

Analysis of heavy metal toxicity in the sediments of deteriorating Pallikaranai Marshland, Chennai, Tamilnadu

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ABSTRACT

Pallikaranai Marshland is a wetland ecosystem in Chennai city with multiple ecological functions. However, due to severe anthropogenic disturbances, the natural ecosystem has undergone several changes in the past few decades. The high concentrations of probably toxic heavy metals like Cd, Cr, Cu, Ni, and Hg beside trace metals like Fe and Mn measured in surface sediment of Pallikaranai Marshland reveal the extent of the damage. The heavy metals in sediments were present in the following order, Fe > Mn > Cr > Cu > Ni > Hg > Cd. The increase in garbage dump area adjacent to the marshland has caused some amount of leachate infusion into the marsh which might probably be a reason to be attributed. Sediment samples were analyzed for significant metals like Cd, Cr, Fe, Mn, Cu, Ni, and Hg along with sediment texture and organic carbon. The main sources of nonpoint supply of pollution contaminating the marshland are the disposal of untreated sewage into the marsh, leachate infusion, and urban run-off. The presence of significant metals has been increasing staggeringly since past study levels. Results indicate that the potential damage can cause bioaccumulation in benthic, soil-dwelling, and aquatic species. The conclusion arrived is based on indices such as enrichment factor, contamination factor, and geoaccumulation index Igeo. These indices are helpful to assess the degree of pollution in sediments. The Enrichment factor results show severe to extremely severe pollution. The contamination factor (CF) shows CF > 1 indicating metal enrichment particularly for Cu, Ni, and Hg and the necessity for close monitoring of the environment and assessment of the marshland to enhance and maintain its aesthetic value.

Keywords: Heavy metals; Pallikaranai Marshland; Sediment; Pollution

1. Introduction

Heavy metals are harmful toxic pollutants that have been on the rise in environments around the world. Their increasing levels cause severe threats to all different forms of life. For instance, in an aquatic environment, heavy metal contamination is a cause of concern given its toxicity and accumulation in aquatic habitats [1]. The behavior of metals in natural water is said to be a function of the substrate sediment composition, the suspended sediment composition, and its water chemistry [2]. These toxic metals participate in numerous bio-geochemical mechanisms by having vital mobilities that affect the ecosystems through bioaccumulation and biomagnification processes and are probably lethal for bio-network. Heavy metals additionally might stay within the atmosphere for years due to their non-biodegradable nature and pose long-term risks to life even after sources of these metal pollution are removed [3]. The fate of these metals largely depends on the biota. The accumulation nature of such toxic metals

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even in traces might have undesirable effects on the physiology, reproductive behavior, and overall health of the organisms that are present in such ecosystems. Sediments are heterogeneous assemblages of natural and organic elements and therefore exhibit a large degree of variability in terms of composition and chemical properties [4]. Metal concentration in sediments shows high levels of anthropogenic influence. The sediment that has a high mixture of contaminants will provide information concerning the activities happening around the region. Sediment contaminants, if bioavailable, will be bioaccumulated by aquatic organisms presumably giving rise to acute or chronic health effects [5]. Humans promote heavy metal pollution through activities like mining, smelting, transportation, military operations, industrial productions, and the application of metal contained pesticides and fertilizers in agriculture. Heavy metals are deposited onto sediment surfaces and prevented from further movement through adsorption, coagulation, flocculation processes, and by forming metal complexes [6]. Since heavy metals are non-biodegradable, they are considered hazardous and have been proved to be carcinogenic in many cases. Heavy metal pollution in an aquatic ecosystem should be of major concern to any government making policies. In water, metal mobility is increased considerably due to soluble inorganic and organic ligands, which are molecules that bind to metals to form metal complexes [7]. The soluble metal concentration in pore water is generally higher than predicted due to the partitioning between metal binding phases and pore waters [8]. Heavy metals that are released into any aquatic environment are finally incorporated into the aquatic sediments, therefore causing the accumulation of these metals to several degrees in the organisms living in these sediments [9]. Heavy metal distribution and abundance in the sediments are important to assess the influence of the high anthropogenic intervention on natural ecosystems. The marsh ecosystem must be protected and preserved for long-term biodiversity and the future generation. Suggested treatment includes phytoremediation through native species of plants and shrubs. Plants that are proved to be effective in the removal of heavy metals include *Typha latifolia* (Bulrush) [10] and *Phragmites australis* (Common reed) [11] which are native to the marshland. This method is profoundly useful if done on a large scale as the roots of such plants tend to absorb heavy metal pollution as well as provide shelter to the nesting marshland birds. Dredging of canals that connect the marshland to increase the water flow which will reduce the movement of metals into the soil as well as encourage the growth of native species of plants and the dump yard extension into the marshland should be stopped immediately. An elaborate study on the potential changes in the environment includes the environmental risk assessment study to examine the extent of the damage created to this sensitive ecosystem.

2. Study area

Pallikaranai Marshland is situated adjacent to the Bay of Bengal in the southern part of the city of Chennai with a total area of 80 km2 . The marshland has shrunk from 5,500 to 600 ha in the past few decades. The Velachery–Tambaram road on the west, the Old Mahabalipuram Road (OMR) on the east, Perungudi on the north, and Pallikaranai on the south are the current boundaries. Pallikaranai Marshland is located at 12.9377° N, 80.2153° E. The region is laden with thick silty loam soil and brackish sediment. There is some amount of water always present in the marshland. The entire region is made up of the weathered Charnokite bed [12] and covered with a layer of alluvial soil of varying thickness [13]. The surface sediments in Pallikaranai Marsh are generally medium to fine in texture, and the estimated sand content varied from 8.6% to 41.2%, with an average value of 25.2%. The recharge potential of the region is immense as the region is proximate to the south Chennai aquifer [14].

2.1. Heavy metal contamination

The study area has a garbage disposal area in its vicinity. Decomposition of organic matter and the matured compost in the landfill produces a dark melanized alkaline extract oozing from the heaps to produce leachates [15]. The leachate was found to have heavy metals by the following studies and thus confirms the origin of heavy metal contamination in this region. Esakku et al. [16] studied leachate characteristics such as pH, EC, TDS, COD, BOD, DOC, ammonia-N, chloride, and heavy metals (Cu, Cr, Ni, Zn, Pb, and Cd) profuse leachate from the dumpsite entering the mainstream wetland may have caused high values in pollution level as the leachate samples from Perungudi dumpsite tested for Leachate Pollution Index (LPI) indicated values higher than 7.5. After the monsoon of December 2002, they found a gradual increase in pollutant concentrations that was evident and have attributed to the enhanced leaching of pollutants from the fresh wastes dumped at the top layer during the initial precipitation followed by the dilution effects of rainfall. Another leachate characterization was done by Parameswari and Mudgal in the year 2014 and reported the order in which they were present at the source level as Pb > Fe > Cr > Zn > Cd and that the occurrence of alternate sandy and clayey layers facilitate the movement of the contaminants as there is no or very less space for the generated leachate. Eventually, the leachate generated has to move out of the dumpsite, with a lateral movement beyond the dumpsite [17].

The major effect of toxicity of the receiving water is seen in the decline of aquatic bird species which use the marshland as foraging and nesting ground. However, due to the loss of food sources, the number of birds visiting the marshland has seen a decline in the past few years. The next effect includes a threat of bioaccumulation of these metals in the living organisms affecting the food web and finally entering humans through metal movement in the soil reaching groundwater aquifers and polluting the same and creating adverse effects such as neurotoxicity due to metals and constitute a major threat to this marshland ecosystem.

3. Methods

3.1. Sample collection procedure and analysis

A total of six samples were collected during the preand post-monsoon seasons in the Pallikaranai Marshland. The sampling locations included places close to two bird

watching towers, adjacent to Perungudi garbage dump yard, near Velachery metro railway station, the backside of NIOT (National Institute of Ocean Technology), and the beginning of Pallikaranai Radial road. Samples were collected during alternate months of pre-monsoon and post-monsoon seasons. During pre-monsoon (January–July), the water level was very low and during the post-monsoon season (August–December); the marsh environment was not suitable for the frequent collection of samples. At each point, composite sediment samples were collected from the center of the marsh for total metal analysis. The sediments were taken at a depth of 0–5 cm from the surface. Grab samples of mass approximately 200 g were collected at each point. Sampling sites were selected based on the point source being the Perungudi garbage dump site. Samples were collected in clean, washed polythene air-tight bottles and stored at −4°C immediately. Sediment analysis was performed as prescribed by the EPA. Analytical grade reagents and deionized water were used during analysis to maintain the quality of analytical procedures to produce reliable data. The sediment samples were oven-dried and the constant weight was measured. The dried samples were then homogenized by grinding in mortar and pestle and stored before analytical procedures were conducted. About 0.25 g of sample from each sampling site was digested by the Aqua Regia method using $HNO₃$ and HCl in a 3:1 v/v ratio, respectively. Each sample was digested until it turned colourless. The solution was made up to 50 mL with ultrapure distilled water. Certified reference materials (CRMs) were used to establish the accuracy of the analytical methods and, to assure quality control. Statistical

analysis was performed using the Mat Lab software and correlation studies were done using the Microsoft excel.

3.2. Instrumental Analysis

Heavy metals in the water samples were analyzed by using a Flame Atomic Absorption Spectrometer (Agilent 200 series AA). The spectrometer was calibrated against a series of solutions of known standards to give a calibration graph. The standard solutions were per NIST (National Institute of Standards and Technology) guidelines to provide traceability. The calibration curves with $R^2 > 0.995$ were accepted for concentration calculation. For each experiment, a run included blank, certified reference materials (CRM) and samples were analyzed in duplicate to eliminate any batch-specific error. A comparison was made with the certified values, which in both cases confirmed that the sample preparation and instrumentation conditions provided good levels of accuracy and precision.

3.3. Sediment texture, chemical properties and total metals in sediments

The sediment types silty clay loam, clay loam, and sandy clay loam were present in tested samples and the estimated sand content varied from 8.6% to 41.2% with an average value of 25.2%. Percentage silt and percentage clay are given in Table 2. Mostly silty clay soil is present and results reveal that it is predominantly found in the central part of marshland. The entire marsh is also covered with mud along with alluvial black topsoil. Most of the regions

Table 1

Analytical methods used to determine soil quality at Pallikaranai Marshland

Table 2

Textural characters and sediment type in surface sediments of Pallikaranai Marshland

were dry and arid due to a lack of freshwater inflow during the pre-monsoon season. However, in the post-monsoon seasons, the soil was clogged with water pockets in the outer regions of the marsh indicating clay soil, where the vegetation was absent.

3.4. Chemical properties

Organic carbon (OC) in soil has a significant effect on the chemical and physical characteristics of soil and it is one of the essential components of soil quality assessment [18]. The correlation of organic carbon in sediments and soil with organic contaminants can be used as a tool to estimate the level of contamination toxicity [19]. Organic carbon has a crucial influence on the migration

Chemical properties of sediments at Pallikaranai Marshland

and transformation of pollutants in soils. Studies show that soil OC can combine with pollutants such as heavy metals and organic pollutants, changing their physical and chemical properties [20]. The OC of the analyzed samples varied from 1.1% to 4.2% in the pre-monsoon season with an average of 2.75% and 5.5% to 8.4% with an average of 6.7% in the post-monsoon season. Rich vegetation and consequent decomposition of plants and animals could be the main source behind increased organic carbon content in the post-monsoon season.

3.5. Heavy metals

Heavy metal concentrations show significant variations from one sampling site to the other. Table 4 represents

Table 4

Table 3

Heavy metal concentrations in the sediments of Pallikaranai Marshland (mg·kg)

Sampling sites	Cd		Cr		Fe		Mn	
	Pre-	Post-	Pre-	Post-	Pre-	Post-	Pre-	Post-
	monsoon	monsoon	monsoon	monsoon	monsoon	monsoon	monsoon	monsoon
	0.5	BQL	31.9	55.8	19,100	11,700	362	136
2	0.7	BQL	12.9	30.9	9,500	20,900	151	239
3	0.8	BQL	4.98	6.9	8,900	10,600	247	253
$\overline{4}$	0.5	BQL	2.49	21.9	5,800	14,200	160	243
5	0.6	BQL	23.3	23.5	12,400	5,100	360	235
6	BQL	BQL	25.6	42.8	15,700	23,000	455	362
$Mean \pm SD$			16.86 ± 11.89	30.3 ± 17.15	$11,900 \pm 4868.26$	$14,250 \pm 6698.28$	289.17 ± 122.78	244.67 ± 71.80

BQL – Below quantification limit

the values of heavy metal concentration in Pallikaranai Marshland at six different locations around the marsh. The concentration graphs of heavy metals Fe and Mn are represented in Fig. 2 and the concentration graphs of heavy metals Cd, Cr, Cu, Ni, and Hg is represented in Fig. 3. The corresponding distribution diagrams are presented in Fig. 5.

4. Metal assessment in sediment

4.1. Pollution load index (PLI) and contamination factor (CF)

The pollution load index (PLI) gives a single value that represents the number of times by which the heavy metal concentrations in the sediments are higher than the background concentration, and gives an interpretation of an overall level of heavy metal toxicity in a particular sample [21]. It is calculated as Eq (1) the *n*th root of the number of multiplied contamination factor (CF).

$$
PLI = (CF1 \times CF2 \times CF3 \times \dots \times CFn) \frac{1}{n}
$$
 (1)

The contamination factor (CF) is the ratio of the content of each metal to the background values (background shale values from the upper continental crustal average, CCA) in sediment. CF is calculated from the ratio of the sediment metal content in a sediment sample to the normal concentration levels; CF values indicate the enrichment of metals in the sediment. CF_{metals} is the metal value in polluted sediment/background value of metal. PLI values in sampling site 2 during pre-monsoon indicate increased pollution and at sampling sites 3 and 4, PLI values indicate increased pollution during both pre- and post-monsoon seasons. Sampling sites 3 and 4 are adjacent to the garbage dump and this may attribute the reason for the increase in pollution level. CF > 1 indicates contamination with concern to that metal and CF < 1 indicates there is no metal enrichment in the sediments.

4.2. Geoaccumulation index (Igeo)

Geoaccumulation index (Igeo) introduced by Muller [22] is used to determine the metal contamination in sediments with the following equation.

Fig. 2. Heavy metals Fe and Mn in sediments from Sampling sites 1–6 in pre-monsoon and post-monsoon season.

Fig. 1. Map of study area of Pallikaranai Marshland (Raj P.P., et al. 2010).

Fig. 3. Heavy metals Cd, Cr, Cu, Ni and Hg in sediments from Sampling sites 1–6 in pre-monsoon and post-monsoon season.

$$
Igeo = \log_2 \frac{C_n}{1.5 \times B_n}
$$
 (2)

where C_{μ} is the concentration of the metal in the sample, B_x is the background value for the metal. Factor 1.5 is used because of geological variations in background values. Igeo calculated using the continental crustal average values CCA is given by Wedephol [23]. The geoaccumulation index is used to refer to the extent of metal pollution [24]. Geoaccumulation index represents seven grades or classes Igeo1-7. Igeo 6 indicates a 100 fold enrichment in pollution above background values. The geoaccumulation index (Igeo) associated with a qualitative scale of pollution intensity, is proposed by Muller. Table 7 provides the Igeo values for the sediment samples from six locations of Pallikaranai Marshland during both pre- and post-monsoon seasons.

4.3. The Enrichment Factor

The enrichment factor (EF) is used to calculate the heavy metal contamination in sediments and it is calculated using the following equation.

 $EF = \frac{C_{\text{SAMPLE}}/A1}{C_{\text{CREIST}}/A1}$ SAMPLE^{/111}SAMPLE CRUST¹¹CRUST $=\frac{C_{\text{SAMPLE}}/A1_{\text{SAMPLE}}}{C_{\text{CUTE}}/A1_{\text{CPIIST}}}$ (3)

 C_{SAMPLE} and C_{CRUST} are the concentration of the metal in the sample and the concentration of the metal in the continental crust, respectively.

 Al_{SAMPIE} and Al_{CRIST} are the Al content in the sample and the Al content in the continental crust, respectively. Al is used as a reference element. Since Al is an element that is particularly stable in soil because of the absence of vertical mobility and lesser degradation, it is chosen as the reference element [25]. Aluminium is also a major constituent of clay minerals and is known as a very conservative element used by many scientists [26].

Concerning EF, mercury contamination is extremely severe in sites 1, 2, and 6. respectively. Mercury contamination should be immediately taken into concern as it is a potential neurotoxicant that may harm the biota of the

Fig. 4. PLI values of heavy metals in sediment of Pallikaranai Marshland in Sampling sites 1–6 (Md. S. Islam, 2015).

Fig. 5. Geoaccumulation index (Igeo) value of heavy metals in sediment of Pallikaranai Marshland.

able 5

Table 6 Element concentrations in ppm in the upper continental crust (Wedephol, 1995)

marshland. The contamination in the sediments of these sampling sites may derive these contaminants mainly from the dumpsite. Cr and Mn show minor to moderate enrichment in the pre-monsoon season while Cr shows severe enrichment in the post-monsoon season in the sampling sites 1, 5, and 6. Fe shows moderate to moderately severe enrichment in the pre- and post-monsoon seasons, respectively. Cu shows severe enrichment in the pre-monsoon season while very severe enrichment in the post-monsoon season. Ni shows severe enrichment in both seasons.

5. Results

5.1. Total metal concentration in sediment

Textural characters of the sediments and chemical properties are presented in Tables 2 and 3, respectively. The concentration of each metal present in the pre- and post-monsoon seasons are represented in Table 4. The average pH is 7.2 which is neutral. The organic carbon content showed only a slight variation between the two seasons. Organic carbon content ranged from 1.1% to 4.2% in the pre-monsoon season and 5.5% to 8.4% during the post-monsoon season. The range of metal concentrations differed widely in the sampling sites. Factors such as proximity to the dump, absence of water inlet source, non-point sources of pollution, and geomorphological land setup may be the reasons for such variations. The concentration of heavy metals in sites 1 and 6 was much higher than other sites since they are close to the adjacent garbage dumping site. Metal concentrations were more or less similar in both pre- and post-monsoon seasons owing to the sediment deposits of heavy metals as a result of constant leachate infusion from the adjacent dump. Chromium, iron, and copper are found to be higher in the post-monsoon season and the reason may be due to the deposits brought down by the floodwaters from the city. Metal concentrations in sediment were higher in post-monsoon than pre-monsoon season. The average concentration of heavy metals in sediments was in the following order Fe > Mn > Cr > Cu > Ni > Hg > Cd. Iron concentration is relatively higher in all sediment at all locations and in both seasons. High iron content may indicate redox conditions in surface waters due to which the sediment has accumulated iron in such high levels. Manganese is present in a considerably

 ≤ Igeo ≤ 2 Moderately polluted ≤ Igeo ≤ 3 Moderate to strongly polluted ≤ Igeo ≤ 4 Strongly polluted ≤ Igeo ≤ 5 Strongly to extremely polluted Igeo ≥ 5 Extremely polluted

Igeo ≤ 0 Unpolluted

Table 9

Enrichment factor values (Zhang and Liu, 2002)

Geoaccumulation Index and categories of sediment quality

0 ≤ Igeo ≤ 1 Unpolluted to moderately polluted

Table 10

Enrichment Factor values of sediments of Pallikaranai Marshland for Pre-Monsoon season (Zhang and Liu, 2002)

BQL – Below quantification limit

Table 11

Enrichment factor values of sediments of Pallikaranai Marshland for post-monsoon season (Zhang and Liu, 2002)

	S. no. Cd Cr Fe Mn Cu Ni				Hg
1.	BOL 20.23 4.81 3.28 36.04 13.79				88.6
2.	BOL 9.37 7.18 4.81 23.22 9.18 37.32				
3.	BOL 2.46 4.29 5.996 22.97 10.01				20.74
4.	BOL 4.85 3.56 3.57 12.62 8.08				4.70
5.	BOL 17.33	4.26 11.51	18.05	7.09	10.6
6.	BOL 12.80 7.79 7.19		16.69 8.95		34.01

BQL – Below quantification limit

high concentration and chromium concentration is also considerably high. An increased amount of tannery wastes may have attributed to the high chromium enrichment. Total cadmium content was relatively less all through the sites. But has increased when compared to the previous study results. Variation in water levels in the marsh and

Table 8

(Muller, 1979)

Fig. 6. Continued

Fig. 6. Distribution of heavy metals in Pallikaranai Marshland (Jayaprakash. M et al., 2010).

restricted water flow may have increased in cadmium concentration. In general, the metal concentrations have $enriched about 3-10 times compared to the previous studies.$

6. Conclusions

The present study reveals that concentrations of some heavy metals Cr, Cu, and Ni are higher than safer limits. The presence of heavy metal Hg is to be considered dangerous and its increase in concentration than previous study levels is alarming. These metals in higher levels in the sediment of the Pallikaranai Marshland indicate severe anthropogenic disturbance and elevated levels of pollution. The contamination factor (CF), Geoaccumulation index Igeo*,*

and the enrichment factor (EF) reveal that the sediments are contaminated considerably which may lead to bioaccumulation by organisms. This can create an adverse effect on the wetland ecosystem if left unchecked. Appropriate remediation techniques suggested may be used to reclaim the land for wildlife usage, especially the winged visitors of Pallikaranai. Mitigation strategies that could reduce the high levels of contamination include phytoremediation, dredging of canals, protection of the marshland from illegal activities such as letting off untreated sewage, stop dumping of waste in the adjacent dumpsite with alternate solid waste technology management and, reclaiming the lost marshland area due to encroachment. Several migratory birds use this marshland as their winter refuge every

year. Hence it is suggested that necessary steps are taken to reclaim the land used as a dumping site, to prevent further damage. Restoration projects have been undertaken by the Conservation Authority of Pallikaranai Marshland (CAPML) in recent days which is welcoming.

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