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HYDROCHEMISTRY OF DISSOLVED METALS IN YAMUNA RIVER AROUND INDUSTRIAL HUB OF HIMACHAL PRADESH, INDIA

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ABSTRACT

The Yamuna river flows through Sirmour district in Himachal Pradesh (India) and receives domestic and industrial effluents from different villages and Ponta-sahib industrial hub situated on its bank. Ninety six surface water samples were collected from eight sites from the river during 2014 and 2015. Water quality with respect to 9 heavy metals (Cr, Cu, As, Cd, Ni, Pb, Mn, Zn & Fe) and 3 non-metals (NO_3, Mg & Ca) was assessed for their variations, source apportionment and pollution indexing. Principal component analysis (PCA) outcome of different factors indicated natural and anthropogenic activities as contributing factors for metals profusion in the river. During 2014, three principal components were identified to be responsible for river water deterioration and accounted for 94.73% of overall total variance. PCA_1 in 2014 accounted for 70.16% of total variance and was characterized by high loading for Cr, Cu, NO_3, As, Mn, Zn, Fe, Mg and Ca. River stretch during 2015 showed high pollution load and two principal components were identified responsible for the impairment of river water which accounted for 88.67% of the overall total variance. PCA_1 in 2015 accounted for 75.74% total variance and registered high loading for Cr, Cu, As, Ni, Mn, Zn, Fe, Mg and Ca. Based on pooled pollution index, in river water Cr, As and Zn were seriously (24.74), moderately (0.616) and slightly (0.380) polluting metals, respectively. Although river quality does not pose any serious threat to human health still various activities domiciled at downstream need to be discontinued to restore water quality.

Key words: Dissolved metals, pollution index, principal component analysis, Yamuna

INTRODUCTION

Water is a universal solvent on the Earth and one of the important needs of life. River water quality monitoring is necessary as they are threatened by pollution resulting from various human activities along the river course (Kashyap *et al*., 2015). Heavy metal contamination in river is one of the major quality issues in many fast growing cities (Amadi *et al*., 2010). Heavy metals contamination is important due to their potential toxicity to the environment and human beings (Vinodhini and Narayanan, 2008). Metals such as Cu, Fe, Mn, Ni and Zn are essential as micronutrients in animals and plants while many other metals such as Cd, Cr, Pb and Co have no known physiological role

(Suthar and Singh, 2008). Metals are non-degradable and can accumulate in human body system, causing damage to nervous system and internal organs (Sharma and Kansal, 2011). The metals enter into river water through various activities like mine discharge, run-off, chemical weathering of rocks, soils, wet and dry fallout of atmospheric particulate matter, etc. (Venugopal *et al*., 2009). They also enter from industrial areas *via*. discharge of untreated industrial effluents (Singh *et al.*, 2008). In India, the rivers passing through urban areas are frequently associated with water quality problems. Thus practice of discharging untreated domestic and industrial waste water into the rivers often leads to intrusion of dissolved metals into the rivers (Bhardwaj and Singh, 2011, Patil *et al*., 2012; Gupta *et al*., 2013). Further, rivers play major role in the assimilation or transportation of municipal, industrial wastewater/effluents, runoff from agricultural and mining areas (Singh *et al*., 2008). These anthropogenic activities result in toxic metal contaminations.

The Yamuna is the largest tributary river of the Ganges (Ganga) in northern India which originates from Yamunotri glacier at a height 6,387 m on south-western slopes of Banderpooch peaks. It has been acclaimed as a holy river in Indian mythology and various pilgrimage centers e.g. Yamunotri (Uttaranchal), Paonta Sahib (Himachal Pradesh), Mathura, Vrindavan, Bateshwar and Allahabad (all in Uttar Pradesh) are located on the banks of this river. On the basis of hydrological and ecological conditions, Yamuna has been classified into five segments that are Himalayan segment, Upper segment, Delhi segment, Eutrophicated segment and Diluted segment (Mishra, 2010). Several studies revealed that Yamuna is highly polluted river of India (Jha *et al*. 1990 Bhargava, 2006; Agarwal, 2009; Mishra, 2010). In Himachal Pradesh the disposal of untreated domestic and industrial wastes around Pontasahib, situated on the bank of Yamuna river, is aggravating the pollution problem. Considering the implications of water pollution on human and aquatic health, the effective management of polluted segment of the river is of prime importance. So far no systematic study has been undertaken to critically assess the water quality with respect to dissolved metals in the river stretch. The present study was undertaken in the Himalayan segment of river Yamuna flowing through Pontasahib, a major industrial town, to evaluate the spatial variations in water quality.

MATERIALS AND METHODS

Sites, sample collection and laboratory analysis

Eight sampling stations (Barotiwala, Rampur ghat, Kunja, Pontasahib, Gurudwara, Kedarpur, Ganguwala and Satiwala lying in between 30^{o} 24'40.63" - 30^{o}26'43.51" N and 77^{o}34' 44.67" - 77^{o} 40'20.19" E at an altitude of 369 - 400 m amsl) were selected along Pontasahib industrial hub/municipal

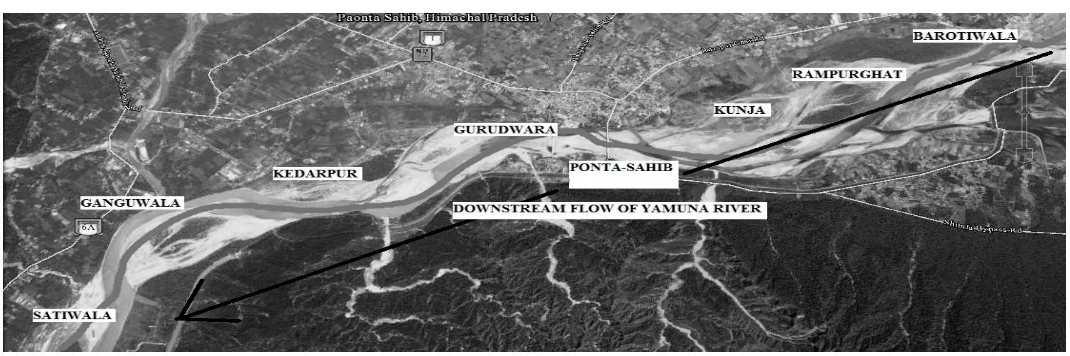

Fig. 1: Location of study area showing stretch of Yamuna river along Pontasahib and water sampling locations (source: *www.googlearth.com*)

town to assess the overall river quality. Water quality was monitored once in pre-monsoon and post-monsoon for a period of two years (2014-2015) by taking samples from each sampling station (Fig. 1). Three replicates were taken from each sampling station. Ninety six water samples were taken 5 to 10 cm below the water surface using acid washed plastic containers to avoid any changes in river quality parameters (APHA, 2011). The samples for dissolved metal analysis were collected and acidified at site (pH < 2) with conc. HNO_3. All the samples, collected in tight capped high quality polyethylene bottles, were immediately transported to the laboratory in ice-boxes for analysis.

For heavy metals analysis, water samples (200 mL) were digested in 5 mL di-acid mixture (HNO_3:$HClO_4$ = 3:1 ratio) on a hot plate and filtered through Whatman No. 1 filter paper and the final volume made to 50 mL by double distilled water. The analysis of metals *viz.*, Fe, Zn, Mn, Cd and As was done using inductively coupled plasma emission spectrophotometer [ICP-ES- 6000 series] (APHA, 2011; Kashyap and Verma, 2015). The methodology followed for ICP-ES was as per Kashyap *et al.* (2015). Cr, Cu, NO_3, Ni, Pb, Ca and Mg were analyzed photo-metrically using Spectroquant Pharo-300 [Merck, India] (APHA, 2011).

Statistical analysis

The samples were analyzed using statistical program XLSTAT-2012 to calculate mean concentrations, ranges, standard deviations, total seasonal mean concentrations and correlation coefficient (r-values) of dissolved metals in water samples collected in two years. Significance was tested at 5% level. The principal component analysis (PCA) was performed on normalized (z-scale transformation) on 12 variables after sorting out the highly correlated variable from the data sets individually for each year. The Bartlett's sphericity test was applied to the correlation matrix of variables for assessing the adequacy of PCA in water quality (Lambarkis *et al*., 2004). The principal components with eigen values > 1 were retained and used to assess the compositional and spatial variations in river quality due to anthropogenic activities domiciled along with the river course (Sundaray, 2009; Amadi *et al*., 2010).

Pollution index

Pollution index (PI) is a technique which shows the pollution status of water/soil quality parameters. It identifies the contribution of individual parameters on overall water quality (Aboud and Nandini, 2009). The pollution index was calculated as per the formula of Amadi (2012).

$$\mathrm{PI} = \frac{\sqrt{\left[\frac{Ci\ max}{Si}\right]^2 - \left[\frac{Ci\ min}{Si}\right]^2}}{2}$$

Where PI = pollution index; Ci = respective maximum and minimum concentration of quality parameters; Si = standard permissible limit of surface water quality. The water quality and its suitability for drinking purpose were examined by determining its metal pollution index (Prasad and Kumari, 2008). The interpretation of PI was based on different classes and their values i.e. class-I = <0.3 (non-polluting), class-II = 0.3-0.5 (slightly polluting), class-III = 0.5-1.0 (moderately polluting), class-IV = 2-3 (strongly polluting) and class-V = >5 (seriously polluting) given by Amadi (2012).

RESULTS AND DISCUSSION

Spatial distribution of dissolved metals in river Yamuna

Chromium (Cr), copper (Cu) and nitrate (NO_3): Copper, despite its toxic effects is reckoned as an essential component of many enzymes. Cu and Cr reach the aquatic environment chiefly from mines, agricultural chemicals, paints, etc. Overall mean Cr content in river water was 0.016 and 0.015 mg

Table 1: Spatial variations of dissolved metals (mg L^{-1}) in water samples at different locations of Yamuna river during 2014

	Locations of water sampling along river stretch								Standards		Grand mean
Parameters	L1	L2	L3	L4	L5	L6	L7	L8	CPCB*	WHO*	±SD
Cr	BDL	0.002	0.005	0.007	0.027	0.020	0.035	0.033	0.1	0.01	0.016 ±0.014
Cu	0.007	0.008	0.007	0.006	0.010	0.010	0.015	0.018	3.0	0.05	0.010 ±0.004
NO_3	7.617	10.000	10.333	13.875	13.000	12.833	14.833	15.500	NA	45.0	12.249 ±2.699
As	0.123	0.128	0.137	0.150	0.150	0.155	0.155	0.167	0.2	0.10	0.146 ±0.015
Cd	0.005	0.005	0.006	0.005	0.004	0.005	0.005	0.006	2.0	0.01	0.005 ±0.001
Ni	0.020	0.010	0.020	0.020	0.010	0.020	0.020	0.030	2.0	0.20	0.021 ±0.004
Pb	0.027	0.017	0.021	0.027	0.015	0.019	0.020	0.021	0.1	0.05	0.021 ±0.004
Mn	0.027	0.030	0.033	0.036	0.039	0.044	0.050	0.052	2.0	0.05	0.039 ±0.009
Zn	1.947	1.998	2.163	2.212	2.273	2.423	2.507	2.537	5.0	5.0	2.258 ±0.221
Fe	0.068	0.088	0.112	0.133	0.140	0.162	0.182	0.205	1.0	NA	0.136 ±0.046
Mg	15.717	15.830	14.868	16.702	17.355	21.238	23.657	26.898	200.0	NA	19.033 ±4.391
Ca	14.687	14.280	15.537	17.760	24.263	31.763	40.025	42.758	150.0	NA	25.134 ±11.653

L_1 - Barotiwala, L_2 - Rampurghat, L_3 - Kunja, L_4 - Pontasahib, L_5 - Gurudwara sahib, L_6 - Kedarpur, L_7 - Gunguwala, L_8 - Satiwala, BDL - Below detectable limit, NA - Not available, SD - standard deviation

L^{-1} during 2014 and 2015, respectively, while similar mean concentrations of Cu 0.010 mg L^{-1} were recorded, irrespective of years. Mean contents of NO_3 in river water were 12.25 and 14.15 mg L^{-1} in 2014 and 2015, respectively (Table 1 and 2). During 2014, highest Cr content of 0.035 mg L^{-1} was observed at Ganguwala, whereas higher Cu (0.018 mg L^{-1}) and NO_3 (15.50 mg L^{-1}) was observed at Satiwala. During 2015, maximum Cr (0.035 mg L^{-1}) was also noticed at Ganguwala while Cu was higher at Satiwala (0.013 mg L^{-1}) and NO_3 (15.17 mg L^{-1}) was higher at Kedarpur. Upstream from Satiwala (Pontasahib industrial town) there was more discharge of untreated industrial effluents, sewage and agricultural runoff into river. Thus slight increase in chromium, copper and nitrate content was noticed at downstream locations (L6, L7 and L8). Suthar and Singh (2008) reported that in agricultural practices urea, copper sulphate is widely used and it may be one of the reasons for alarming concentration of nitrate and copper in water bodies. Dissolved metals were below permissible limits of surface water quality (CPCB, 2010).

Table 2: Spatial variations of dissolved metals (mg L^{-1}) in water samples at different locations of Yamuna river during 2015

	Locations of water sampling along river stretch								Standards		Grand mean
Parameters	L1	L2	L3	L4	L5	L6	L7	L8	CPCB*	WHO*	±SD
Cr	0.002	0.005	BDL	0.007	0.022	0.018	0.035	0.033	0.1	0.01	0.015 ±0.014
Cu	0.010	0.009	0.009	0.008	0.010	0.010	0.012	0.013	3.0	0.05	0.010 ±0.002
NO_3	14.500	13.333	12.333	14.417	14.833	15.167	14.333	14.167	NA	45.0	14.135 ±0.902
As	0.075	0.080	0.080	0.082	0.102	0.103	0.115	0.120	0.2	0.10	0.095 ±0.018
Cd	0.001	0.002	0.001	0.001	0.000	0.001	0.001	0.004	2.0	0.01	0.001 ±0.001
Ni	0.020	0.016	0.019	0.022	0.022	0.025	0.026	0.030	2.0	0.20	0.022 ±0.005
Pb	0.041	0.043	0.039	0.039	0.041	0.040	0.041	0.020	0.1	0.05	0.038 ±0.008
Mn	0.017	0.017	0.018	0.022	0.022	0.027	0.025	0.025	2.0	0.05	0.022 ±0.004
Zn	1.467	1.488	1.375	1.507	1.547	1.585	1.662	1.693	5.0	5.00	1.540 ±0.105
Fe	0.068	0.073	0.107	0.112	0.132	0.160	0.188	0.213	1.0	NA	0.132 ±0.052
Mg	13.115	14.088	14.600	16.040	18.183	19.642	22.137	23.872	200	NA	17.710 ±3.932
Ca	18.647	19.262	17.737	20.300	26.282	26.035	29.162	31.732	150	NA	23.644 ±5.329

L_1 - Barotiwala, L_2 - Rampurghat, L_3 - Kunja, L_4 - Pontasahib, L_5 - Gurudwara sahib, L_6 - Kedarpur, L_7 - Gunguwala, L_8 - Satiwala, BDL - Below detectable limit, NA - Not available, SD - standard deviation

Arsenic (As), cadmium (Cd) and nickel (Ni): The mean As content in river water was 0.146 mg L^{-1} and 0.095 mg L^{-1} during 2014 and 2015, respectively. The As content was maximum at Satiwala i.e. 0.167 mg L^{-1} and 0.120 mg L^{-1} in 2014 and 2015, respectively. The significant increase in As content may be attributed to natural geogenic sources or is possibly aggravated by over powering aquifers by phosphorus from different chemical fertilizers, industrial and sewage effluents (Macklin *et al*. (2003). Landfill leachates also serve as an important source of As in the environment (Vinodhini and Narayanan, 2008). Cd is a poisonous metal and may cause serious health problems even if ingested in small amounts. Effluents from industries making batteries, dyes, pigments and alloys are major sources of Cd in water bodies (Kashyap *et al*., 2015). Mean Cd content in Yamuna water was 0.005 and 0.001 mg L^{-1} during 2014 and 2015, respectively. The maximum Cd content during 2014 (0.006 mg L^{-1}) was observed at Satiwala and Kunja, whereas in 2015 it was at (0.004 mg L^{-1}) at Satiwala. Exceptionally, irrespective of the years, Ni content was high (0.030 mg L^{-1}) at Satiwala. The mean Ni content in river water was 0.021 and 0.022 mg L^{-1} in 2014 and 2015, respectively (Table 1 & 2). The aggravated concentrations of Ni and Cd (downstream) might be ascribed to the industries particularly those dealing with electrical goods, household appliances, catalysts, pigments, paints and batteries which appeared to have acted as potential sources of Ni and Cd contamination in river Yamuna (Bhardwaj and Singh, 2011).

Lead (Pb), manganese (Mn) and zinc (Zn): The major industrial sources for Pb and Mn are iron and steel related industries, power plants and coke ovens, paper and newsprint mills, manufacturing of heating equipment, fossil fuel power generation and welding industries (Bhardwaj and Singh, 2011). In river Yamuna overall mean Pb content in 2014 and 2015 was 0.021 and 0.038 mg L^{-1}, respectively; whereas mean Mn content in 2014 was 0.039 mg L^{-1} and in 2015 it was 0.022 mg L^{-1}. Maximum Pb content (0.027 mg L^{-1}) was at Barotiwala and Pontasahib during 2014. In 2015 the maximum Pb (0.043 mg L^{-1}) was at Rampurghat (Table 2 & 3). Various units of newsprint, iron-ferro-alloy industries and dumping of treated waste occur near the river bank which could possibly be the reason for high Pb values at upstream (L1 and L2) in Yamuna river. On the other hand, mean Zn content was 2.258 mg L^{-1} in 2014 and 1.540 mg L^{-1} in 2015 (Table 1 & 2). Zn is an essential micronutrient required in traces to sustain life, however its high concentration in water bodies may prove toxic to aquatic flora and fauna (Kashyap and Verma, 2015; Kashyap *et al*., 2015). It is a common contaminant in industrial effluents (Gupta *et al*., 2011).

Calcium (Ca), magnesium (Mg) and iron (Fe): Ca and Mg are most abundant ions in fresh water and are important in shell construction, bone building and precipitation of lime. Both enter in river waters through surface run-off from agricultural land use. Mg is often associated with Ca in all kinds of waters, but its concentration remains generally lower than Ca. The mean Ca and Mg contents in river stretch were (25.13 mg/L^{-1}; 23.64 mg/L^{-1}) and (19.03 mg/L^{-1}; 17.71 mg L^{-1}) during 2014 and 2015, respectively (Table 1 & 2). Within the sites studied, the maximum Ca during 2014 and 2015 (42.75 mg/L^{-1} and 31.73 mg L^{-1}) was at Satiwala. Similarly, maximum Mg contents during 2014 and 2015 (26.89 mg/L^{-1} and 23.87 mg L^{-1}) was also at Satiwala. Hardness of water in river is directly related to the amount of Ca and Mg present in water. Ca and Mg content of river did not exceed 120 mg L^{-1} so water was soft in nature which is in agreement with Dwivedi and Pathak, (2007) and Patil *et al*.(2012), who studied abundance of these ions in fresh waters. Fe in water may be present in varying quantities depending upon the geology of the area. Further, the ferrous and ferric ions are the primary forms of concern in aquatic environment (Kashyap *et al.*, 2015). The mean Fe content in river water during 2014 and 2015 was 0.136 and 0.132 mg L^{-1}, respectively. Gupta *et al*. (2013) have reported similar results in Yamuna river at Agra, India. The water runoff may carry higher concentrations of these metals, which arise due to municipal waste water disposal, use of chemical fertilizers and pesticides, etc. (Avvannavar and Shrihari, 2008).

Principal component analysis

During 2014, three principal components were identified to be responsible for the deterioration of river water which accounted for 94.73% of the overall total variance. The first principal component (PCA_1) accounted for 70.16% of the total variance and showed high loads of Cr, Cu, NO_3, As, Mn, Zn, Fe, Mg and Ca (Table 3). The presence of these elements showed some degree of mineralization in river water due to high rates of loading and dissolution processes (Prasad and Kumari, 2008). The second principal component (PCA_2) accounted for 15.47% and comprised of Ni and Pb with moderate loadings. Naturally, chemical weathering of rock-forming minerals may be a source of their enrichment in surface water (Mohan *et al*., 1996). Third principal component (PCA_3) noticed less loading and consisted of only Cd which accounted for 9.16% of overall total variance.

Table 3: Principal component analysis of data set of water quality during 2014 and 2015

Dissolved metals	2014 PCA_1	2014 PCA_2	2014 PCA_3	2015 PCA_1	2015 PCA_2
Cr	**0.330**	-0.092	-0.132	**0.317**	0.091
Cu	**0.308**	0.040	0.239	**0.280**	-0.139
NO_3	**0.312**	0.070	-0.197	0.155	**0.580**
As	**0.323**	0.096	-0.114	**0.323**	0.047
Cd	0.046	0.085	**0.918**	0.205	**-0.558**
Ni	-0.124	**0.683**	-0.064	**0.317**	0.021
Pb	-0.124	**0.683**	-0.064	-0.231	**0.489**
Mn	**0.343**	0.049	-0.041	**0.285**	0.268
Zn	**0.336**	0.058	-0.058	**0.317**	0.083
Fe	**0.339**	0.075	-0.020	**0.319**	-0.011
Mg	**0.323**	0.141	0.130	**0.327**	0.013
Ca	**0.335**	0.068	0.026	**0.326**	0.068
Eigen value	8.413	1.856	1.099	9.089	1.553
Variability (%)	70.105	15.468	9.159	75.742	12.940
Cumulative %	70.105	85.574	94.732	75.742	88.682

Compared to the year 2014, the river stretch during 2015 showed high pollution load as the two principal components were found responsible for the impairment of river water which accounted for 88.67% of overall total variance. PCA_1 accounted for 75.74% of total variance and showed high loading for Cr, Cu, As, Ni, Mn, Zn, Fe, Mg and Ca (Table 3). Especially this year the disposal of untreated domestic sewage from villages, effluents from industrial area and chemical weathering may have been the major point sources of pollution for these heavy metals in river water (Mohan *et al*., 1996). At different sampling points PCA_2 showed very less loading of NO_3, Cd and Pb in water along the river course registered with 12.94% of overall total variance.

Correlation coefficients (r-values)

Different dissolved metal attributes (Cr, Cu, As, Mn, Zn Fe, Mg and Ca) of water between 2014 and 2015 registered positive/highly significant correlation (r-values = 0.979, 0.926, 0.879, 0.906, 0.826, 0.986, 0.961 and 0.965), respectively. It showed that these metals observed with almost similar concentrations during each year. However, other metals *viz*., NO_3, Ni and Pb showed non-significant correlation, which implied that metals were recorded with different concentrations during 2014 and 2015. The results indicated that the concentrations of heavy metals in water affected the quality of river water during the study period. Giri and Singh, (2014) reported similar results in their studies on heavy metal pollution in Subarnarekha river.

Classification based on Pollution index (PI)

Based on pooled PI of two years, Cr, As and Zn were found seriously (24.74), moderately (0.616) and slightly (0.380) polluting metals, respectively; whereas other metals were non-polluting (Table 4). Results were in line with Mishra (2010) who reported similar high metal content in other segments of Yamuna (Upper, Eutrophicated and Delhi segment). During each year's data analysis of

Table 4: Calculated pollution index of pooled values during 2014 and 2015

Dissolved metals	Pollution index values	Status
Cr	24.74	Seriously polluting
Cu	0.004	Non polluting
No_3	0.289	Non polluting
As	0.616	Moderately polluting
Cd	0.002	Non polluting
Ni	0.012	Non polluting
Pb	0.274	Non polluting
Mn	0.016	Non polluting
Zn	0.380	Slightly polluting
Fe	0.155	Non polluting
Mg	0.103	Non polluting
Ca	0.144	Non polluting

PCA verified results of pollution index that Cr, As and Zn metals were polluting in river segment keeping these metals in first principal component (PCA_1).

Comparison with standard limits

In present study amongst the dissolved metals only Cr and As were above the permissible limits for drinking water (WHO, 2010) and the rest metals were within the permissible limits. The people in the study area do not use river water for drinking purpose, so dissolved metals in water do not pose any health concern to the human. The regular monitoring of metals needs to be undertaken by the government agencies. Earlier studies on heavy metal contamination in river Yamuna in Haryana and Delhi states of India have indicated that river water possessed heavy metals. Textile, engineering, chemical, electrical and electronic industries contribute to metals deposition in river water. Several metals present in industrial effluents and municipal sewage around Haryana find their way into river Yamuna (Agarwal, 2009). Jha *et al*. (1990) reported metals like Fe (0.07 - 0.94 mg L^{-1}) in upper Delhi segment, Ni (0.05 - 0.25 mg L^{-1}) near Sonepat-Gohana industrial area and lead (0.12 - 0.15 mg L^{-1}) from Hathnikund to Pallaghat area.

Conclusion: The concentrations of dissolved metals (Cr, Cu, As, Cd, Ni, Pb, Mn, Zn and Fe) and non-metals (Mg, Ca and NO_3) in surface water of river Yamuna at different locations showed significant spatial variations. Irrespective of year and location of sampling, elemental concentrations differed from each other. When compared with the drinking water guidelines established by WHO, in India much attention needs to be paid to Cr and As elements, though their concentrations were slightly above permissible limits. In fact, concentrations of heavy metals were below permissible limits of surface water quality given by CBCB, India. The pollution index suggested that Yamuna river water was slightly affected, except with respect to Cr. During 2014 and 2015 three and two possible sources of pollution to river water were revealed by PCA which were categorized into natural and anthropogenic factors. The augmented concentrations of dissolved metals in selected stretch of river Yamuna in Himalayan segment is greatly affected by direct discharge of industrial, municipal and sewage water. Thus adequate management planning needs to be formulated to limit the intrusion of dissolved metals from different point or non-point sources.

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