as a natural choice to save cost) during cleaning operation, then the

soil may become contaminated. This is highly undesirable. Furthermore, it is also important to note that sediments play a crucial role in water quality. They remove elemental pollutants from the water; however, the pollutant elements that have accumulated in the sediments may diffuse back into the surface waters or penetrate into the underground waters. Therefore, the sediments may become “reservoirs” for toxic and heavy metals, created from anthropogenic activities (Huy et al., 2002). As a follow up study, in this work I therefore report major and trace element (mostly heavy metal) concentrations in sediments of the Kolleru Lake. These sediment samples were collected along with the water samples from the same locations, and the trace element contents in water samples were reported earlier (Das Sharma & Sujatha, 2016). The contamination level in sediments is estimated using geochemical indices such as metal enrichment factor (EF_{metal}), geoaccumulation indices (I_{geo}), contamination factor (CF) and pollution load index (PLI). An assessment is then made to evaluate the extent of anthropogenic contribution on the measured parameters.

MATERIALS AND METHODS

In view of international significance of the area (a Ramsar site), a brief account of the study area is presented first. The Kolleru Lake, located between the latitudes $16^{\circ} 32'$ and $16^{\circ} 47'$ N and longitudes $81^{\circ} 05'$ and $81^{\circ} 27'$ E, is the largest natural freshwater lake in the southern state of Andhra Pradesh. The lake lies between the deltas of two main rivers, viz., the Godavari and the Krishna and it is approximately 35 km inland from the present day shoreline (Fig. 1). There are three main streams that drain into the lake. These streams are known as the Budimeru, the Thammileru and the Ramileru. The lake finally drains itself into the Bay of Bengal through the Upputeru

river (see also Rao et al., 2004; Das Sharma & Sujatha, 2016). It is a shallow freshwater body with a natural spread of $\sim 674 \text{ km}^2$. However, its areal extent goes up to $\sim 954 \text{ km}^2$ during the rainy seasons when highest floods are recorded in the area. On the other hand, during dry seasons the areal extent comes down to as low as $\sim 66 \text{ km}^2$. It is significant to note that the lake also serves as a natural flood-balancing reservoir for the two major rivers— the Godavari and the Krishna. Fluvio-marine deposits (sand, silt and clay) characterize the lake surroundings from all sides. The nearest rock formations occur about 10 km away from the lake. The geological formations include khondalite, Gondwana, Deccan traps and tertiary sediments (Amaraneni, 1997; Das Sharma & Sujatha, 2016). Various types of macrophytes such as water hyacinth (*Eichhornia crassipes*), elephant grass (*Pennisetum purpureum*), *Salvania sp.*, *Ipomea aquatica* and many others have invaded the lake. It has also been documented that prolific use of fertilizers and pesticides, discharges from fish pond, disposal of domestic wastes and sewage, and discharge of industrial effluents and agricultural runoff carrying inorganic nutrients have severely affected and altered the ecological character of the lake ecosystem (Azeez et al., 2011).

Eighteen sampling points were selected to cover the lake area (Fig. 1). Uniform sampling could not be carried out due to inaccessibility of some of the locations. The water samples were collected during summer of 2013 following standard procedure (USEPA, 1983). Polypropylene bottles (1 litre) for sample collection were soaked in 5% HNO_3 for 24 h followed by rinsing them a number of times with deionized water. The bottles were dried and water samples were collected in duplicate after filtration using Whatmann No. 40 filter paper. The pH and total dissolved solids (TDS) were measured

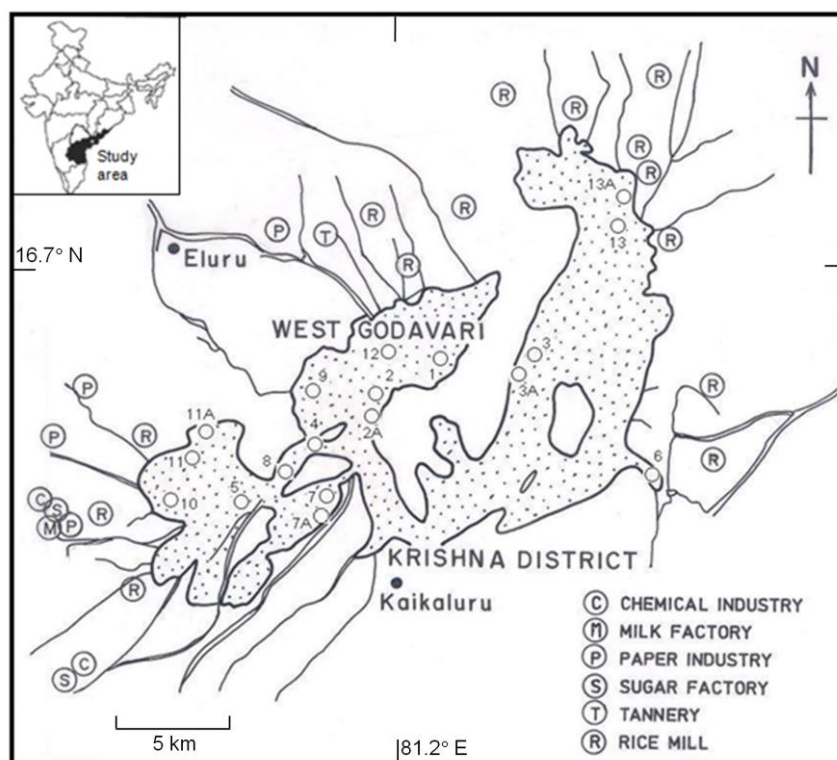


Fig. 1. Map showing Kolleru Lake. Two prominent towns close to the lake, viz., Eluru and Kaikaluru together with various anthropogenic inputs surrounding the lake area are shown (modified after Amaraneni & Pillala, 2001). Eighteen sampling points for water and surface sediments are marked by open circles. The horizontal scale (5 km) displayed in the figure corresponds to 0.045°. The geographical location of the lake (study area) in the state of Andhra Pradesh, India is marked in the inset.

in the field. The pH electrode was calibrated with pH 4 and pH 9.2 buffer solutions. The samples were then acidified with 5 ml HNO₃ to prevent the adsorption of heavy metals onto the bottle walls and transported to the laboratory. Until use, preservation of these water samples was done following the recommendations of USEPA (1983). The heavy metal analyses were done in the laboratory within 2 to 3 weeks of the sample collection. The concentrations of cations such as Fe, Mn, Cr, Cu and Zn were determined by ICP-OES (model Optima 4300 DV) using the standard procedure (see APHA, 1995; Das Sharma & Sujatha, 2016).

The sediment samples were collected along with water samples from the same locations as that of the water samples. These sediments are primarily alluvium and made up of fine particles of silt, clay and relatively coarse grained sand.

Carbonate constitutes only a minor fraction in them compared to silt, clay and sand (Das Sharma & Sujatha, 2016). Samples were collected from a depth of 0–10 cm in self-locked polythene covers and were analyzed within 2 months from the collection date for 10 major (Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K and P) and 10 trace elements (As, Co, Cr, Cu, Mo, Ni, Pb, V, Zn, and Zr). The sediment samples were air dried and kept in an oven for two days at 60°C temperature. The dried sediments were then disaggregated using an agate mortar and pestle and sieved through a 2 mm sieve, homogenized and stored at 4°C until use. Before analysis, the sample was finely powdered to –250 mesh size (US Standard). Pressed pellets were prepared by using collapsible aluminium cups. These cups were filled with boric acid and about 1 gm of the finely ground powdered sample was put on the top of the boric acid

in each cup and pressed under a hydraulic press at 20 tons pressure to get a pellet (Rao & Govil, 1995). The major and trace element concentrations were measured on the sediment samples using a Philips PW-2440 fully automated microprocessor-controlled, 168-position automatic PW 2540 vrc sample changer X-ray fluorescence spectrometer. Suitable software 'Super Q' was used to define dead-time correction and inter element matrix-effects. International sediment reference materials from the US Geological Survey and Canadian Geological Survey (LKSD 1, TILL 1 and TILL 3) were used to prepare the calibration curves for trace elements (for details of XRF procedure, see Rao & Govil, 1995; Krishna et al., 2007). During the course of XRF analyses, the QA/QC plan was strictly adhered to using international sediment reference material, thereby ensuring quality data.

Four pollution indices are estimated for the purpose of environmental risk assessment. These indices are: enrichment factor (*EF*), index of geoaccumulation (*I_{geo}*), contamination factor (*CF*), and pollution load index (*PLI*). A brief account of these parameters is presented in Appendix-A.

RESULTS AND DISCUSSION

The water samples from Kolleru Lake are near neutral to alkaline (pH varying from 6.8 to 8.8). The total dissolved solids (TDS) in these waters are variable ranging from 1006 to 5285 mg/L, indicating that they belong to brackish water type, which has typical TDS contents of ~1000 mg/L or more (Freeze & Cherry, 1979). The range, average and standard deviation of the concentrations of a few selected trace metals in the water samples of the Kolleru Lake are presented in Table-1. The normalized concentration ratio of these metals is presented in Fig. 2a. The normalization has been carried out with respect to the median values of

concentration of these toxic elements that were reported for clean and uncontaminated surface waters (data from Langmuir, 1997). Values in excess of 1 indicate enrichment and those below 1 specify depletion. It can be seen from Fig. 2a that Cr is enriched and Fe is depleted, while the other elements like Cu, Mn and Zn show both enrichment as well as depletion at various locations (see Figs. 1 and 2a).

Figure 2a points to the fact that the distribution of the analyzed elements in water of the lake is highly heterogeneous; indicating that both free flow and internal mixing of water in the lake is obstructed. Furthermore, the observed heterogeneous distribution could be due to the presence of various types of macrophytes such as water hyacinth (*Eichhornia crassipes*), elephant grass (*Pennisetum purpureum*), *Salvania sp.*, *Ipomea aquatica* and many others, some of which have the potential to purify water from various toxic metals (see also Das Sharma & Sujatha, 2016). To cite an example, the toxic metal removal potential of water hyacinth was evaluated by Stephenson et al. (1980). It was found by these researchers that water hyacinths can be used as remediation plants for several toxic metals like Cd, Co, Cr, Cu, Fe, Pb etc. Likewise, Dhote & Dixit (2009) reviewed the role of macrophytes in order to improve the water quality by preferentially scavenging different heavy metals. In view of the above, it is inferred that the observed heavy metal concentration in water samples of Kolleru Lake may be considered as the minimum limit. In the absence of macrophytes, it is possible that the concentration of heavy metals in water would have been much higher. In order to understand the overall contamination of waters in the lake, the average normalized ratios corresponding to each element is also plotted in Fig. 2b. From this plot it is evident that the average levels of Cr, Cu and Mn are definitely

high, whereas the level of Zn is marginally high. Although with the available data it is difficult to point out the exact source/sources and assess the relative contribution of each source for the observed high concentration of toxic heavy elements in the lake water, it is certain that unchecked anthropogenic activities in and around the Kolleru Lake area are responsible for the degradation of its water quality (cf. Das Sharma & Sujatha, 2016).

In view of the phytoremediation potential of various species discussed here, it is relevant to ask whether these species in the lake ecosystem need to be maintained or they should be eliminated. As proposed in our previous study, it is stressed that after the phytoextraction processes, these species need to be disposed off safely during the initial stages of restoration phase of the lake (see also Das Sharma & Sujatha, 2016).

Table 1. Range, average and standard deviation of 5 selected trace metal concentrations in water samples collected from the Kolleru Lake in Andhra Pradesh. Similar data on 10 major elements and 10 heavy metals pertaining to the surface sediments of the Kolleru Lake are also presented. For comparison, range, average and standard deviation on heavy metal contents in sediment reported earlier by Chandra Sekhar et al. (2003) and Vijayalakshmi & Brahmaji Rao (2017) are also included in the table.

Trace metal content in water (this study)										
µg/L	Cr	Cu	Fe	Mn	Zn					
Maximum	80.0	20.0	20.0	313.2	57.0					
Minimum	4.5	1.0	4.0	1.0	1.2					
Average	12.5	6.9	9.2	55.5	21.3					
Std. deviation	17.06	6.0	4.5	96.74	17.0					
Major element content in sediment (this study)										
Oxide (wt%)	SiO ₂	TiO ₂	Al ₂ O ₃	Fe ₂ O ₃	MnO	MgO	CaO	Na ₂ O	K ₂ O	P ₂ O ₅
Maximum	59.9	1.3	19.0	11.7	0.2	2.6	7.8	0.8	2.50	4.47
Minimum	53.1	0.7	11.4	6.1	0.1	1.7	1.4	0.4	1.38	0.13
Average	55.7	1.1	15.7	10.2	0.1	2.2	3.0	0.5	1.88	0.50
Std. deviation	1.89	0.13	1.93	1.41	0.04	0.23	1.63	0.14	0.29	1.01
Heavy metal content in sediment (this study)										
mg/kg	As	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	Zr
Maximum	30.1	44.1	154.1	95.3	3.4	61.1	58.3	228.6	169.5	229.2
Minimum	1.1	22.6	97.0	54.6	0.4	31.1	1.2	127.7	81.2	136.5
Average	6.1	37.9	126.0	74.0	1.2	50.1	13.3	190.1	97.3	174.5
Std. deviation	6.34	5.36	12.72	9.97	0.7	8.37	12.23	25.28	20.69	23.25
Heavy metal content in sediment (Chandra Sekhar et al., 2003)										
mg/kg	As	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	Zr
Maximum	-	4.4	66.0	572.0	-	2.2	5.6	-	671.0	-
Minimum	-	2.0	44.0	270.0	-	0.2	2.5	-	356.0	-
Average	-	3.0	52.3	409.9	-	1.1	4.1	-	510.1	-
Std. deviation	-	0.6	5.7	86.3	-	0.8	0.9	-	98.2	-
Heavy metal content in sediment (Vijayalakshmi & Brahmaji Rao, 2017)										
mg/kg	As	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	Zr
Maximum	-	-	160.0	79.9	-	135.2	4.3	-	88.8	-
Minimum	-	-	43.0	14.0	-	5.2	0.3	-	26.2	-
Average	-	-	127.6	55.7	-	84.8	1.5	-	69.6	-
Std. deviation	-	-	43.0	22.13	-	42.5	1.59	-	19.63	-

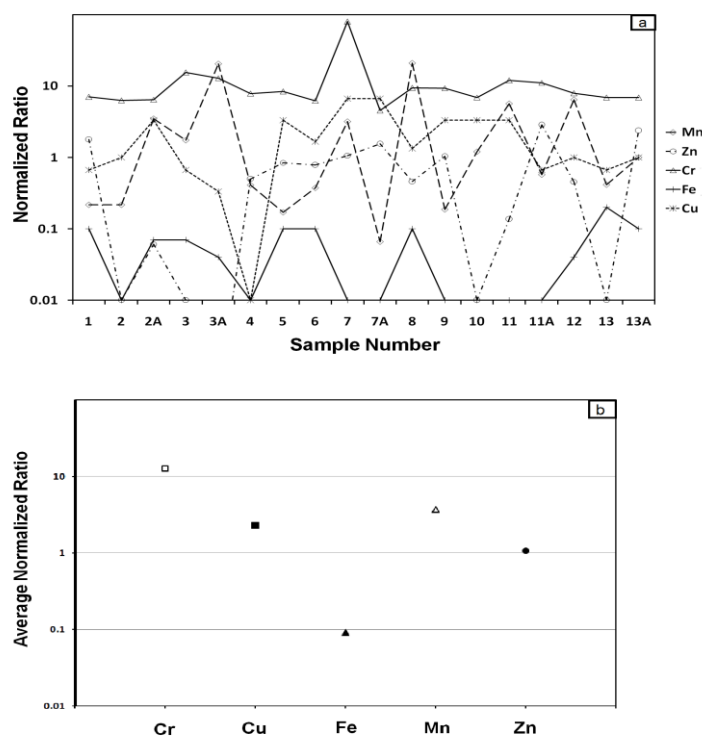


Fig. 2. Plot showing heavy metal contaminated water chemistry of the Kolleru Lake. The normalized and average normalized ratios of some toxic trace metals in the water samples of the Kolleru Lake are plotted for (a) individual sampling site and (b) the entire lake, respectively. Note that values greater than 1 indicate enrichment and those lower than 1 specify depletion (for normalization, data from Langmuir, 1997 is used).

The range, average concentration and standard deviation corresponding to each major and trace element analyzed in this study pertaining to 18 sediment samples are presented in Table–1. It is important to mention here that of all the sediments, sample number 8 is highly anomalous. In that it shows the maximum concentrations for As, Cr, Mo and Pb. Four other elemental concentrations (viz. Co, Cu, Ni and V) for this sample are also high (more than 0.8 times of the maximum value). Such high values perhaps reflect point source contamination and could originate from localized anomaly due to illicit dumping/discharge of contaminants.

It has been documented that the abundance of heavy metals in sediments normally follows the order as: Fe > Mn > Zn > Cu > Pb > Cr > Ni, which corresponds to the natural progressive concentration of heavy metals in sediments (Turekian & Wedepohl, 1961). Almost similar ranking

was reported by many researchers in sediments from different aquatic bodies (e.g., Krupadam et al., 2003; Kumar et al., 2011; Vaezi et al., 2016; Amira & Leghouchi, 2018). It is interesting to note that with the exception of Cr, similar order was also observed for the Kolleru Lake sediments (see Table–1 for comparison) by Chandra Shekar et al. (2003). However, the ranking order observed in the present study differs for Cr and Ni, with the average concentration of Cr being very high (~126 mg/kg, Table–1). The ranking order in the present study is almost similar to that of Anzali Wetland sediments reported by Esmailzadeh et al. (2016), where industrial, agricultural and municipal wastewaters flow into the wetland. In order to get an idea as to how the average abundances of various heavy metals analysed in the present study compare with those of one and half decade old data, the average abundances of heavy metals reported by Chandra Shekar et al. (2003) are presented in

Table-1. It can be noted from Table-1 that while the average abundances of Cr, Ni, Co and Pb of Kolleru Lake sediments exhibit considerable increase, those of Zn and Cu show depletion when compared with the data of Chandra Shekar et al. (2003). Therefore the present study points out that the average abundances of Cr, Ni, Co and Pb in the Kolleru Lake sediments have enhanced significantly with time, whereas those of Zn and Cu reduced considerably. Recent data on Zn and Cu reported by Vijayalakshmi & Brahmaji Rao (2017) for Kolleru Lake sediments also exhibit lower average abundances, whereas those of Cr and Ni show higher average concentrations (see Table-1). The results obtained in the present study are therefore in good agreement with those of Vijayalakshmi & Brahmaji Rao (2017) (see Table-1).

In the present study total metal concentration was measured by XRF method. However, it is now well established fact that in order to evaluate the possible toxicity or risk of environmental pollution due to heavy metals present in sediments, the types of association between metals and the sediment must be assessed. This can be undertaken by adopting sequential extraction techniques (e.g., Tessier et al., 1979). Therefore in order to understand the abrupt drop in the average abundances of Cu and Zn as documented in the present study, reference can once again be made about the study of Chandra Shekar et al. (2003), who employed sequential extraction of metals from sediments. Based on the extractable contents of metals in sediments using sequential extraction scheme, these researchers argued that all metals were mainly associated with the oxidizable (organic matter bound) fraction and residual fraction as indicated by the partitioning patterns for Zn, Cd, Cu, Pb, Cr, Ni and Co in sediment samples. Such observation also allowed them to predict the mobility of metals in sediments and thereby their entry into human food chain. According to the results obtained by them, four metals

(Zn, Cd, Cu and Pb) are dominantly associated with the fraction which is mobile (exchangeable and carbonate bound). In other words, the bioavailability of these metals is more to aquatic life. The abrupt reduction in the average abundances of Zn and Cu in Kolleru Lake sediments reported in the present study may possibly be attributed to their bioavailability to aquatic life. The increase in the average abundances of Cr, Ni, Co and Pb with time is unambiguously a reflection of continued anthropogenic activities in the vicinity of the lake.

The data on heavy metal concentration was also subjected to statistical analyses. Based on the correlation matrix, analyses were performed to understand the interrelationship between heavy metals in the Kolleru Lake sediments. The results of the analysis are shown in Table-2. The correlation matrix shows positive correlations for the element pairs: As-Pb ($r=0.98$), As-Mo ($r=0.71$), Cr-Mo ($r=0.6$), Cr-Ni ($r=0.59$), Cr-Pb ($r=0.59$), Cu-Zn ($r=0.59$), Mo-Pb ($r=0.74$), Zr-V ($r=0.73$), As-Cr ($r=0.56$), and Co-Ni ($r=0.53$) etc. This suggests that these metal pairs are (i) either associated to the lake sediments because they probably have similarity in geochemical behaviour and a similar origin related to the chemical and physical weathering (Vaezi et al., 2015b) and/or (ii) an anthropogenic origin related to the discharge of wastewaters from domestic, industrial and agricultural activities (Zhang et al., 2011). In the case of Kolleru Lake sediments, it is inferred that the observed high metal contents originated from anthropogenic activities. This interpretation is supported by (i) observed increase in the average abundances of Cr, Ni, Co and Pb over a period of one and half decade (see data presented in Table-1 pertaining to this study and those reported earlier by Chandra Sekhar et al., 2003) and (ii) many analysed heavy metals are largely associated to the exchangeable phase as documented by Chandra Sekhar et al. (2003).

Table 2. Correlation matrix of different heavy metals in Kolleru Lake sediments.

Element	As	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	Zr
As	1									
Co	-0.11	1								
Cr	0.56	0.44	1							
Cu	0.26	0.07	0.17	1						
Mo	0.71	0.21	0.60	0.23	1					
Ni	0.10	0.53	0.59	-0.01	0.27	1				
Pb	0.98	-0.04	0.59	0.18	0.74	0.21	1			
V	-0.16	0.59	0.33	0.23	-0.15	0.06	-0.20	1		
Zr	0.33	-0.15	0.38	0.59	0.38	0.16	0.27	-0.13	1	
Zn	-0.30	0.18	0.04	0.09	-0.28	-0.24	-0.39	0.73	0.06	1

Lake sediments can act as sink for a large number of contaminants that can be stored in the bed sediments. If the environmental conditions change within the sedimentary column, these sediments can become the source of pollution. In view of all these, four pollution indices have been estimated for the purpose of environmental risk assessment of the Kolleru Lake sediments. It is to be borne in mind that for the estimation of various pollution indices, the choice of an appropriate background assumes extreme importance. Different researchers have made use of compositions of various background references for the estimation of pollution indices (Förstner & Müller, 1973; Sinex & Helz, 1981; Salomon & Förstner, 1984; Chakravarty & Patgiri, 2009; Perunović et al., 2015; Poh & Tahir, 2017). These background references include (i) average shale (Turekian & Wedepohl, 1961), (ii) composition of the average continental crust (Taylor, 1964; Rudnick & Gao, 2003), (iii) composition of world soil baseline reference (Ure & Berrow, 1982), (iv) soil in relatively unpolluted areas and borehole sediment composition as a standard (Perunović et al., 2015), and (v) composition of world surface rock average (Martin & Meybeck, 1979).

In the present study in order to choose a suitable background, an ACNK diagram (Nesbitt & Young, 1984) corresponding to 18 sediment samples collected in this study from the Kolleru Lake is considered. Since degradation of feldspar and concomitant formation of clay minerals are the dominant processes during chemical weathering,

elements such as Ca, Na and K that are labile get removed easily from the feldspar during weathering. Therefore the abundance of these elements with respect to resistant elements can be used as a measure to deduce the extent of chemical weathering (Nesbitt & Young, 1984). The most intensely weathered samples plot in the top corner of the diagram, reflecting preponderance of Al-silicates. As plagioclase is more susceptible to weathering than K-feldspar, Ca and Na are leached preferentially over K, so that the slope of the weathering trend becomes sub-parallel to the A-CN boundary (see Nesbitt & Young, 1989). Figure 3 shows the ACNK diagram of Kolleru Lake sediments along with the compositions of average continental crust (Taylor, 1964) and average sediment (Taylor & McLennan, 2001). It can be seen that both the chosen backgrounds do represent well the weathering trend of the Kolleru Lake sediments, which is sub-parallel to the A-CN boundary. This diagram (Fig. 3) therefore forms the basis of choosing the suitable backgrounds (viz. average continental crust and/or average sediment) for this study in order to assess the environmental risk of the Kolleru Lake sediments.

Table-3 presents the background values of major element and heavy metal concentrations of average continental crust (Taylor, 1964) and average sediment (Taylor & McLennan, 2001). In addition to the above two backgrounds, a third background, viz., world surface rock average (Martin & Meybeck, 1979), has

also been considered in the present study (see Table–3). Compositions of all these have been used as references in this study

for the estimation of enrichment factor, geoaccumulation index, contamination factor and pollution load index etc.

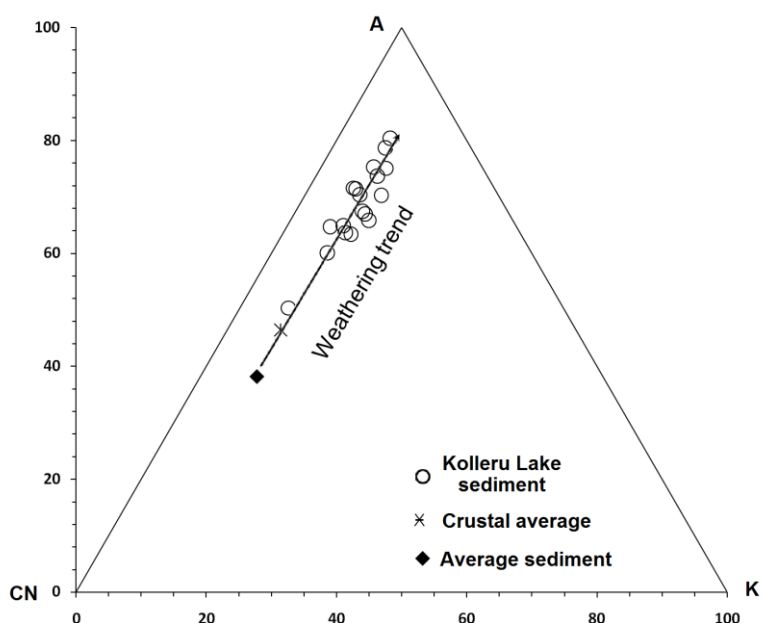


Fig. 3. ACNK diagram for the Kolleru Lake sediments. The compositions of average crustal rock (Taylor, 1964) and average sediment (Taylor & McLennan, 2001) are also plotted in the diagram. The line sub-parallel to the A-CN join indicates the predicted weathering trend (Nesbitt & Young, 1989). Note that most of the sediment samples from Kolleru Lake plot along the predicted trend originating from average crustal rock or average sediment. This diagram therefore forms the basis for selection of suitable background in the present study for estimation of different pollution indices (see text for details).

Table 3. Average values of major element and heavy metal concentrations of various rock materials used as background. Assessment of metal pollution on surface sediments of Kolleru Lake was made by estimating enrichment factor, geoaccumulation index, contamination factor and pollution load index etc (see Table–4 and text).

Major elements										
(wt%)	Si	Ti	Al	Fe	Mn	Mg	Ca	Na	K	P
Average crustal rock (Taylor, 1964)	28.15	0.57	8.23	5.63	0.095	2.33	4.15	2.36	2.09	0.105
Average sediment (Taylor & McLennan, 2001)	30.0	0.45	7.1	4.0	0.068	1.85	6.40	1.25	2.35	0.0665
World surface rock average (Martin & Meybeck, 1979)	-	0.38	6.93	3.59	0.072	-	-	-	-	-
Heavy metals										
mg/kg	As	Co	Cr	Cu	Mo	Ni	Pb	V	Zn	Zr
Average crustal rock (Taylor, 1964)	1.8	25	100	55	1.5	75	12.5	135	70	165
Average sediment (Taylor & McLennan, 2001)	-	16	74	40	-	40	17	110	65	210
World surface rock average (Martin & Meybeck, 1979)	-	-	97.0	32.0	-	49.0	20.0	-	129.0	-

The average EF values of measured trace metals relative to the concentrations of background elements used in this study for the Kolleru Lake sediments are presented in Table-4. If $1 < EF_{metal} < 5$, then the contamination level is not very significant and it only means minimal to moderate metal accumulation at the sampling sites relative to the background (Sutherland, 2000). Using this criterion it can be inferred that the Kolleru Lake sediment is not much contaminated with the studied metals ($EF < 5$). However, there are some metals which show slight enrichment as indicated by their

$EF > 1$ value (see Table-4). When average crustal rock is considered as the background (Taylor, 1964), the mean values of $EF > 1$ are exhibited by As, Co, Cr, Cu, Pb, V, Zn and Zr. Using average sediment compositions as the background (Taylor & McLennan, 2001), the metals which are characterized by average EF of more than 1 are Co, Cr, Cu, Mn, Ni, V and Zn etc. For world surface rock (Martin & Meybeck, 1979) as background, the metals which show minor enrichment are Cr, Cu and Mn (see Table-4). The estimated average

Table 4. Estimated average values of enrichment factors, geoaccumulation indices, contamination factors and pollution load index for various toxic elements present in the Kolleru Lake sediments. It is important to note that according to Müller (1969) instruction, wherever and whenever a negative value is obtained for geoaccumulation index, it should be replaced by zero. The choice of average crustal rock and average sediment as background references is justified based on weathering trend of Kolleru Lake sediments in the ACNK diagram. In addition, a third background reference, world surface rock, is also chosen for estimation of the pollution indices (see text for details).

Considering aluminium as immobile element, average enrichment factors (EF) for various toxic elements											
Background taken as	EF_{As}	EF_{Co}	EF_{Cr}	EF_{Cu}	EF_{Mn}	EF_{Mo}	EF_{Ni}	EF_{Pb}	EF_V	EF_{Zn}	EF_{Zr}
ACR	3.3	1.5	1.3	1.4	1.0	0.8	0.7	1.1	1.4	1.4	1.1
AS	-	2.0	1.5	1.6	1.3	-	1.1	-	1.5	1.3	0.7
WSR	-	-	1.1	2.0	1.2	-	0.9	0.6	-	0.7	-
Average geoaccumulation indices (I_{geo}^{metal}) for various toxic elements											
Background taken as	I_{geo}^{As}	I_{geo}^{Co}	I_{geo}^{Cr}	I_{geo}^{Cu}	I_{geo}^{Mn}	I_{geo}^{Mo}	I_{geo}^{Ni}	I_{geo}^{Pb}	I_{geo}^V	I_{geo}^{Zn}	I_{geo}^{Zr}
ACR	1.05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
AS	-	0.64	0.18	0.29	0.0	-	0.0	0.0	0.19	0.0	0.0
WSR	-	-	0.0	0.61	0.0	-	0.0	0.0	-	0.0	-
Average contamination factors (CF) for various toxic elements											
Background taken as	CF_{As}	CF_{Co}	CF_{Cr}	CF_{Cu}	CF_{Mn}	CF_{Mo}	CF_{Ni}	CF_{Pb}	CF_V	CF_{Zn}	CF_{Zr}
ACR	3.4	1.5	1.3	1.3	0.7	1.0	0.7	1.1	1.4	1.4	1.1
AS	-	2.4	1.7	1.9	1.4	-	1.3	-	1.7	1.5	0.8
WSR	-	-	1.3	2.3	1.4	-	1.0	0.7	-	0.8	-
Pollution Load Index (PLI) based on equation (4). CF values for different backgrounds have been used.											
PLI (based on average $CF_{As}, CF_{Co}, CF_{Cr}, CF_{Cu}, CF_{Pb}, CF_V, CF_{Zn}$, and CF_{Zr} with ACR as background)											1.45
PLI (based on average $CF_{Co}, CF_{Cr}, CF_{Cu}, CF_{Mn}, CF_{Ni}, CF_V$, and CF_{Zn} with AS as background)											1.66
PLI (based on average CF_{Cr}, CF_{Cu} and CF_{Mn} with WSR as background)											1.60

ACR= Average crustal rock (Taylor, 1964)

AS= Average sediment (Taylor & McLennan, 2001)

WSR= World surface rock average (Martin & Meybeck, 1979)

geoaccumulation indices (I_{geo}^{metal}) of various trace metals in the sediments of the Kolleru Lake are also presented in Table-4. Taking average crustal rock composition as the background (Taylor, 1964), it can be seen that with the exception of arsenic, the average I_{geo}^{metal} values for all the studied trace metals are characterized by $I_{geo}^{metal}=0$, indicating mostly unpolluted sediment quality. This is in contrast with our previous observation where trace metals such as As, Co, Cr, Cu, Pb, V, Zn and Zr in Kolleru Lake sediments are characterized by average EF values of >1 , indicating minor to moderate enrichment. Therefore I_{geo}^{metal} values have been estimated using compositions of average sediment (Taylor & McLennan, 2001) as well as world surface rock average (Martin & Meybeck,

1979) as backgrounds. It can be seen that the estimated I_{geo}^{metal} values for at least four elements, viz., Co, Cr, Cu and V are characterized by moderate positive values and are in conformity with the estimated average EF values of these trace metals (see Table-4 and Fig. 4). It may also be pointed out that both average EF_{As} and I_{geo}^{As} values are more than one (3.3 and 1.05, respectively; see Table-4) when composition of the average continental crust is taken as the background. However, it is believed that such an observation has nothing to do with arsenic contamination of Kolleru Lake sediments and it may be an artefact of extremely low content of arsenic (1.8 mg/kg) in the average crustal rock.

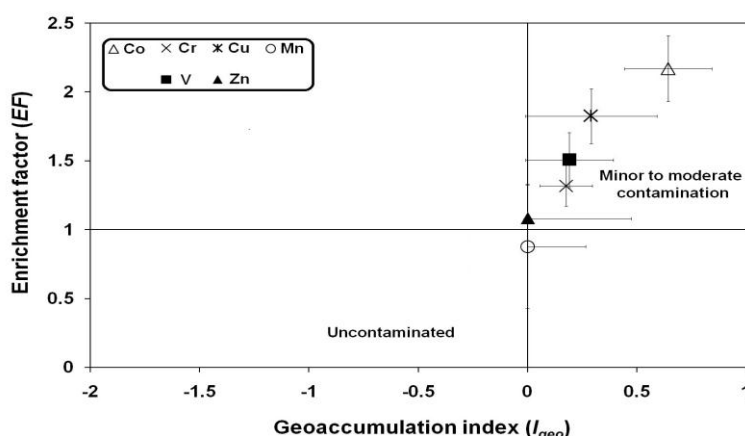


Fig. 4. Average enrichment factor plotted against average geoaccumulation index with respect to average sediment as the background. Concentrations of various toxic metals are considered in the present study to assess the risk associated with the sediment quality of the Kolleru Lake. The horizontal and vertical error bars indicate the standard deviations on average values corresponding to each point. At least four elements (viz. Co, Cr, Cu and V) present in the Kolleru Lake sediments are characterized by average $EF > 1$ and average $I_{geo} > 0$, indicating minor to moderate contamination. It may be noted that according to Müller (1969) instruction, wherever and whenever a negative value is obtained for geoaccumulation index, it should be replaced by zero (Mn and Zn).

The pollution load index (PLI) was proposed by Tomlinson et al. (1980). This parameter provides a measure to assess the sediment quality over an area if the average contamination factor (CF) is considered. For example, if PLI value is zero then the quality of the sediment is considered to be perfect. On the other hand, if this parameter is close

to 1.0 then it is inferred that the contaminants are present in the study area, but it is only at the baseline levels. However, $PLI > 1.0$ is indicative of progressive deterioration of the study area (Tomlinson et al., 1980). In order to estimate PLI value, the contamination factors with respect to any suitable background need to be determined first. The

average contamination factors (CF) for Kolleru Lake sediments are estimated for various toxic elements using compositions of average continental crust (Taylor, 1964), average sediment (Taylor & McLennan, 2001) and world surface rock average (Martin & Meybeck, 1979) as backgrounds (see Table-4). Using these average CF values, PLI in sediments of Kolleru Lake has been calculated. In the computation of PLI , average CF values of only those toxic metals are used for which $CF^{metal}_{average} > 1$. These PLI values corresponding to various backgrounds are presented in Table-4. It can be seen that all the values are greater than 1.0, indicating initiation of deterioration of the study area. Toxic metals such as Co, Cr, Cu, Mn, V and Zn are of major concern. It is also important to note that the results on PLI are in conformity with those of average EF_{metal} and I^{metal}_{geo} values presented above (see also Table-4 and Fig. 4).

Summarizing the results obtained from EF_{metal} , I^{metal}_{geo} , CF_{metal} and PLI (Table-4 and Fig. 4), the following inferences can be drawn: (i) the average concentrations of some heavy metals (Co, Cr, Cu, Mn, V and Zn) in the Kolleru Lake sediments are higher than the background values, (ii) these results are also consistent with the dissolved heavy metal contents in water samples of the Kolleru Lake that were collected from the same location along with the sediments, (iii) the average contents of all other heavy metals are low relative to the background and they do not pose any threat to the lake ecosystem (see Table-4), (iv) the choice of background is extremely important to assess the extent of metal pollution in the sediment, and (v) the use of ACNK diagram in conjunction with concentrations of average sediments (Taylor & McLennan, 2001) and/or world surface rock average (Martin & Meybeck, 1979) as backgrounds are found to yield the most consistent results pertaining to enrichment factors, geoaccumulation

indices, contamination factors for the Kolleru Lake sediments.

The bioavailability of any toxic trace metal present in the sediment depends upon the chemical form and concentration of the metal (Kwon et al., 2001). Toxic trace metals in sediments with higher labile fractions are potential sources for mobility and bioavailability in the aquatic ecosystems (Mohiuddin et al., 2010). Chandra Sekhar et al. (2003) carried out a study on the bioaccumulation of sediment-bound heavy metals by some edible fishes of the Kolleru Lake. These researchers concluded that large fractions of Zn, Cd and Cu were associated with mobile fraction of sediment and therefore these metals showed greater bioaccumulation in fish. They further suggested that the fishes of Kolleru Lake are contaminated and are not suitable for human consumption.

Based on the results obtained in our previous study (Das Sharma & Sujatha, 2016), several measures were recommended for the environmental rehabilitation of the lake and its surroundings. One of the suggestions was to clean up of lake bed at regular intervals in order to prevent piling up of huge sediments and residues that drain into the lake. It was envisaged that such periodic cleaning measure would ensure deep lake bed and would also facilitate recharge of large volumes of freshwater during the rainy seasons, besides ensuring free flow of water in the lake. However, based on the present study, where it has been demonstrated that the lake sediments are minor to moderately contaminated with certain toxic metals such as Co, Cr, Cu, V and Zn two relevant questions arise. Firstly, where to dump these contaminated sediments? Secondly, how to get rid of the toxic metals from the sediments? As regard to the first question, the sediments can be dumped at some designated site, chosen in such a way that there is perfect balance amongst the cost of transport, proximity of

the site to the lake and ecology of the area. Coming to the second question, it is envisaged that since the degree of contamination due to presence of toxic metals like Co, Cr, Cu, V and Zn range from minor to moderate (see Table-4), phytoremediation of these heavy metals appears to be the most suitable option for several reasons as highlighted below.

- (i) Phytoremediation is an appropriate technique if the level of contamination is low;
- (ii) More than 400 plant species are presently known (Lasat, 2000; Schmidt, 2003) which have the natural tendency to accumulate very high metal concentrations;
- (iii) It is an extremely cost-effective method (Garbisu & Alkorta, 2001) compared to the other remedial processes;
- (iv) The method poses little imminent risk to human health or the environment;
- (v) It offers a green and environmentally friendly technique and hence likely to be accepted by everyone.

With the above-cited advantages of phytoremediation, it is also significant to note that the method is simple, and it uses living green plants for removal, degradation or containment of contaminants in soils, sludges, sediments, surface water and groundwater (Cunningham & Ow, 1996; Helmisaari et al., 2007). As a follow-up action of the phytoremediation process, it is recommended that the domestic sewage and the industrial effluents originating from various anthropogenic activities (sugar factories, distilleries, tanneries, agrochemical establishments, paper mills and rice mills etc.) need to be treated on a regular basis with proper monitoring so that the heavy metal pollution of the lake sediments and waters is avoided to a great extent.

CONCLUSIONS

The results obtained in this study indicate

that the lake water is contaminated to variable degrees with certain heavy metals. These metals are chromium, copper, manganese and zinc. An assessment of sediment quality through several pollution indices (e. g., enrichment factor, geoaccumulation index, contamination factor and pollution load index) also corroborate the contamination of lake sediments with heavy metals like Co, Cr, Cu, Mn, V and Zn. The observed heavy metal contents in lake water and sediment have increased with time when compared to the one and half decade old data. Therefore these metals owe their origin to diverse nature of industrial effluents that enter the lake through different channels, besides domestic sewage and agricultural runoff. The extent of sediment contamination, however, is in its initial stage ranging from minor to moderate. These results are expected to be of significant help to future researchers in order to evaluate the relative availability of these elements to biological communities and their impact on reactions and transformations. It is satisfying that the Government of Andhra Pradesh has been trying its best for the restoration of this fragile ecosystem using various measures. However, concerted efforts are needed on local and regional scales to promote mass awareness program and save the lake from its impending disaster. This should be followed by restoration measures where research organizations, industries, government and private agencies can join and work together in harmony.

APPENDIX-A

Characterization of the extent of anthropogenic contamination is often carried out by estimating enrichment factor (*EF*), which provides the enrichment ratio. The *EF* values can be estimated using the equation

$$EF = \frac{[(C_{metal}/C_x)_{sample}]}{[(C_{metal}/C_x)_{background}]}$$

where C_{metal} and C_x represent the concentrations of studied metal and any immobile element, respectively. Several

immobile elements (both major and trace) have been considered by different researchers. The major immobile elements include aluminium (Chatterjee et al., 2007), iron and titanium (Zhang et al., 2007). Immobile trace metals like Sc and Zr have also been used in the estimation of *EF* (e. g., Blaser et al., 2000). In the present study, aluminium is chosen as immobile metal for the estimation of *EF*.

The geoaccumulation index (I_{geo}) was defined by Müller (1969) to determine the metal contamination in sediments and is expressed as $I_{geo} = \log_2[(C_n)/(1.5 \times B_n)]$, where C_n is the measured concentration in the sample for the metal n and B_n is the background value (in shale) for the metal n . The factor 1.5 is used to account for the possible variations of background data originating from lithological variations.

Contamination factor is estimated to ascertain the level of contamination (Häkanson, 1980) and is expressed as $CF = (\text{Conc. of metal X})_{\text{sample}}/(\text{Conc. of metal X})_{\text{background}}$

The pollution load index was proposed by Tomlinson et al. (1980). The pollution load index is calculated according to the equation $PLI = [CF_1 \times CF_2 \times CF_3 \dots \times CF_n]^{1/n}$, where n is the number of analyzed metals and CF_n is the contamination factor of metal n (see equation on *CF*). In other words, *PLI* is the n th root of the product of n contamination factors (*CF* values).

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