

HEAVY METAL TRANSPORT IN THE HINDON RIVER BASIN, INDIA

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Abstract. Total mass transfers of heavy metal in dissolved and particulate form has been determined in the downstream section of river Hindon, an important tributary of river Yamuna (India). The contribution of different point sources to the river Hindon has also been assessed. The river Kali has the largest contribution to the river Hindon. The highest metal loads were related to the highest flow of the river and thereby increased both by surface runoff and sediment resuspension. The contribution of monsoon months to the total transported load was also calculated and it was observed that monsoon months contributes more than 40% of total loading annually for all the metals. The metal fluxes from the river Hindon were compared with other rivers of Indian sub-continent.

Keywords: heavy metal, load, river system, transport

1. Introduction

The elevated levels of trace metals in natural water systems pose a severe threat to the aquatic environment (Holeman, 1968; Meybeck, 1976; Martin and Meybeck, 1979; Forstner and Wittmann, 1983; Milliman and Meade, 1983; Jha *et al.*, 1988). Foster and Charlesworth (1996) have reviewed the major sources and transport characteristics of heavy metals in the hydrological cycle. Many different metals have been studied in environmental pollution studies, particularly Hg, Cu, Zn, Pb, Cr and Cd (Rippey *et al.*, 1981). Davide *et al.* (2003) studied the characteristics of bed sediments and suspension of the river Po (Italy) with special reference to grain size distribution, major elements, nutrients and trace metals under normal and high flow conditions and concluded that both nutrient and trace metal particulate concentrations substantially decreases under high flow conditions.

The continental contribution of heavy metals to the world's ocean is quite large, as >97% of the mass transport of metals is associated with river sediments (Gibbs, 1977). A variety of factors such as basin geology, physiography, chemical reactivity, lithology, mineralogy, hydrology, vegetation, land use pattern and biological productivity regulate the metal load of a river system (Dahlberg, 1968; Garrels *et al.*, 1975; Warren, 1981; Aurada, 1983). Due to the relative mobility of metals during transport processes, metal content in sediments can reflect the present and the historical industrial and urban development of the basin.

The metal contribution from Indian rivers, which carry 20% of the global supply of sediments to the oceans has not been properly assessed (Subramanian, 1979; Martin and Meybeck, 1979; Sarin *et al.*, 1979). Subramanian *et al.* (1985) attempted to estimate an average chemical composition of Indian river sediments based on large rivers. Borole *et al.* (1982) and Subramanian *et al.* (1988) have reported metal concentrations and their non-conservative behaviour in estuarine sediments, on the west and east coast of India. Recent geochemical studies of some basins (Ramesh, 1985; Ramesh and Subramanian, 1986; Raymahashay, 1987; Subramanian *et al.*, 1987; Biksham and Subramanian, 1988; Ramanathan *et al.*, 1988; Subramanian and Jha, 1988) have yielded additional data to improve and update information on the metal contribution of Indian rivers to the adjacent ocean.

The river Yamuna contributes 64×10^6 t year⁻¹ of suspended sediments and 42×10^6 t year⁻¹ of dissolved load to the Ganges (Jha *et al.*, 1988). Subramanian and Sitasawad (1984) reported that annually, 86 t Ni, 64 t Cr, 61 t Pb, 45 t Fe and 36 t Zn, derived from industrial effluents and city wastes are disposed into the river Yamuna in the vicinity of Delhi alone. In earlier papers, we have studied distribution and adsorption characteristics of heavy metals on bed sediments of river Hindon (Jain and Sharma, 2001, 2002). In this paper an attempt has been made to estimate the contribution of different point sources to the river Hindon and to calculate the mass transfer within the downstream portion of the river Hindon.

2. Study Area

The river Hindon is among 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 in western Uttar Pradesh and covers a distance of about 200 km before joining the river Yamuna downstream of Delhi. The major land use in the basin is agriculture and there is no effective forest cover. On the basis of land use map the study area can be demarcated into five categories: agriculture (78.94%), urban area (6.63%), barren land (12.32%), forest cover (2.09%) and water bodies (0.02%) (Sharma, 2001). The climate of the region is moderate subtropical monsoon type. The average annual rainfall is about 1000 mm, major part of which is received during the monsoon period (June to September).

A general plan of sampling locations with respect to different outfalls of municipal and industrial effluents in river Hindon is shown in Figure 1. In all eight stations in the waste effluents, tributaries and canal joining the river and one station in the downstream of the river at Mohannagar were selected for monitoring.

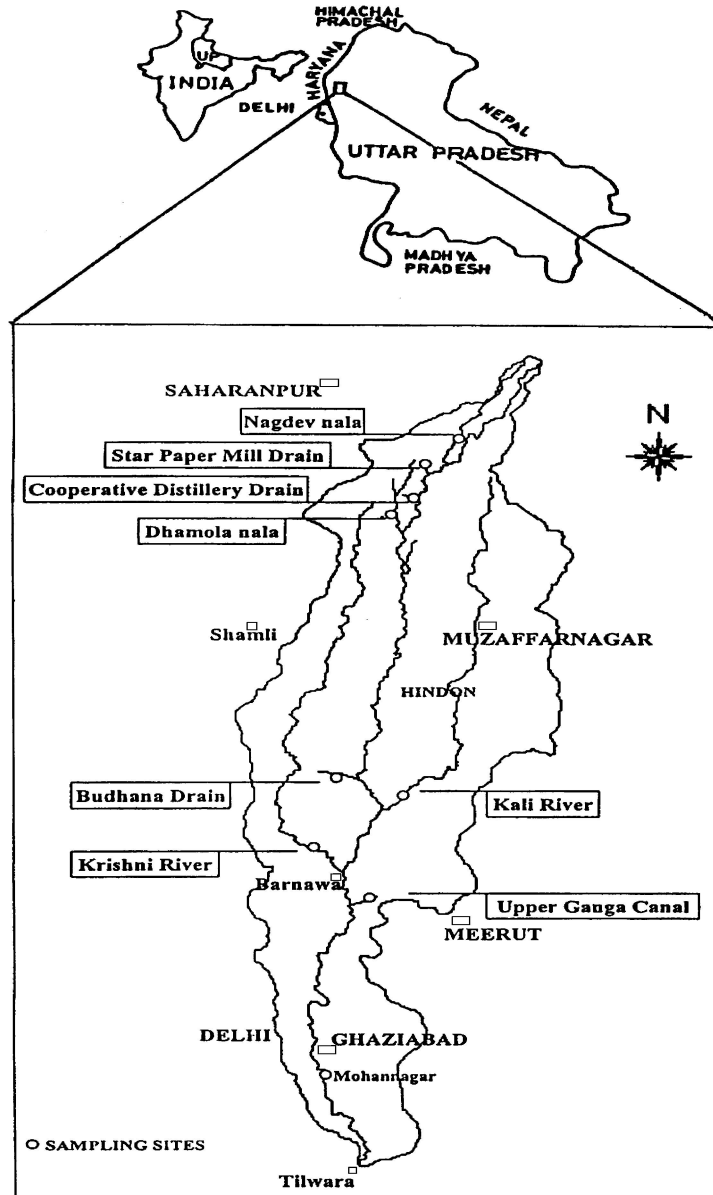


Figure 1. The Hindon River system showing location of sampling sites.

3. Material and Methods

For the pollution survey of the river, twelve sets of samples were collected from various locations on alternate months for a period of two years (April 1997 to March 1999) by dip (or grab) sampling method. The water and wastewater samples were

collected in polyethylene bottles from 1/3, 1/2 and 2/3 width of the river and mixed together to obtain a composite sample. All the samples were collected at a depth of 15 cm to avoid introduction of floating particles, using standard water sampler (Hydro Bios, Germany). Discharge of waste effluents and the river downstream was measured using current meter by area-velocity method (Sharma, 2001) at the time of sampling.

For trace metal analysis, sample bottles and other containers were soaked in 10% nitric acid for 48 h and rinsed with deionized water several times prior to use. For dissolved metal analysis, water samples were filtered through Whatmann 0.45 μm pore diameter membrane filters. The filtered samples were then 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 till the volume reduced to around 30 mL. The digested samples were then filtered and final volume was made upto 100 mL with deionised water and stored at 4 °C for total metal analysis. The difference between the total and dissolved metal concentrations gives the concentration of particulate metal.

Perkin-Elmer Atomic Absorption Spectrometer (model 3110) was used for metal analysis of water. Average values of five replicates 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 metal standard solutions (Merck, Germany). The standards were checked with standard reference materials obtained from National Bureau of Standards, USA. The detection limit for different metals were 3, 1, 1, 4, 0.8, 2, 10 and 0.5 $\mu\text{g/L}$ for Fe, Mn, Cu, Ni, Zn, Cr, Pb and Cd. The relative standard deviation (r.s.d.) ranged from 5 to 10% for different metals. Precision for the analyses of standard solutions was better than 5%.

4. Result and Discussion

The dissolved and particulate metal loads transported by different point sources to the river Hindon and their percentage contribution are given in the Tables I and II. By far the Kali river contributes the largest load (>50%) to the river Hindon with 347.3 t, 139.8 t, 59.4 t, 34.8 t, 16.4 t, 15.5 t, 7.39 t and 2.09 t of dissolved load of Fe, Mn, Zn, Pb, Ni, Cr, Cu and Cd respectively and 2650 t, 206.9 t, 157 t, 77.9 t, 58.4 t, 34.2 t, 23 t and 13.9 t of particulate load of Fe, Mn, Zn, Pb, Ni, Cd, Cu and Cr respectively. It can be inferred that higher loadings of river Kali, which carries municipal waste water and industrial effluents from a variety of industries of Muzaffarnagar region, were observed for all metals in particulate form which may be attributed to sediment transport function. The particulate loading contains highest load of iron and manganese as compared to other metals, which are important carrier

TABLE I
Dissolved metal loads transported by different point sources of river Hindon and their percentage contribution during the sampling period (April 1997 to March 1999)

Point sources	Discharge (m ³ /s)	Metal load (ton)									
		Zn	Ni	Pb	Cu	Fe	Mn	Cr	Cd		
Nagdev nala (D1)	0.11–1.04	0.675 (0.7)	0.711 (2.5)	0.445 (0.8)	0.058 (0.5)	1.982 (0.4)	1.651 (0.9)	0.168 (0.6)	0.080 (1.7)		
Star paper mill drain (D2)	0.27–0.56	0.991 (1.0)	1.015 (3.6)	1.128 (2.1)	0.528 (4.3)	7.183 (1.5)	2.260 (1.2)	0.444 (1.6)	0.166 (3.6)		
Cooperative distillery drain (D3)	0.09–0.15	0.460 (0.5)	0.275 (1.0)	0.227 (0.4)	0.036 (0.3)	6.932 (1.5)	2.535 (1.3)	0.079 (0.3)	0.031 (0.7)		
Dhamola nala (D4)	3.07–8.80	10.90 (11.0)	3.417 (12.1)	5.595 (10.3)	2.978 (24.2)	13.88 (3.0)	16.27 (8.4)	4.360 (16.1)	0.707 (15.2)		
Budhana drain (D5)	0.13–0.31	0.378 (0.4)	0.659 (2.3)	0.388 (0.7)	0.059 (0.5)	1.985 (0.4)	2.943 (1.5)	0.190 (0.7)	0.037 (0.8)		
Kali river (D6)	4.95–51.81	59.38 (60.1)	16.36 (57.8)	34.78 (64.2)	7.385 (60.1)	347.3 (74.4)	139.8 (72.2)	15.50 (57.2)	2.089 (44.9)		
Krishni river (D7)	0.98–10.40	11.70 (11.8)	5.894 (20.8)	5.107 (9.4)	0.844 (6.9)	68.21 (14.6)	23.09 (11.9)	3.265 (12.0)	0.288 (6.2)		
Upper Ganga canal (D8)	2.32–15.97	14.36 (14.5)	BDL (Nil)	6.547 (12.1)	0.397 (3.2)	19.56 (4.2)	4.998 (2.6)	3.103 (11.4)	1.260 (27.1)		

BDL – Below detection limit (values given in parenthesis shows % contribution of point source).

TABLE II
Particulate metal loads transported by different point sources of river Hindon and their percentage contribution during the sampling period (April 1997 to March 1999)

Point sources	Metal load (ton)									
	Zn	Ni	Pb	Cu	Fe	Mn	Cr	Cd		
Nagdev nala (D1)	3.082 (1.1)	0.281 (0.4)	0.594 (0.6)	0.362 (1.0)	39.97 (0.9)	10.81 (2.4)	0.239 (0.9)	0.082 (0.2)		
Star paper mill drain (D2)	2.580 (0.9)	0.481 (0.6)	1.265 (1.3)	1.956 (5.5)	40.76 (0.9)	4.771 (1.1)	0.322 (1.2)	0.055 (0.1)		
Cooperative distillery drain (D3)	0.714 (0.3)	0.088 (0.1)	0.312 (0.3)	1.837 (5.2)	7.364 (0.2)	0.921 (0.2)	0.219 (0.8)	0.037 (0.1)		
Dhamola nala (D4)	28.53 (10.1)	3.902 (4.8)	8.495 (8.5)	3.854 (10.8)	310.9 (7.0)	69.18 (15.4)	1.362 (5.1)	1.229 (3.0)		
Budhana drain (D5)	3.179 (1.1)	0.158 (0.2)	1.643 (1.7)	0.661 (1.9)	32.92 (0.7)	1.227 (0.3)	0.104 (0.4)	0.073 (0.2)		
Kali river (D6)	157.0 (55.8)	58.43 (72.2)	77.94 (78.3)	23.01 (64.6)	2650 (59.8)	206.9 (46.0)	13.86 (51.7)	34.23 (83.1)		
Krishni river (D7)	38.26 (13.6)	6.855 (8.5)	3.789 (3.8)	2.896 (8.1)	495.7 (11.2)	76.33 (17.0)	8.243 (30.8)	4.410 (10.7)		
Upper Ganga Canal (D8)	48.04 (17.1)	10.7 (13.3)	5.451 (5.5)	1.050 (3.0)	857.3 (19.3)	79.36 (17.7)	2.446 (9.1)	1.085 (2.6)		

Values given in parenthesis shows % contribution of point source.

for the transport of metal ion. The loading of Kali river is followed by Dhamola nala, which carries municipal waste of Saharanpur city as well as waste effluents from several small scale industrial units.

The monthly variation of total loads and flow of these point sources contributing to the river Hindon is shown in Figure 2. The general behavior of monthly metallic loads of Nagdev nala, Kali river, Krishni river and Upper Ganga Canal is almost same with small deviations. Further metallic load increased with increase in flow in these sources i.e. higher loads were observed in the monsoon months. It is also supported by the fact that the correlation coefficients among different metallic loads and flow of these sources is positive and greater than 0.5 for almost all metals (Table III) at level of significance of 5%. But the metallic loads of Star paper mill drain, Cooperative distillery drain, Dhamola nala and Budhana drain did not show systematic trend and are not significantly correlated with discharge.

The discharge profile of river Hindon at Mohannagar is depicted in Figure 3. The heavy metal concentrations in the river Hindon at downstream site (Mohannagar) during the study period from April, 1997 to March, 1999 is given in Table IV. The monthly variation of observed metallic load and flow at the downstream site of river Hindon is depicted in Figure 4. Higher metallic loads were always observed during higher flow months. By comparing Figures 2 and 4, it can be inferred that the general behaviour of the monthly metallic loads of different metals was almost similar between downstream site of river Hindon and its point sources Nagdev nala, Kali river, Krishni river and Upper Ganga Canal.

It was assumed that the metal concentrations remain constant between two sampling periods (60 days) but using the daily water discharge data, daily metallic load at downstream site, at Mohannagar was calculated. The other way to calculate

TABLE III
Correlation coefficients between metallic loads and flow of point sources and river Hindon downstream site

Point sources	Zn	Ni	Pb	Cu	Fe	Mn	Cr	Cd
Nagdev nala (D1)	0.855	0.925	0.922	0.831	0.877	0.945	0.549	0.775
Star paper mill drain (D2)	-0.097	0.661	0.406	0.418	-0.119	0.311	0.040	-0.400
Cooperative distillery drain (D3)	0.226	0.441	0.134	-0.008	0.300	0.425	-0.606	-0.138
Dhamola nala (D4)	0.530	0.179	0.418	-0.117	0.027	0.176	0.576	0.070
Budhana drain (D5)	0.608	0.802	0.784	0.327	0.024	0.668	0.680	0.134
Kali river (D6)	0.476	0.640	0.509	0.549	0.626	0.872	0.627	0.459
Krishni river (D7)	0.692	0.844	0.445	0.649	0.829	0.799	0.008	-0.248
Upper Ganga Canal (D8)	0.674	0.874	0.022	-0.195	0.740	0.701	0.775	0.579
D/S site of river Hindon (at Mohannagar)	0.683	0.637	0.237	0.327	0.633	0.610	0.540	0.341

Level of confidence = 5%.

the metallic load using monthly mean values of discharge instead of taking daily discharge. The results obtained by these two methods showed almost same values of total monthly loads. The higher load of Fe, Mn, Zn, Pb and Ni may be attributed to the composite waste of municipal and industrial effluents specially from sugar mills and steel factories which utilises these metals in their manufacturing process. The monthly metal loads were highly varied. Generally, the highest loads were related to the months of highest discharge and thereby to the highest suspended particulate

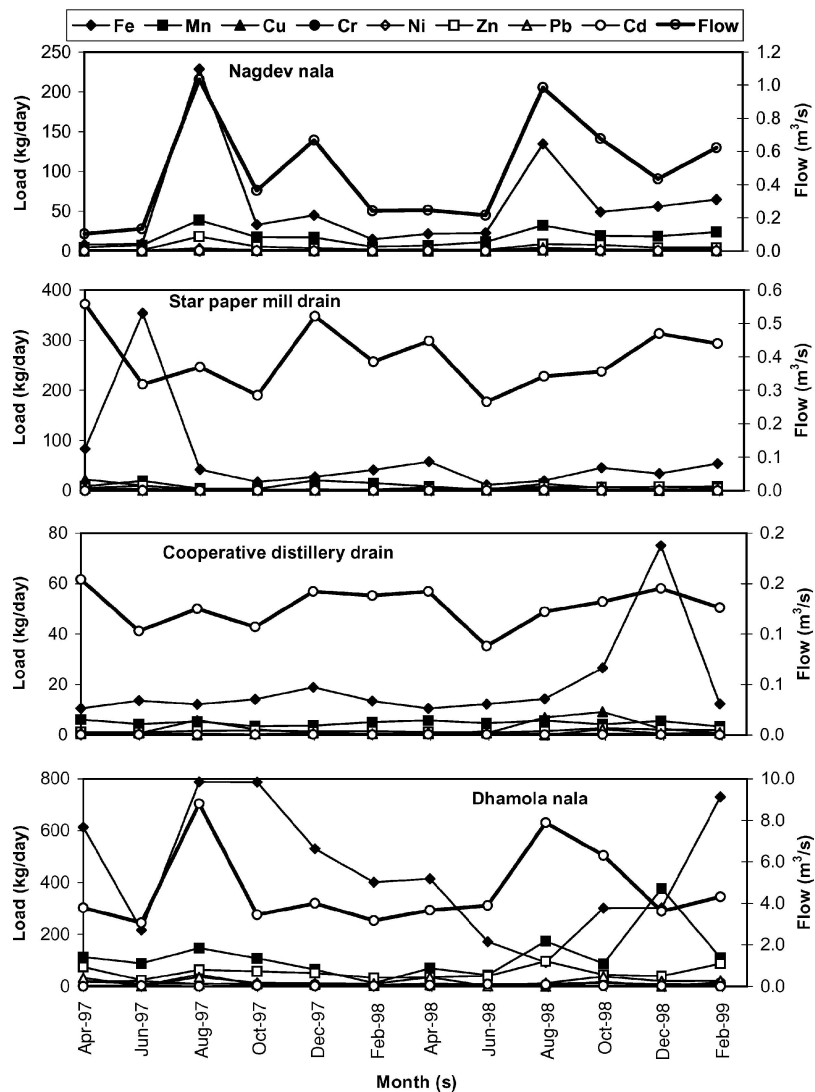


Figure 2. Monthly load and flow variation of different point sources contributing the river Hindon. (Continued on next page)

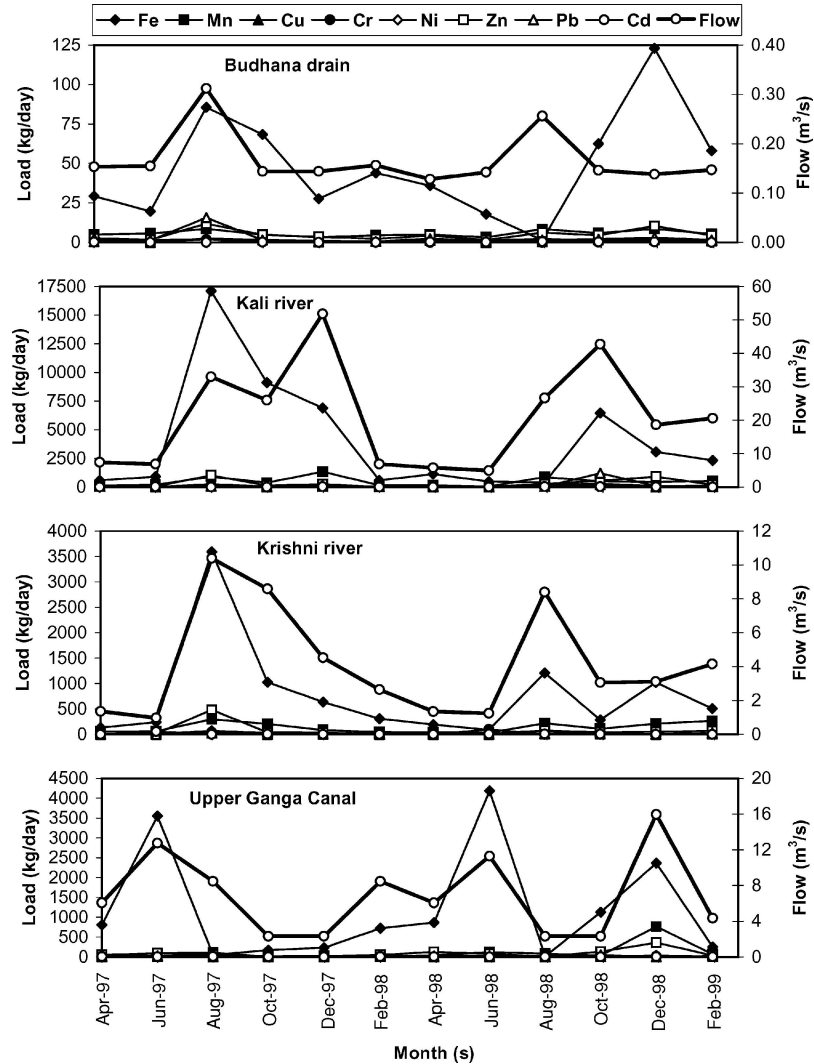


Figure 2. (Continued)

matter loads. This observation emphasizes the importance of suspended particulate matter as the main carriers for heavy metals in rivers. Similar conclusions were drawn by other workers (Jenne, 1968).

The seasonal pattern of different constituents is dominated by annual cycles of the year as well as release of canal water into the Hindon river. Generally, higher values of loading are observed during monsoon months which may be attributed to additional contribution of runoff from the agricultural fields. Higher load observed in the month of December may be attributed to the release of Upper Ganga Canal water into the river through Khatauli and Jani escapes. The particulate fraction

TABLE IV
Heavy metal concentrations in the river Hindon at downstream site (Mohannagar)
during the study period from April 1997 to March 1999

Metal	Form	Concentration ($\mu\text{g/L}$)			
		Min.	Max.	Mean	SD
Fe	Dissolved	44	356	226	90
	Particulate	467	9973	2438	2669
	Total	793	10164	2667	2653
Mn	Dissolved	14	423	129	110
	Particulate	45	621	214	170
	Total	59	743	344	231
Cu	Dissolved	BDL	22	6.6	8.3
	Particulate	BDL	43	8.3	12
	Total	BDL	62	15	17
Cr	Dissolved	1	36	15	10
	Particulate	BDL	19	5.3	6.2
	Total	1	38	20	12
Ni	Dissolved	4	43	24	12
	Particulate	BDL	23	10	8
	Total	13	54	34	12
Zn	Dissolved	6	127	58	34
	Particulate	20	144	82	38
	Total	26	271	139	63
Pb	Dissolved	6	69	37	21
	Particulate	BDL	31	10	10
	Total	8	90	47	28
Cd	Dissolved	BDL	9	3.8	2.6
	Particulate	BDL	2	0.5	0.8
	Total	BDL	11	4.3	3.0

BDL – Below detection limit.

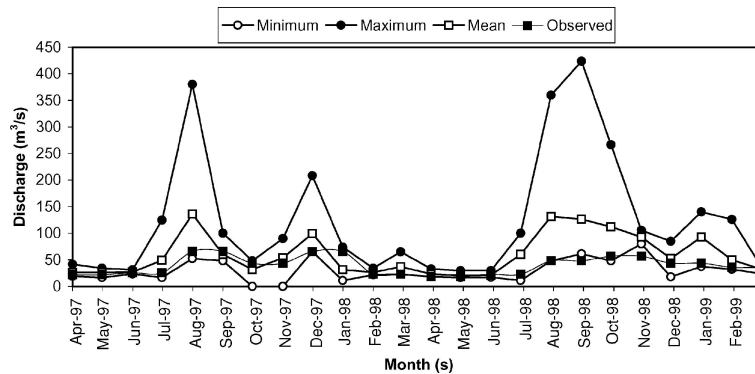


Figure 3. Discharge profile of Hindon river at Mohannagar during the study period (April, 1997 to March, 1999).

contains a higher loading of metal ions during monsoon months. This may be attributed to simple sediment transport functions, whereby increase in flow are associated with increased water turbulence and shear velocities resulting in an increased capacity to erode and transport particulate from the upstream catchment and channel network.

Daily rainfall and discharge pattern at downstream site, at Mohannagar is shown in Figure 5. The highest flow months have importance in calculation of total

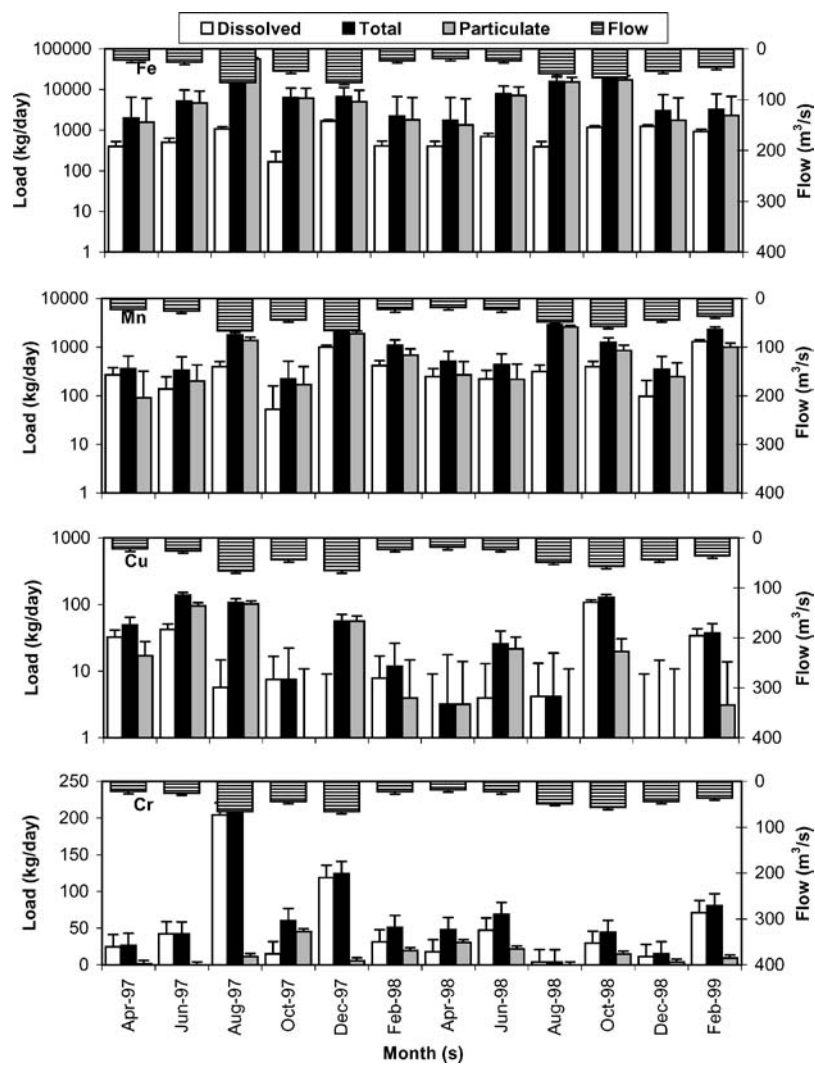


Figure 4. Monthly observed load and flow variation at downstream site of the river Hindon, Mohannagar.

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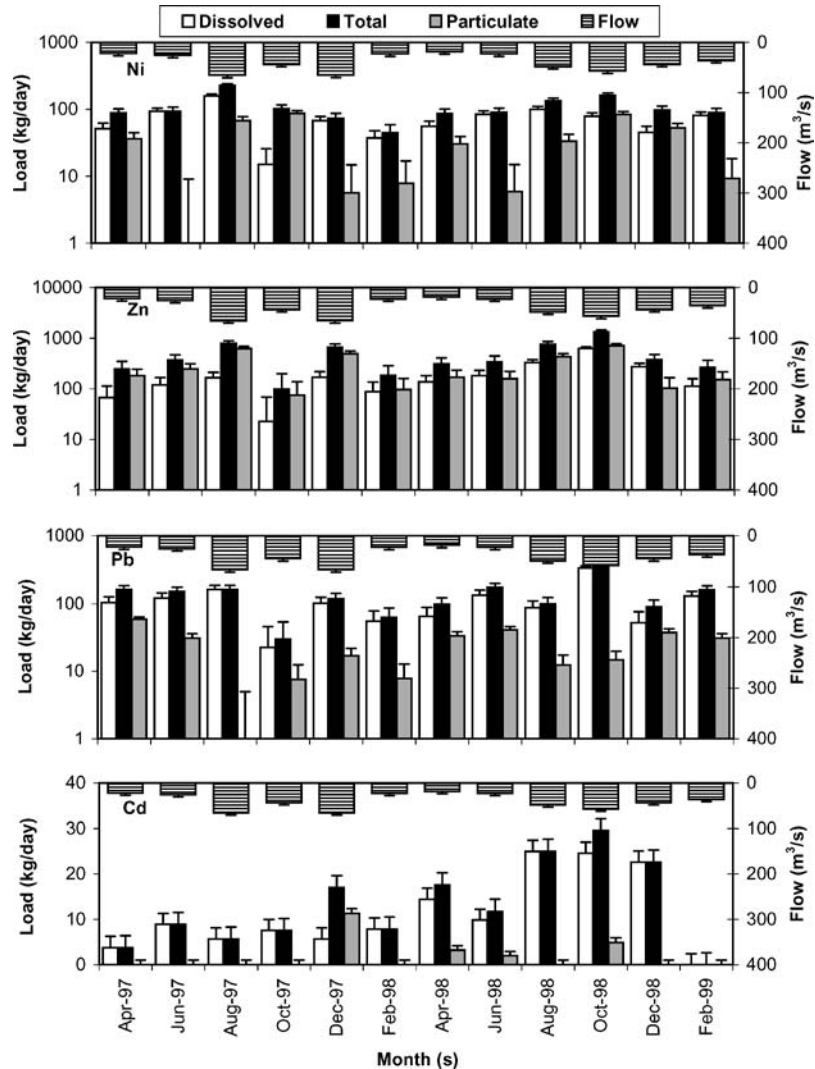


Figure 4. (Continued)

transported load. Calculations were carried out for the monsoon month contribution (Table V). Out of the transported load at Mohannagar by river Hindon, monsoon month contributed dissolved load about 29 to 65% during the year 1997–98 and 7 to 54% during the year 1998–99 while that of particulate load about 18 to 85% during the year 1997–98 and 22 to 70% during the year 1998–99. Comparing two periods (Table IV), it is observed that although the loads were more in the second year (April 1998 to March, 1999) than the first year (April 1997 to March 1998), but the monsoon month contribution in the second year (April 1998 to March, 1999) is

TABLE V

Annual heavy metal loads transported by river Hindon at downstream site (Mohannagar) during the study period from April 1997 to March 1999

	Zn	Ni	Pb	Cu	Fe	Mn	Cr	Cd
April, 1997 to March, 1998								
Dissolved load (ton)	50	35	46	8	322	167	35	3
Particulate load (ton)	135	15	9	23	6564	336	6	1
Total load (tons)	185	50	55	31	6886	504	41	4
Monsoon month contribution (%)								
Dissolved load	52	65	56	56	45	29	64	43
Particulate load	58	41	31	78	85	43	18	Nil
April, 1998 to March, 1999								
Dissolved load (ton)	192	50	87	15	493	241	16	12
Particulate load (ton)	204	24	17	5	5835	685	7	1
Total load (tons)	396	74	103	20	6328	926	24	13
Monsoon month contribution (%)								
Dissolved load	41	54	35	7	30	33	36	47
Particulate load	45	27	40	48	60	70	33	22
Net load (ton) from April, 1997 to March, 1999								
Dissolved	242	85	133	23	814	408	52	15
Particulate	339	40	26	28	12399	1021	13	2
Total	581	124	159	52	13213	1429	65	16

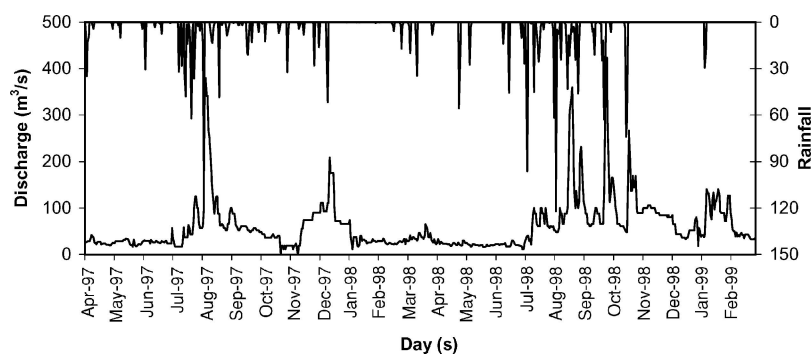


Figure 5. Daily rainfall and discharge pattern at downstream site of river Hindon at Mohannagar.

less than the first year (April 1997 to March 1998). Further, the first period (April 1997 to March 1998) had lesser rainfall (1050 mm) as compared to the second period (April 1998 to March, 1999) having rainfall (1276 mm). Also, out of the total rainfall occurred during two consecutive year, 63% rainfall received in monsoon months (June to September) in the first study period (April 1997 to March 1998) while 76% rainfall occurred in the second period (April 1998 to March, 1999).

TABLE VI
Heavy metal flux (in tons) by major Indian rivers

	Fe	Mn	Zn	Pb	Cu	Cr	Ni	Source
Brahmaputra	65×10^6	2.7×10^6	5.5×10^5	—	6.4×10^4	1.3×10^4	1.1×10^5	Subramanian (1979)
Ganges	30×10^6	1.1×10^6	6.0×10^5	—	8.2×10^4	8.6×10^4	4.5×10^4	Abbas and Subramanian (1984)
Ganges	4.1×10^6	5.6×10^4	1.4×10^5	1.4×10^4	0.7×10^4	1.5×10^4	5×10^4	Modak <i>et al.</i> (1990)
Godavari	9.7×10^6	1.8×10^5	9180	1870	1.310^4	2.1×10^4	8670	Biksham and Subramanian (1988)
Mahanadi	8.0×10^5	7240	1590	929	803	331	110	Chakrapani and Subramanian (1990)
Krishna	5.4×10^5	1.0×10^4	1112	185	908	1212	732	Ramesh and Subramanian (1988)
Cavery	4.4×10^4	923	355	28	43	107	107	Subramanian (1978)
Hindon	6607	715	290	79	26	32	62	Present study

— = data not available.

It may be inferred that the monthly load variations were strongly dependent on rainfall and consequently, with diffuse pollution from soil.

Further, annual heavy metal fluxes from Hindon river are compared with those other rivers from Indian sub-continent into the Bay of Bengal (Table VI). For individual heavy metals such as Pb, the contribution of the river Hindon to the chemistry of the Bay of Bengal could be significant.

5. Conclusion

The river Hindon transports significant amount of metal load to the river Yamuna. Out of eight point sources contributing to the river Hindon, river Kali contributes more than 50% of total metallic load to the river Hindon. The metallic load is related to the flow, thereby particulate matter and rainfall. Monsoon months contributes significant load to the river. The contribution of river Hindon could be significant in comparison of other rivers of India specially from Pb metal fluxes.

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