Distribution and source identification of heavy metal concentration in Chilika Lake, Odisha India: an assessment over salinity gradient

Satabdi Banerjee, Arnab Pramanik, Sanghamitra Sengupta*, Dhrubajyoti Chattopadhyay* and Maitree Bhattacharyya*

Department of Biochemistry, University of Calcutta, 35, Ballygunge Circular Road, Kolkata 700 019, India

Extent of heavy metal contamination in Chilika Lake was assessed to conserve the eco-dynamics of the lake. Distribution of heavy metals, cations and total organic carbon were analysed in the sediment samples collected from three sites that represent a salinity gradient. Analysis of environmental indices, viz. enrichment and contamination factors, geo-accumulation ($I_{geo}$) and pollution load indices showed that the concentrations of Cd and Hg were significantly high with geo-accumulation index being greater than one in all three stations. Levels of Pb and Co were also found to be elevated with $I_{geo}$ value of 0.025 and 0.048 respectively. Among the three sampling stations, Kaluparaghat witnessed the highest pollution load index followed by Balugaon and Rambhartia. Principal component analysis revealed that the high-salinity zone was associated with high concentration of Hg and decreased concentration of other toxic metals, indicating that source of pollution of Hg is different from that of other metals.

Keywords: Heavy metal, lagoon, pollution load index, salinity.

COASTAL lagoons are superficial water bodies attached to the ocean through one or more inlets. Most coastal lagoons act as a sink of heavy metals deposited by the freshwater tributaries. Sediments are the basic repositories of different metals. The quality of soil sediment depends on a number of factors, including pH, conductivity, organic carbon and heavy metal content. Contamination of toxic and nondegradable heavy metals in the environment represents a major ecological risk. Physicochemical adsorption, biological uptake and physical accumulation are the key mechanisms influencing concentrations of heavy metals in the sediments. Various anthropogenic activities of the expanding population also exert a thrust on the soil environment. To conserve the eco-dynamics of the lake, an assessment of the geochemical distribution and the pattern of accumulation of heavy metals in the sediment should be considered a priority.

Lagoons from all over the world have been reported to be contaminated by heavy metals. Various lakes and lagoons from Egypt, Portugal, Indonesia and Mexico have been severely affected. In most of the cases, the pollution sources resulted due to anthropogenic activity. Pulicat Lake in India, a Ramsar Convention site, is also facing the problem of heavy metal pollution due to over-exploitation, mismanagement and improperly treated industrial effluents from more than 25 industries. These industries continuously discharge waste water into the North Chennai coastal region, which is a great challenge for ecosystem balance. Like Pulicat Lake, Ennore estuary is also contaminated due to heavy metal pollution.

Rapid economic development in the areas adjoining Chilika Lake has resulted in tremendous pressure on the associated environment. In an estuary like Chilika, where the biodiversity is significantly high, any small level of pollution may exert a major impact on the ecosystem. Here we present an analysis of heavy metal concentration in three regions of the lake, that represent a salinity gradient along the length of the estuary. Effort has also been made to provide a comparative account of heavy metal contents of Chilika Lake to gauge any anthropogenic impact over the past 20 years.

Methodology

Study area

Chilika lagoon (19°28′–19°54′N and 85°05′–85°28′E), the largest brackish water lagoon in Asia, is one of the few Ramsar site wetlands of international importance. Unlike other coastal lagoons, the salinity of Chilika Lake fluctuates over a high range resulting in variations in physical and chemical parameters of lagoon water.

In this study, water and sediment samples were collected from three sampling stations representing three salinity zones. Freshwater emerging from River Mahanadi flows into the Chilika Lake and mixes with saline water...
entering from the Bay of Bengal. This generates three zones with varying salinity such as freshwater, brackish water and saline water environments. The first station, Rambhartia (N1, 19°39'25"N; 85°27'43"E) is situated near the sea mouth. The other two stations, Balugaon (N2, 19°44'37"N; 85°12'59"E) and Kaluparaghat (N3, 19°50'41"N; 85°24'19"E) are located in the brackish water and freshwater zones respectively. Stations N2 and N3 lie in the central and northern sectors of the lake. Thus these three sampling sites were selected from different geographical zones with salinity gradient and represent three sectors of the lake (Figure 1).

**Sediment sampling**

Sampling locations were selected according to salinity variation. Surface sediments were collected from the three stations (N1–N3) before monsoon in June 2014. Sampling was done in triplicate from each sampling station at a depth of 30 cm with a grab sampler. Samples were then transferred to sterilized plastic bags and brought to the laboratory. For metal analysis, the soil samples were oven-dried at 50–60°C. The dried soil samples were disaggregated with mortar and pestle and passed through a 100 mesh nylon sieve (0.25 mm) before chemical treatment. pH and conductivity (mS cm⁻¹) were recorded in all water and sediment samples.

**Sediment analysis**

Analysis of heavy metals was performed with each sample of 400 mg dried sediment. Each sample was digested with a mixture of HNO₃, HCl and H₂O₂ in the ratio of 4:2:2 in an Anton Paar MULTIWA 300 microwave. The reaction vessels were cleaned using 5% HNO₃, heated for 30 min at 1000 W. The sample was then filtered and the volume made up to 50 ml. Heavy metal concentrations were measured using an inductively coupled plasma optical emission spectrometer (ICP-OES, Thermo Scientific iCAP 6000 series). Calibration standard solutions and internal standards were prepared by successive dilution of a high-purity ICP multi-element standard solution supplied by Merck Millipore (Darmstadt, Germany). The concentrations of internal standards were 0.02, 0.1, 0.2, 0.5 and 1 ppm. The wavelength of Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni, Pb and Zn was 226.5, 228.6, 267.7, 324.7, 259.9, 184.9, 257.6, 231.6, 220.3 and 213.8 nm respectively. Calibration was performed with a blank and standard solution samples. For accuracy of the method, NCS ZC 73006 Certified Reference Material was used and for digestion of this reference material, an acid mixture (3 ml HNO₃, ultrapure 60%, 2 ml HF 40%) was added to 0.1 g of the sample. For total organic carbon analysis, 300 mg of each sample was digested with 250 μl H₂SO₄ (5%) and 250 μl HCl (5%) at 100°C for...
1 h. The sample was then filtered and soil was scraped and dried. Final weight was measured and analysed in a TOC analyzer (O.I. Analytical Solids Model 1030). Organic matter was derived from total organic carbon according to Walkley–Black method.

Assessment of sediment contamination and ecological risk parameters

Degree of heavy metal contamination in Chilika lagoon was analysed using different indices such as contamination factor (CF), enrichment factor (EF)\(^{14}\), geoaccumulation index \((I_{\text{geo}})^{15}\) and pollution load index (PLI)\(^{14}\). The following equations were used

\[
EF = \left( \frac{C_x}{C_{Al}} \right) \text{sample}/\left( \frac{C_x}{C_{Al}} \right) \text{background},
\]

where \(C_x\) and \(C_{Al}\) denote the concentration of metal \(X\) and aluminium in the sample and the upper crust continent respectively. In this study iron was taken as the background value because natural sources vastly dominate its input\(^{15}\).

\[
I_{\text{geo}} = \log_2 \left( \frac{C_n}{1.5B_n} \right),
\]

where \(C_n\) is the measured concentration of metal \(n\) and \(B_n\) is the background concentration of metal \(n\) (ref. 15).

\[
\text{CF} = \frac{C_m}{C_{m\text{background}}},
\]

where \(C_m\) sample is the concentration of a given metal in river sediment and \(C_{m\text{background}}\) is value of the metal equal to the world surface rock average\(^{14}\).

\[
\text{PLI} = \left( CF_1 \times CF_2 \times CF_3 \times \ldots \times CF_n \right)^{1/n},
\]

where \(n\) is the number of metals\(^{14}\).

The world rock average values were chosen for the reference background values\(^{14}\).

Statistical analysis

Each sample was analysed in triplicate and comparisons were performed using the mean values. Pearson correlation matrix and cluster analysis were performed using SPSS for Windows Version 20.0 (SPSS Inc. Chicago, USA). To analyse the sources of heavy metals and their correlation, principal component analysis (PCA) was carried out using Past Version 3.06c software\(^ {16}\).

Results and discussion

Physical characteristics

Sediment structure of Chilika lake varies from sea mouth to freshwater zones. Sediment is mainly composed of clayey silt. Overall, the sediment structures of the three stations are similar, except for N2 which was enriched with fine sand. pH values of water samples collected from N1, N2 and N3 were 7.9, 8.1 and 7.5 respectively, whereas the corresponding values measured from soil sediments were 6.95, 7.92 and 6.38 respectively. Salinity followed a decreasing trend across the three sampling stations, with the indices 36, 21.4 and 4.66 PSU in N1, N2 and N3 respectively. The temperatures of water samples were 27°C, 28.8°C and 28.5°C in N1, N2 and N3 respectively. The highest level of salinity of the sediment was recorded in N1 (4.6 PSU) and negligible (<0.1 PSU) in N3, which was located in the freshwater zone. The total organic carbon was 4.03% at Rambhartia, 11.87% at Balugaon and 16.36% at Kaluparaghat. The organic matter was 6.94% at Rambhartia, 20.46% at Balugaon and 28.90% at Kaluparaghat.

Spatial distribution of heavy metals

Among the 20 metals analysed, Fe showed significantly high concentration at all three sampling sites with the highest concentration at N3 (Table 1). Among the three stations, CF and EF values for Pb were higher at N3 (Table 2). Pb concentration displayed a significant positive correlation with that of Mn, indicating that these two metals might have originated from common sources (Table 3). Estimated \(I_{\text{geo}}\) trend in the three stations indicated that N1 and N2 might be categorized as unpolluted while N3 could be considered as an unpolluted to moderately polluted zone according to Muller’s classification\(^ {15}\) (Table 2). The concentration of Pb estimated by this study was higher than the world rock average at N3. The elevated level of Pb at N3, located in the freshwater zone might be attributed to the municipal wastes carried along with the sediments of river Mahanadi.

Total concentration of Cd was higher than world rock average at all three stations (2.11–3.45 mg kg\(^{-1}\)) (Table 1) and exhibited significant positive correlation with that of organic carbon, Fe, Ba and Cu (Table 3). CF and EF values of Cd ranged between 10.55 and 17.25 as well as 1.10 and 1.18 respectively, and according to Muller’s classification all three stations were categorized as moderate to strongly polluted area for Cd contamination (Table 2). N3 was recorded to harbour the highest Cd enrichment, with \(I_{\text{geo}}\) of 3.52 (Table 2). Cr concentration at N3 (87.77 mg/kg) and N2 (86.29 mg/kg) was higher than the world rock average concentration (Table 1). The estimated environmental indices in the present study indicate that N2 and N3 should be categorized as moderately contaminated (Table 2). Cr contamination in freshwater zone might arise due to cement dust, contaminated landfill and effluent from chemical plants brought by the River Mahanadi.

EF and \(I_{\text{geo}}\) values for Ni were <1 for all stations, (Table 2) and total concentration of Ni showed a statistically
significant negative correlation with Hg (Table 3). In this study, very high concentration of Co was observed at all three stations, which was even higher than the world rock average (Table 1). However, \( I_{geo} \) and EF values of cobalt were >1 only at N3, indicating station N3 to be more polluted with cobalt compared to the other two stations (Tables 1 and 2). Co showed strong positive correlation with Cd, Be and Cu (Table 3). CF, EF and the negative \( I_{geo} \) values for Zn indicate that Chilika is not polluted with Zn (Table 2). The total concentration of Cu and Mn ranged from 18.26 to 40.66 and 183.37 to 1214.49 mg kg\(^{-1}\) respectively (Table 1). Stations N2 and N3 displayed higher Cu concentration than world rock average, while total concentration of Mn was higher than the world rock average only at N3 (Table 1). For Cu contamination, CF values at N2 and N3 were >1 indicating a moderate level of contamination while the lower EF values of Cu indicated minimal enrichment at all three stations (Table 2). Distribution of Cu showed strong positive correlation with that of Fe, Cd, Ba and Co (Table 3). Copper is released in the environment mostly from industrial and domestic waste. Increased concentration of copper at stations N2 and N3 points towards siltation of sediment carried by Mahanadi. Mn concentration showed a strong positive correlation with that of Pb (Table 3). According to Muller’s classification, only N3 was moderately
Table 3. Correlation matrix

<table>
<thead>
<tr>
<th>Metals</th>
<th>pH</th>
<th>Salinity</th>
<th>OC</th>
<th>Fe</th>
<th>Pb</th>
<th>Cd</th>
<th>Cr</th>
<th>Hg</th>
<th>Ni</th>
<th>Co</th>
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<td>0.957</td>
<td>1.000**</td>
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*Correlation significant at 0.05 level (two-tailed); **Correlation significant at 0.01 level (two-tailed).
polluted with Mn as indicated by a positive *I*<sub>geo</sub> value (Table 2).

Total concentration of Hg at all three stations was higher than world rock average (Table 1). This study shows a significantly high concentration of Hg in Chilika (N1 and N2). CF, EF and *I*<sub>geo</sub> values for Hg were remarkably high at N1, where salinity was the highest (Table 2). Since N1 is located near the sea mouth, the concentration of other metals is relatively low. The negative correlation of Hg with the remaining metals indicates that the sources of Hg and other metals are likely to be different (Table 3). The Ba, Sb, V, Be and Tl values measured had no significant contamination on the Chilika environment.

In summary, Chilika Lake is observed to be polluted mainly with four metals, including Hg, Cd, Co and Pb of which only Hg and Cd show positive *I*<sub>geo</sub> indices indicating the lake to be severely contaminated with these metals. Overall, pollution load indices calculated followed the order: N3 (1.71) > N2 (1.34) > N1 (0.839). Presence of fishery market and fish catchment activities may be responsible for enhanced metal concentration at Kaluparaghat (N3), whereas metal contamination in Balugaon (N2) is attributed to the neighbouring township. In addition, concentration of various metals is increased due to discharge from River Mahanadi. Kaluparaghat which is located in the freshwater zone has the lowest salinity. It shows the highest total organic carbon and organic matter contributed by the teragenous materials and the decomposition of plants and animals by the action of bacteria.

### Multivariate analysis

Hierarchical cluster analysis based on nearest neighbour method using the mean of metal contaminants from three sampling stations yielded three clusters. Cd, Pb, Hg and Co which were present in elevated concentrations in Chilika Lake formed a cluster (Figure 2). In PCA, PC1 and PC2 explained 90.33% variance of the data. PC1 was positively loaded with all toxic metals, except Hg (Figure 3). PCA biplot implied that Hg was derived from a different source (Figure 3). It also supported the higher concentration of Hg at Rambhartia, which placed the station in the positive side of PC1 (Figure 3). Kaluparaghat, on the other hand, was positioned on the positive side of both the components in PCA due to high concentration of most toxic metals (Figure 3). Balugaon lying on the positive side of PC2 and negative side of PC1 showed higher concentration of Cr and Ni (Figure 3). Salinity which increased along the negative axis of PC2 showed a correlation with Hg at N1. Rambhartia (N1) situated near the sea mouth was not affected by the sediment carried by River Mahanadi. Thus increased Hg contamination at N1 presumably originated from natural sources which may include volatilization of ocean and/or atmospheric deposition.

Cluster analysis (Figure 2) reveals that the metals having high concentration form a group, as Pb, Co, Hg, Cd form a cluster followed by Ni, Cu and Cr, Zn. However, when the heavy metal concentration was compared with salinity, pH and total organic carbon in Pearson correlation matrix (Table 3), all the metals except Hg showed negative relation with salinity. This result was supported by PCA (Figure 3), where mercury was positively loaded with salinity and both were on the positive side of N1 sampling site. Thus these multivariate results depict that only mercury had a positive correlation with salinity and its source is also different compared to metals. Thus it is at high concentration in N1 sampling site. The other metals are high in N3 sampling sites where salinity is low.

### Cations

The distribution of cations followed a specific trend such that concentration of Mg < Ca < Na < K with the highest deposition of all metals at N2, except Ca (Table 1). Concentration of Ca was highest at N3, which ranged from 1866.37 to 10853.87 mg kg<sup>−1</sup> (Table 1). Mg content in the sediments depends on the nature and type of sediment. In general, Mg to Ca ratio is higher in sea water than river water<sup>17</sup>

Heavy metal concentration of Chilika was compared with other important Indian rivers and estuaries which also support a large number of populations. Chilika...
Table 4. Comparative analysis of concentration of heavy metals in different rivers and estuaries

<table>
<thead>
<tr>
<th>Metals</th>
<th>Chilika lagoon</th>
<th>Chilika lagoon (present study)</th>
<th>Mahanadi river</th>
<th>Godavari river</th>
<th>Pulicat Lake</th>
<th>Ennore Estuary</th>
<th>World rock average</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>24</td>
<td>89.8</td>
<td>43.33</td>
<td>30.94</td>
<td>57</td>
<td>73</td>
<td>32</td>
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<tr>
<td>Cd</td>
<td>–</td>
<td>–</td>
<td>2.06</td>
<td>2.87</td>
<td>–</td>
<td>–</td>
<td>0.2</td>
</tr>
<tr>
<td>Ni</td>
<td>67</td>
<td>66</td>
<td>61.33</td>
<td>45.46</td>
<td>9</td>
<td>52</td>
<td>49</td>
</tr>
<tr>
<td>Pb</td>
<td>&lt;25</td>
<td>–</td>
<td>25.76</td>
<td>17.73</td>
<td>60</td>
<td>13</td>
<td>23.7</td>
</tr>
<tr>
<td>Zn</td>
<td>–</td>
<td>72</td>
<td>83.33</td>
<td>77.02</td>
<td>125</td>
<td>53</td>
<td>127</td>
</tr>
<tr>
<td>Cr</td>
<td>64</td>
<td>158</td>
<td>124</td>
<td>77.60</td>
<td>15</td>
<td>126</td>
<td>81.3</td>
</tr>
</tbody>
</table>

All values are presented as mean concentration (mg kg⁻¹). The heavy metal concentration in the present study is shown in boldface.

Chilika lagoon was found to be less polluted than rivers Mahanadi and Godavari. Sediment of River Mahanadi was contaminated with high level of Pb. It is notable that the anthropogenic pressure is less in Chilika lagoon than that in Pulicat and Ennore estuaries (Table 4). The comparative account of different pollutants presented here indicates that all metal ions except Cd have decreased over the past 20 years, indicating a positive outcome of Chilika management plan (Table 4). In the 1990s the sea mouth of Chilika Lake was clogged as a result of which salinity declined. In 2000, a new opening of the Lake to the Bay of Bengal by the Chilika Development Authority allowed mixing of saline water with freshwater, which enhanced the saline condition of the lake. This also divided the lake into three major distinct saline environments such as freshwater, brackish water and saline water, which form the basis of selection of sites in this study. The concentrations of several heavy metals were observed to be reduced. This study shows that concentration of Pb has decreased over the years, although the present concentration is still higher than the world rock average. Cobalt concentration is still high, particularly in Balugaon and Kaluparaghat, probably due to the impact of Balugaon township, thus corroborating with the finding of Panda et al. Conclusion

Chilika lagoon is an important wetland which spreads over a vast area with varying physical and chemical characteristics, and political and socio-economic importance. Balugaon township and the fish catchment areas/markets have profoundly affected the health of Chilika Lake as indicated by this study. High concentrations of Cd, Hg, Co and Pb have been recorded in the soil sediments.
Slow absorption of these heavy metals in the sediments may induce severe damage to the ecosystem vis-à-vis biodiversity of the lake. The correlation between heavy metal concentration and salinity gradient indicates that the source of contamination is predominantly the sediments carried by the Mahanadi riverine system. A suitable management plan comprising regular monitoring of the sediment quality of Chilika Lake and pre-treatment of sediment carried by the river Mahanadi needs to be implemented as indicated by this study. Further studies are necessary for risk analysis of the lagoon and detailed mapping of pollution sources.


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