

METAL POLLUTION ASSESSMENT OF SEDIMENT AND WATER IN THE RIVER HINDON, INDIA

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Abstract. The metal pollution in water and sediment of the River Hindon in western Uttar Pradesh (India) was assessed for Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn. The metal concentrations in water showed wide temporal variation compared with bed sediment because of variability in water discharge and variations in suspended solid loadings. Metal concentrations in bed sediments provided a better evaluation of the degree and the extent of contamination in the aquatic environment, Santagarh and Atali being the most polluted sites of the river. The ratio of heavy metals to conservative elements (Fe, Al, etc.) may reveal the geochemical imbalances due to the elevated metal concentrations normally attributed to anthropogenic sources. Metal/Al ratios for the bed sediments of the river Hindon were used to determine the relative mobility and general trend of relative mobility occurred Fe > Mn > Zn > Cr > Ni > Pb > Cu > Cd.

Keywords: bed sediments, metal pollution, mobility

1. Introduction

The introduction of metallic pollutants into a river, whether it is natural (erosion) or artificial (anthropogenic), can occur in dissolved and particulate form. Depending on physico-chemical conditions, the pollutants in dissolved form can later precipitate. They can also be adsorbed by the iron or manganese oxides and hydroxides or co-precipitated with these, or form dissolved organic or inorganic complexes (Salomons and Forstner, 1984; Drever, 1988). Metal partitioning appears to be metal specific and the eventual fate of various metals is a function of the distribution between the aqueous phase, suspended sediments and bed load of the river (Salomons and Forstner, 1984; Forstner, 1985; Luoma, 1990). The analysis of heavy metals in sediments permits us to detect pollution that might not be detected by analysis of single water samples (Forstner and Salomons, 1980; Salomons and Forstner, 1984; Erel *et al.*, 1991).

Sakai *et al.* (1986) analyzed the distribution of Cd, Cr, Cu, Mn, Pb and Zn, in water and sieved sediment samples taken from the main stream of the Toyohira River, Japan and reported that the heavy metal concentrations generally increased with decreasing particle size of sediments. Sabri *et al.* (1993) determined the concentrations of various metals in water, suspended solids and surficial sediments of

the River Tigris at Samarra impoundment and found that the concentrations of most of the elements in the surficial sediments (except for Mn and Fe) were lower than those in the suspended solids indicating the importance of the suspended solids in transportation of heavy metals. Combest (1991) evaluated sediment trace metals in White Rock Creek watershed located in Dallas and Collin Counties of north central Texas, in relation to sediment sorption characteristics. Bertin and Bourg (1995) studied the geochemical characteristics of sediments in the Lot River basin contaminated by heavy metals (Cd, Pb and Zn) and reported that the heavy metal transport in the river takes place mainly in particulate form. Dupre *et al.* (1996) studied the three main phases (suspended load, dissolved load, and bed load) of materials carried by Congo River and its main tributaries for twenty five major and trace elements and reported that 80% of the material is transported through dissolved load. Viers *et al.* (2000) studied chemical weathering processes and element transport mechanisms in the Nyong River basin of the Congo craton in Central Africa and found that there was a progressive increase of chemical weathering intensity with the decrease of the watershed drainage area. Chemical weathering mainly occurred in swamp zones where organic matter favors mineral dissolution.

Horowitz *et al.* (1999) analysed bed sediment, suspended sediment and fresh floodplain samples from the Seine River basin in France. The concentrations associated with floodplain sediments indicate that within the basin, trace element concentrations vary spatially by as much as three orders of magnitude, which may be attributed to increase in the population as well as concomitant increases in industrial activity.

In our earlier papers, we have reported the role of river bed sediments in controlling the metal pollution (Jain and Ram, 1997a,b; Jain and Ali, 2000; Jain and Sharma, 2001, 2002). In the present work, an attempt has been made to establish the role of bed sediments as indicators for assessing the metal pollution. The extent of contamination has been determined and relative mobilities of different metals presented.

2. The River System

The River Hindon is one of the important rivers in western Uttar Pradesh (India) having a basin area of about 7000 km² and lies between latitude 28°30' to 30°15' N and longitude 77°20' to 77°50' E (Figure 1). The river originates from Upper Shivaliks (Lower Himalayas) and flows through four major districts, viz., Saharanpur, Muzaffarnagar, Meerut and Ghaziabad, a distance of about 200 km before joining the river Yamuna downstream of Delhi. Physiographically the area is generally flat except the Shivalik hills in the north and north east. Deep gorges have been cut by *nalas* and rivers flowing through the area.

The major land use in the basin is agriculture, with little forest cover. On the basis of land use map, the study area can be demarcated into five categories: agriculture

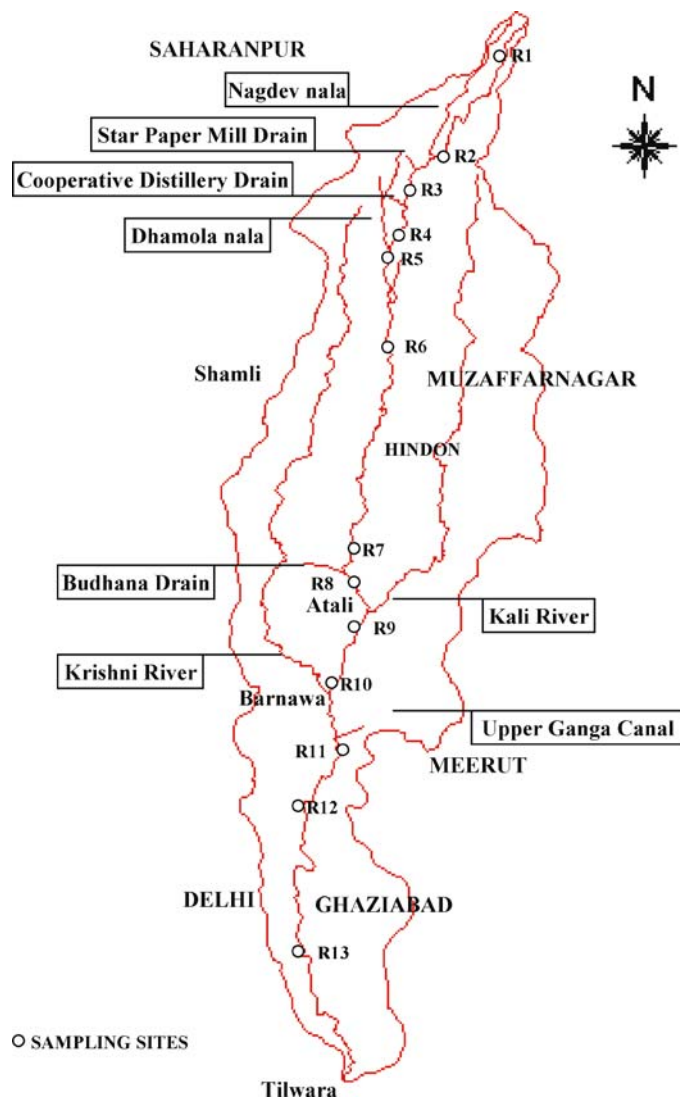


Figure 1. The Hindon River system showing location of sampling sites: R1-Khajnawar; R2-Beherki; R3-Santagarh; R4-Nanandi; R5-Sadhauri Hariya; R6-Maheshpur; R7-Budhana; R8-Chandheri; R9-Atali; R10-Barnawa; R11-Daluhara; R12-Surana; R13-Mohannagar.

(78.94%), urban area (6.63%), barren land (12.32%), forest cover (2.09%) and water bodies (0.02%). The climate of the region is moderate subtropical monsoon type. The average annual rainfall is about 1000 mm, most part of which occurs during the monsoon period (June to September).

The basin is densely populated because of rapid industrialization and agricultural growth during last few decades. The main sources of pollution in River

Hindon include municipal wastes from Saharanpur, Muzaffarnagar and Ghaziabad urban areas and industrial effluents of sugar, pulp and paper, distilleries and other miscellaneous industries through tributaries as well as direct outfalls. In summer months the river is dry from its origin up to Saharanpur town. The effluents of Nagdev *nala* and Star Paper Mill at Saharanpur generate the flow of water in the river. The municipal wastewater generated from the Saharanpur city is discharged to the Hindon River through Dhamola *nala*. The industrial effluents from the Cooperative Distillery and municipal wastewater from Budhana town also join the river in this stretch.

The River Kali joins the River Hindon near the village of Atali. It carries municipal wastewater and effluents of industries located in the Muzaffarnagar city. The Krishni River meets Hindon at Barnawa in Meerut district and transports the waste water from a sugar mill and distillery. River Hindon joins River Yamuna at Tilwara.

3. Material and Methods

Twelve sets of samples were collected from 13 locations on the River Hindon on alternate months during April 1997 to February 1999. The water samples were collected in polyethylene bottles from 1/3, 1/2 and 2/3 width of the river at a depth of 15 cm and combined. All the sample bottles and other containers were soaked in 10% nitric acid for 48 h and rinsed with deionized water several times prior to use. Water samples were filtered through Whatmann 0.45 μm pore diameter membrane filters. The filtered samples were preserved by acidifying with concentrated ultra pure nitric acid to $\text{pH} < 2$ and stored at 4 °C in polyethylene bottles. For total metal analysis, 100 mL of unfiltered water samples were acidified with 2 mL ultra pure nitric acid and digested on a hot plate until the volume was about 30 mL. The digested samples were filtered and made up to 100 mL with deionised water and stored at 4 °C. The difference between the total and dissolved metal concentrations gives the concentration of particulate metal.

Sediment samples were collected using an Ekman dredge and stored in pre-cleaned polyethylene bags for processing. The size distribution of the sediment samples was determined with nylon sieves to obtain various fractions (0–75, 75–150, 150–210, 210–250, 250–300, 300–425, 425–600 μm) by dry sieving. Organic matter was determined by wet oxidation in an acid dichromate solution, followed by back titration of remaining dichromate with ferrous ammonium sulphate solution. Bed sediments of 0–210 μm size were digested using acid ($\text{HF} + \text{HClO}_3 + \text{HNO}_3$) mixture for metal analysis. The concentrations of various metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) were determined in this digested sediment fraction of the river.

All chemicals used in the study were obtained from Merck, India/Germany and were of analytical grade. Deionised water was used throughout the study. All glassware and other containers were thoroughly cleaned and rinsed with deionised water several times prior to use. The metal standards prepared were checked with

standard reference material obtained from National Bureau of Standards (NBS), U.S.A. before each metal analysis.

Perkin-Elmer Atomic Absorption Spectrometer (model 3110) was used for metal analysis of both water and sediment. Average values of five replicates (re-analysis of the same digestate) were taken for each determination. Operational conditions were adjusted in accordance with the manufacturer's guidelines to yield optimal determination. Quantification of metals was based upon calibration curves of standard solutions of metals. These calibration curves were determined several times during the period of analysis. The detection limit for different metals were 0.0005, 0.002, 0.001, 0.003, 0.001, 0.004, 0.01 and 0.0008 mg/L for Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn, respectively. The precision of the analytical procedures, expressed as the relative standard deviation (rsd) ranged from 5 to 10% for different metals. Precision for the analyses of standard solutions was better than 5%.

4. Results and Discussion

4.1. METAL CONCENTRATIONS IN RIVER WATER

The longitudinal variations of dissolved, suspended and total metal concentrations of Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn along the course of River Hindon are presented in Figures 2–4. The higher concentrations of Fe, Cu, and Zn occurred in particulates whereas Mn, Cr, Ni, Pb and Cd were observed in higher concentrations in dissolved form in the river water. Rafael *et al.* (1990), Modak *et al.* (1992) and Vertacnik *et al.* (1995) also observed similar trends for different river systems. Higher concentration of total Fe and Mn in the upper stretch of the river may be attributed to the effluent of the Cooperative distillery and runoff from agricultural fields, respectively, whereas in the downstream section the concentration of Fe and Mn decreased substantially due to dilution effect. Cu, Cr, Zn and Cd were occurred in low concentrations at all the sites. The higher concentration of these metals in the upper section of the river, may be linked to combined effluents of the paper mill and distillery. In the downstream section, these concentrations decreased considerably in the mid-portion due to dilution from Dhamola *nala*, which has significant flow throughout the year. Further downstream, higher concentration of these metals may be attributed to the mixing of water of River Kali and Krishna, which carry municipal and industrial effluents of various types of industries of Muzaffarnagar region and sugar mill effluent from Shamli. The higher concentration of Ni and Pb in the upper stretch of the river may be attributed due to the discharge of paper mill effluents. Downstream, dissolved concentrations remained almost constant.

In general, the concentrations of all dissolved metals are lowest in winter months and highest during summer months. The concentration of dissolved metals decreased in the monsoon months due to dilution during higher flow. Higher percentages of almost all metals in particulate form occurred during the post-monsoon

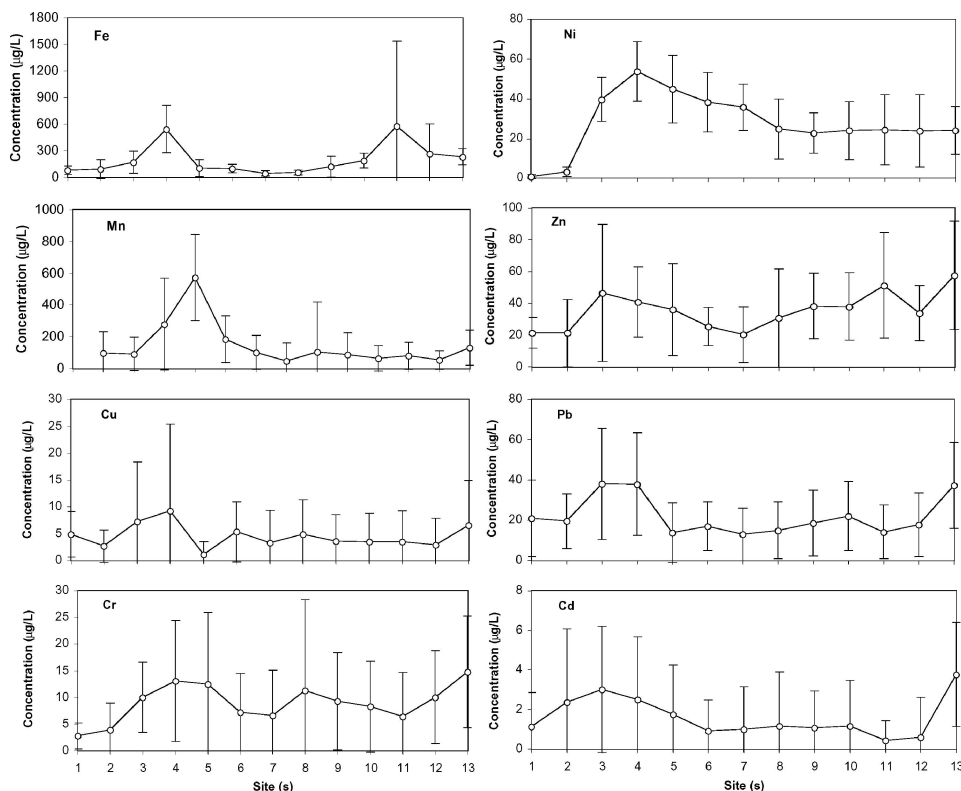


Figure 2. Longitudinal variation of dissolved metal concentrations.

months due to suspended load carried by surface runoff during monsoon season. Any deviations from these trends may be attributed to the site-specific activities, which are likely to increase suspended solid concentration in the water column and thereby decreasing the dissolved metal.

4.2. METAL CONCENTRATIONS IN RIVER BED SEDIMENTS

The sediment of River Hindon consists of more than 90% sand and <10% silt plus clay in the upper portion of the river. However, the concentration of clay and silt (0–75 μm) increased in the down stream section of the river. Most of the sediment transported and deposited by the river consists of fine (0–75 μm) to medium (210–250 μm) grained sand. The organic matter of the sediment occurred of the order of 0–1%.

Heavy metal concentrations in sediments are affected by particle size and composition of sediments (Thorne and Nickless, 1981; Kristensen, 1982; Thomson *et al.*, 1984; Krumlgalz, 1989). The concentrations of various metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn) in 0–210 μm sediment fraction of the river are presented in

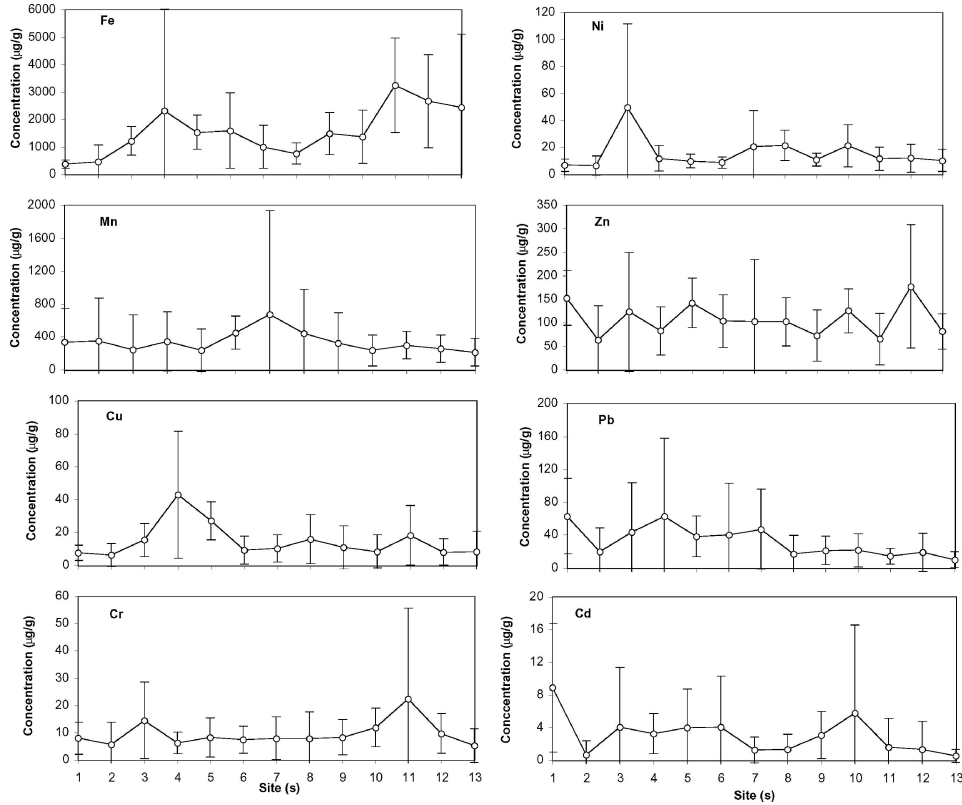


Figure 3. Longitudinal variation of particulate metal concentrations.

Figure 5. Higher concentrations of iron and manganese occurred in the upstream section of the river, which indicates the possibility of the presence of Fe and Mn minerals other than hydroxides. In the middle portion of the river, the concentration of iron and manganese remains almost constant. However, in the downstream section of the river the concentration of iron and manganese increases below the confluence of the Kali and Krishni rivers which carry composite waste effluents from different kinds of industries. The maximum concentration of Fe occurred at Mohan Nagar due to water impoundment for longer periods and site specific activities.

The trend of other metals at different sites of the river, viz., copper, chromium, nickel, zinc and lead occurred to be the same as that of the metal concentrations in the associated water column. The maximum concentrations of these metals occurred downstream site of the river at Mohan Nagar due to water impoundment for longer periods.

The elevated levels of metals occurred during summer months and lower concentrations occurred during monsoon months. During monsoon season, polluted sediment particles (specially fine fraction) may disperse through suspension in the

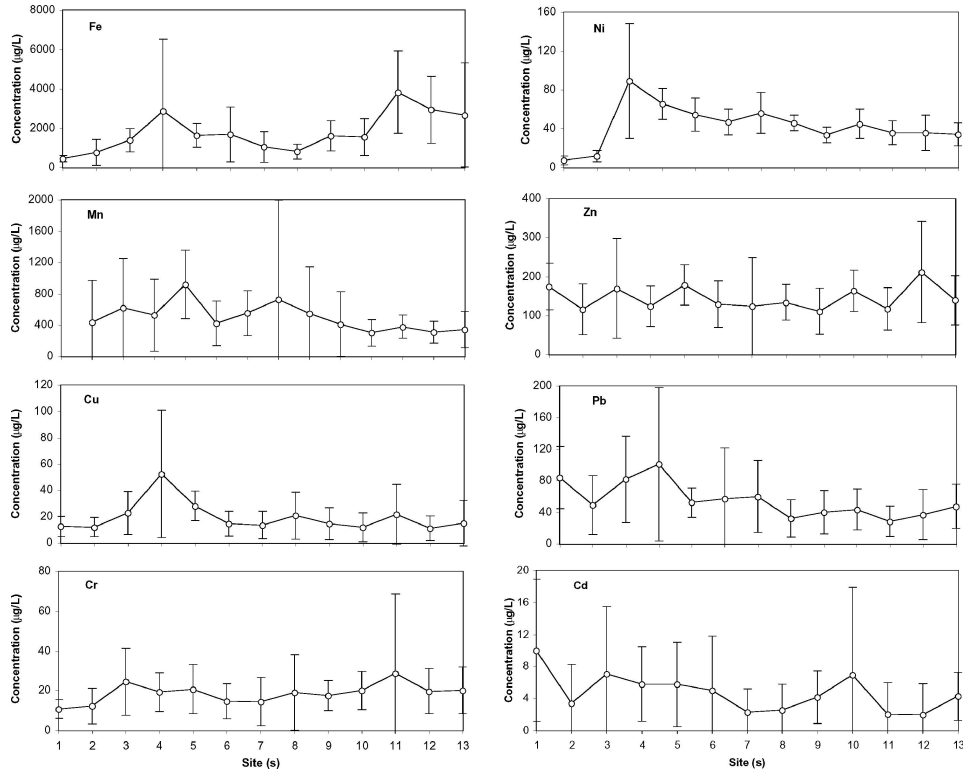


Figure 4. Longitudinal variation of total metal concentrations.

bottom sediment layer, and thus their distribution in the surface sediment was expected to become similar to effluent dilution or dispersion in the water of the river. This leads to lower concentrations of metals in the sediments during monsoon season. Similar findings were also reported by Geesey *et al.* (1984). Following peak discharge, the concentration of the metals in bed sediments increased as the flow again decreased. The highest accumulation of metals occurs in low flow period and the lowest accumulation of metals occurs during the high flow period, suggesting that the concentration of heavy metals in bed sediments is brought about by changes of water flow.

By comparing Figures 2 and 5, the metal concentrations (except Fe and Cr) in sediment were lower than the levels of concentration in associated water columns. The variability in metal concentrations in sediment during different months was also lower (as is indicated by the standard deviation of metal concentration) at all the sites as against the metal concentrations in the associated water column which is highly influenced due to variability in flow conditions. Thus it can be inferred that the bottom sediment provide a more stable base for contaminative studies.

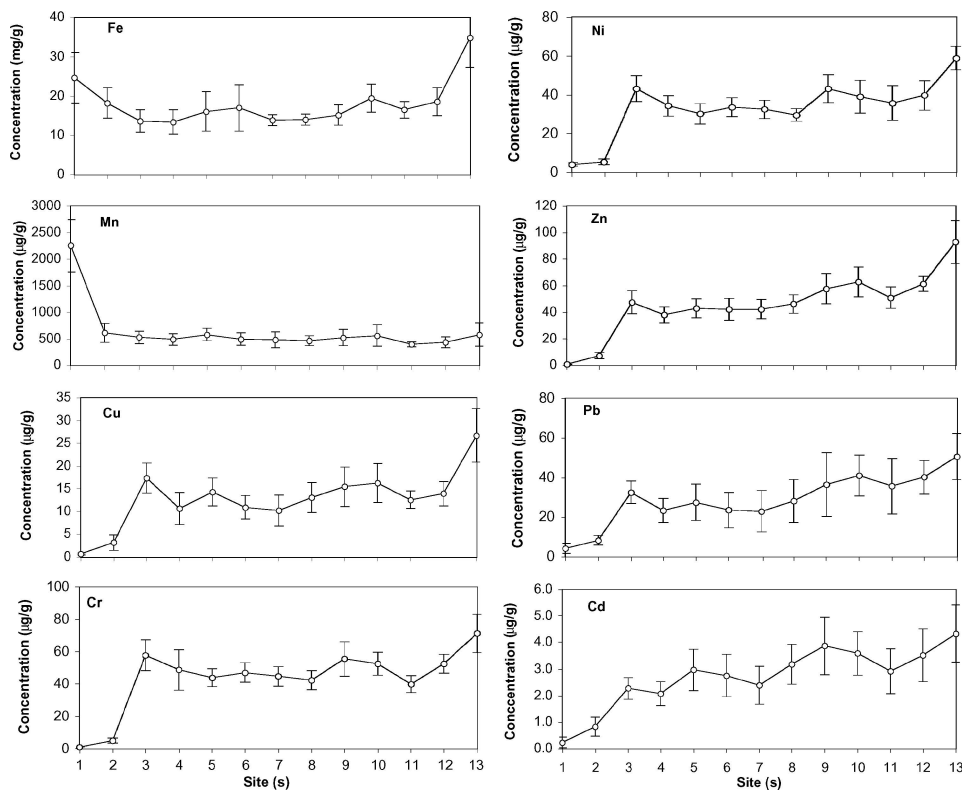


Figure 5. Longitudinal variation of metal concentrations in bed sediments.

The heavy metal data has also been subjected to simple linear regression analysis to examine the possible correlation among different metal ions. Table I shows the correlation coefficients among different metals in the bed sediments of the River Hindon. High correlation coefficients (>0.9) between Ni–Cr and Ni–Zn indicate common source, their mutual dependence and identical behavior during transport. The absence of strong correlation among other metals suggests that the concentrations of these metals are not controlled by a single factor, but a combination of geochemical support phases and their mixed associations. Depending on the degree of contamination of the site and the heavy metal investigated, high metal concentrations may be associated with iron, manganese, organic and inorganic carbon, calcium, sulfide and small particles (Bertin and Bourg, 1995).

The concentration of heavy metals is affected by the particle composition of sediments. In sediments of polluted streams the largest amounts of heavy metals are associated with organic matter (humic and fulvic acids, colloids, synthetic organic substances), the fine grained sediment fraction (clay, silt and fine sand) and Fe/Mn hydrous oxides, or are precipitated as hydroxides, sulphites or carbonates (Forstner, 1981). Interactions between metals and organic matter in bed sediment have often

TABLE I
Correlation among different metals and organic matter (OM) in bed sediments

	Fe	Mn	Cu	Cr	Ni	Zn	Pb	Cd	OM
Fe	1.000								
Mn	0.381	1.000							
Cu	0.272	-0.415	1.000						
Cr	0.085	-0.540	0.845	1.000					
Ni	0.180	-0.494	0.858	0.926	1.000				
Zn	0.319	-0.471	0.872	0.885	0.920	1.000			
Pb	0.247	-0.374	0.802	0.791	0.859	0.863	1.000		
Cd	0.177	-0.463	0.760	0.789	0.787	0.831	0.864	1.000	
OM	0.609	0.035	0.457	0.374	0.430	0.563	0.427	0.353	1.000

been recorded (Langston, 1982; Wren *et al.*, 1983; Stephenson and Mackie, 1988; Rada *et al.*, 1989; Coquery and Welbourn, 1995).

Metal concentrations in the bed sediments of River Hindon are positively correlated with organic matter content, although the correlation is not very strong (Table I). Only two metals, iron and zinc, have correlation coefficient >0.5 indicating the partial affinity of these metals for organic fraction of the sediments. Dissolved and particulate organic carbon in the water column act as scavengers for metals, and the scavenged metals may then be incorporated into the bottom sediments.

4.3. PARTITION COEFFICIENTS

The mean values of partition coefficient (P_c) were calculated using the following equation and are given in Table II.

$$P_c \text{ (L/kg)} = (\text{Metal in solid, } \mu\text{g/kg}) / (\text{Metal in water, } \mu\text{g/L})$$

It was observed from the results that the partition coefficients increased in the downstream section of the river indicating higher adsorption capacity of the sediment. This may be attributed to the presence of fine fraction of the sediment in the downstream section of the river.

4.4. MOBILITY OF METALS

The mobility of metal ions is dependent upon their sorption by sediments and their redistribution with deposition at the sediment–water interface. Further, both mobility and persistence are controlled by the nature of the metal bonding, sediment type and water chemistry (Jha *et al.*, 1990). Some factors cause mobilization of

TABLE II
Partition coefficients ($P_c \times 10^5$ L/Kg)

Site no.	Location	Fe	Mn	Zn	Ni	Pb	Cu	Cr	Cd
1	Khajnawar	0.585	0.120	0.001	0.005	0.001	0.001	0.001	0.002
2	Beherki	0.470	0.050	0.001	0.006	0.002	0.009	0.008	0.007
3	Santagarh	0.113	0.019	0.004	0.006	0.006	0.023	0.045	0.009
4	Nanandi	0.082	0.007	0.004	0.006	0.004	0.012	0.031	0.008
5	Sadhauli Hariya	0.012	0.019	0.003	0.006	0.006	0.018	0.033	0.013
6	Maheshpur	0.147	0.010	0.004	0.008	0.007	0.020	0.058	0.013
7	Budhana	0.208	0.031	0.005	0.006	0.006	0.021	0.049	0.016
8	Chandheri	0.220	0.034	0.004	0.007	0.016	0.023	0.060	0.020
9	Atali	0.114	0.023	0.007	0.014	0.017	0.034	0.037	0.017
10	Barnawa	0.189	0.026	0.004	0.010	0.012	0.063	0.036	0.019
11	Daluhera	0.069	0.014	0.006	0.011	0.017	0.036	0.052	0.022
12	Surana	0.085	0.017	0.004	0.016	0.017	0.047	0.072	0.027
13	Mohannagar	0.229	0.028	0.009	0.020	0.016	0.074	0.091	0.017

metals from sediments and make them available to living organisms, changes in water and sediment conditions (pH, redox potential, dissolved oxygen, etc.), re-suspension of deposited particulates, microbial activity, textural characteristics of sediments and partitioning equilibria involving water, clay, organic substances and lipids. The mobility of metals is also regulated by the chemically mobile fraction of the sediments.

In the present study, iron and aluminium have been chosen as conservative elements for the analysis, because of their relative abundance in the earth's crust and thus their decreased tendency to be greatly influenced by human activities. The metal/Fe and/or metal/Al ratios minimize the grain size effects on heavy metal data and hence are used to study the mobility of heavy metals in the riverine environment. It is generally assumed that Fe and Al has reached a steady state and is not being accumulated by the soil layer and is derived only from land erosion (Martin and Meybeck, 1979). Thus, by studying the metal/Fe and/or Metal/Al ratio, it is possible to determine the relative mobility of different metals. Metal/Al ratios for the bed sediments of the River Hindon are depicted in the Figure 6 and general trend of relative mobility is observed to be $Fe > Mn > Zn > Cr > Ni > Pb > Cu > Cd$ in both the cases.

In a natural riverine sediment system, elements as well as metals exist together in relative proportions to each other and the metal pair ratio is influenced by various geo-chemical processes. The ratio of heavy metals to conservative elements (Fe, Al, Si, etc.) may reveal the geochemical imbalances due to the elevated metal concentrations normally attributed to anthropogenic sources.

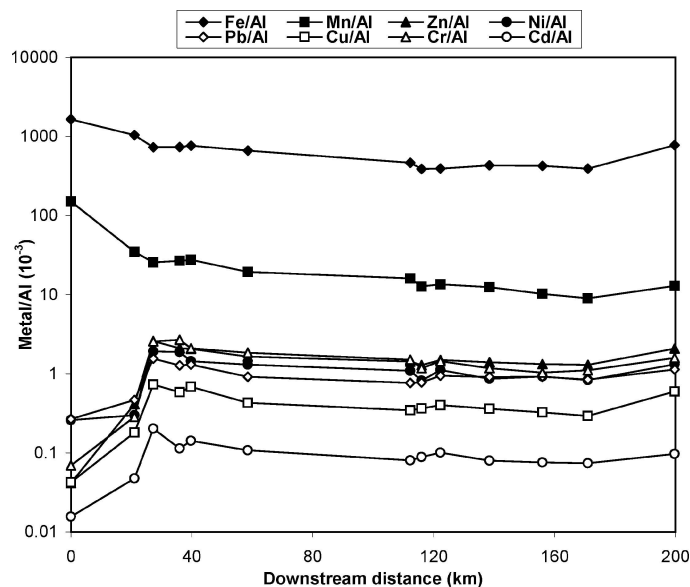


Figure 6. Variation of metal/Al ratio at different sites.

The metal pair ratios are dependent on a large number of processes in the geochemical cycle including weathering, transport and deposition. An attempt has also been made in the present study to assess heavy metal contamination in sediments based on geochemical elemental concentration ratios. Table III show the heavy

TABLE III
Metal/Al ratio for different metals in bed sediments

Site no.	Location	Fe/Al ($\times 10^{-3}$)	Mn/Al ($\times 10^{-3}$)	Zn/Al ($\times 10^{-3}$)	Ni/Al ($\times 10^{-3}$)	Pb/Al ($\times 10^{-3}$)	Cu/Al ($\times 10^{-3}$)	Cr/Al ($\times 10^{-3}$)	Cd/Al ($\times 10^{-3}$)
1	Khajnawar	1647	150.7	0.04	0.25	0.27	0.04	0.07	0.01
2	Beherki	1037	34.7	0.41	0.29	0.47	0.18	0.29	0.04
3	Santagarh	730.6	25.6	2.59	1.92	1.55	0.73	2.57	0.20
4	Nanandi	736.2	26.7	2.08	1.88	1.28	0.59	2.68	0.11
5	Sadhuli Hariya	767.3	27.6	2.05	1.43	1.31	0.68	2.10	0.14
6	Maheshpur	664.9	19.3	1.64	1.30	0.92	0.43	1.84	0.10
7	Budhana	466.4	16.1	1.42	1.09	0.77	0.34	1.50	0.08
8	Chandheri	388.5	12.7	1.28	0.81	0.78	0.36	1.17	0.09
9	Atali	392.7	13.4	1.49	1.11	0.95	0.40	1.44	0.10
10	Barnawa	432.2	12.4	1.39	0.86	0.91	0.36	1.17	0.08
11	Daluhera	426.1	10.2	1.31	0.92	0.92	0.25	1.03	0.08
12	Surana	390.4	9.0	1.29	0.83	0.84	0.29	1.10	0.07
13	Mohannagar	777.1	12.8	2.07	1.31	1.12	0.57	1.59	0.10

metal to aluminium ratios for different sampling sites of the river. It can be seen from the results that the site no. 3 (Santagarh, site at River Hindon just downstream of the confluence of Nagdev *nala* and Star Paper Mill drain) is the most polluted site followed by site no. 9 (Atali, site at River Hindon just downstream of the confluence of Kali River) having enrichment of almost all the metals.

5. Conclusion

The River Hindon is subjected to varying degree of pollution caused by numerous untreated and/or partially treated waste inputs of municipal and industrial effluents. The river is highly influenced due to heavy metals, which enter the river system, by direct discharges of municipal and industrial effluents and surface runoff. Higher concentrations of metals in river water in the upper stretch are largely due to the mixing of effluents from Star Paper Mill and Cooperative Distillery. The variability of metal concentrations in sediments during different months was quite low as against the metal concentrations in the associated water column. Thus sediments existing at the bottom of the water column provide a stable base for contaminative studies. The general trend of relative mobility of heavy metals is observed to be $Fe > Mn > Zn > Cr > Ni > Pb > Cu > Cd$. The metal ion pair ratio suggests that Santagarh (site no. 3) is the most polluted site of the river.

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