



Assessment of heavy metal pollution in water and surface sediment and evaluation of ecological risks associated with sediment contamination in the Ganga River: a basin-scale study

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Abstract

We investigated eight heavy metals (Cr, Cd, Cu, Ni, Pb, Zn, Mn, and Fe) in water and bed sediment at 9 study sites along with 2320 km stretch of the Ganga River. Principal component analysis (PCA) and indices such as geo-accumulation index (I_{geo}), contamination factor (CF), enrichment factor (EF), pollution indices, and sediment quality guidelines were used to assess source apportionment and magnitude of contamination. Concentrations of Cr, Cd, Pb, Ni, Cu, and Fe in water have exceeded their respective standards in the middle and lower reaches of the river. Sediment Cr and Ni have reached probable effective concentration (PEC) at Kannauj, imposing likely threats to sediment dwellers. Highest I_{geo} values were recorded for Cr, Cd, and Pb at Kannauj, Rajghat, and Howrah. We further tested ecological risks (E_r) and potential ecological risks (PERI) to assess individual and cumulative effects and found the Kannauj, Rajghat, and Howrah sites under the high-risk category. The modified pollution index (MPI) and the modified degree of contamination (mCd) also revealed the middle and lower river reaches under moderately to the heavily polluted category. Our study provides the first detailed watershed-scale database on heavy metal concentration in water and bed sediment, the magnitude of contamination, and likely ecological risks to aquatic organisms in the Ganga River. Given that the Ganga water is used for drinking and irrigation and the river harbors a diversity of habitats for fisheries, the study merits attention from a human health perspective as well.

Keywords Ecological risk · Effective concentration · Ganga River · Heavy metal · Pollution indices · Sediment quality guidelines

Introduction

Urban-industrial-driven economic development has dramatically altered aquatic environments with increasingly high input of metals and other pollutants. Terrestrially derived metals, of geogenic or anthropogenic origin, drain into rivers and streams and accumulate in sediments (Omwene et al. 2018).

Bottom sediment regulates trophic status (Singh et al. 2005; Suresh et al. 2015), ecological cycling, and detritus food web in aquatic ecosystems (Burton Jr 2002). Metal contamination of the aquatic environment has received global attention due to their abundance, persistence, long-lasting availability, and toxicity (Dong et al. 2011; Li et al. 2014). Sediment acts as an ultimate receptor of pollutants and a potential secondary source to overlying water. Re-suspension and feedbacks reload the toxicants to overlying water generating one of the most critical problems in the aquatic environment (Zhang et al. 2014). River sediment provides important cues for the assessment of pollution sources, river health, and ecological risks (Praveena et al. 2015).

Ganga, the largest river system of India, traverses ~ 2525 km from Gangotri Glacier to the Bay of Bengal, and support over 400 million people of India. During the last few decades, the river experienced tremendous water quality degradation along its course (Pandey et al. 2014; Dwivedi et al. 2018). Urban-industrial discharge is the main source of

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the Ganga River pollution (CPCB 2013; Yadav and Pandey 2017). Overall, 222 towns in the basin generate ~ 8250 MLD of sewage, of which only ~ 3500 MLD is disposed after treatment. Of the total, 2460 MLD is discharged directly into the Ganga River, 4510 MLD into tributaries, and the remaining amount on land or low-lying areas and leach downstream (CPCB 2013). According to the Environmental Information System (ENVIS), total sewage generation from cities in 5 main states of the Indo-Gangetic Plains is ~ 15,435 MLD while the treatment capacity is only 3458 MLD (ENVIS 2016). Despite all efforts by the Government of India, the untreated sewage being flushed to the river has increased by over ninefold in recent years compared to 1985 (Dwivedi et al. 2018). In addition to urban sewage, hundreds of industrial units including textiles, electroplating, power plants, sugar mills, distilleries, paper and pulp, and several other directly discharge the waste into the river. About 2500 MLD industrial waste is generated in the Ganges Basin (Trivedi 2010). Middle segment (Kannauj to Varanasi) is particularly responsible for most of the industrial effluents added to the river. Tannery sector constitutes about 58% of grossly polluting industries in the middle segment. The atmospheric deposition also adds a sizeable amount of metals (Pandey et al. 2010).

Various studies have quantified metal pollution load in rivers including Gomti, India (Singh et al. 2005), Krotova, Bangladesh (Islam et al. 2015), Tigris River, Turkey (Varol 2011), Brisbane River, Australia (Duodu et al. 2016), Liaohe River, China (Ke et al. 2017), and Wen-Rui River, China (Xia et al. 2018). Studies on metal pollution in the Ganga River are either restricted to water (Pandey et al. 2010; Aktar et al. 2010) or bed sediment (Singh et al. 2003; Singh et al. 2013) or performed considering small stretches only. Watershed-scale variations in metal contamination of water and bed sediment in the Ganga River are yet to be discovered. This watershed-scale study was performed selecting 9 study sites, three in each of upper, middle, and lower parts of the Ganga River to investigate metal pollution load, possible natural and anthropogenic sources, spatiotemporal variations in water and bed sediment, and ecological risk associated with these contaminants. Simultaneous assessment of metals in water and in bed sediment can help to assess anthropogenic impacts and risks posed by waste discharged into the river. To our knowledge, there is no systematic database available, so far, addressing explicitly, on a watershed scale, the simultaneous measurement of heavy metals in water and sediments of the Ganga River. Watershed-scale study will provide detailed mapping, inter-segmental comparison, source apportionment and cues to policy planners for holistic evaluation of metal pollution, abatement, and rejuvenation of the Ganga River. Furthermore, because the Ganga River water is used for drinking and irrigation, the study has relevance in a toxicological perspective as well.

Materials and methods

Study area

The Ganges basin (1,086,000 km²) covers ~ 26.2% geographical area of India. Before entering to the Bay of Bengal, the Ganga River forms the largest mangrove-rich estuarine delta of the world. The basin has a sub-tropical to tropical monsoonal climate. The year constitutes a hot and dry summer (March to June), a moist rainy (July to October), and a cold winter season (November to February). The soils are sandy, loamy, clay, and different combinations of these. The annual rainfall ranges from 78 cm in the upper part through 144 cm in the middle stretch to 182 cm in the lower delta region. The present study was conducted at 9 study sites (Fig. 1) considering 2320 km river stretch, from Devprayag (30.14° N, 78.59° E) to Ganga Sagar (21.66° N, 88.04° E) covering over 90% of the river length.

Sampling and analysis

Sediment samples (0–10 cm depth) were collected using corers from 25 to 50 m reach in summer and winter 2016 and 2017. The samples were collected in triplicate from three sub-sites of each study site. The water samples were collected in polyethylene terephthalate (PET) bottles from the mid-stream of the river, stored at 4 °C, and transported to the laboratory for analysis.

Sediment samples were air dried, sieved (2 mm mesh), homogenized, and stored at 4 °C. A 0.5 g of sediment sample was digested in the ternary acid mixture and volume maintained to 50 ml. Water samples were filtered through 0.45-μ cellulose filter, digested in nitric acid, and filtered again. The concentration of metal was determined in atomic absorption spectrophotometer (AAnalyst 800 Perkin Elmer). Quality assurance was maintained through blank and repeated comparison with a standard. All the reagents used were of high grade and the glassware were properly rinsed with nitric acid followed by ultrapure Millipore water.

Sediment quality guidelines

We consider threshold effect concentration (TEC) below which adverse effect does not occur, and probable effect concentration (PEC) above which adverse effect does occur and often frequently, as criteria sediment quality guidelines (MacDonald et al. 2000).

Heavy metal pollution indices

Geo-accumulation index

The I_{geo} used to assess metal pollution was calculated using the following empirical relations:

$$(I_{geo}) = \log_2 \left(\frac{C_n}{1.5B_n} \right) \quad (1)$$

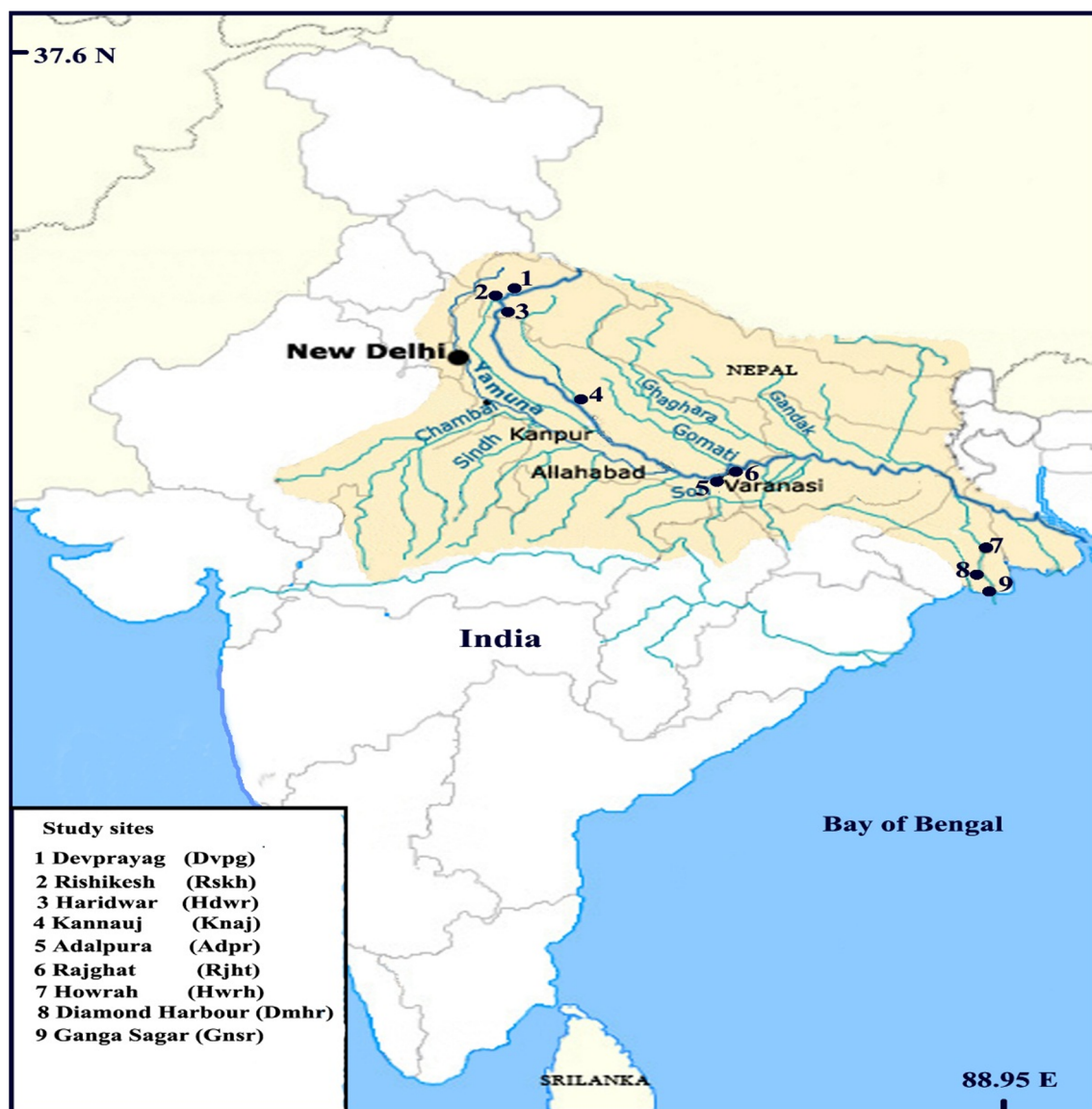


Fig. 1 Map showing the Ganges Basin and sampling stations (1–9) on the Ganga River

where C_n is the concentration of study metal and B_n is the background concentration. For the background, we considered Devprayag as reference site (Pandey and Yadav 2017). Factor 1.5 was used for the background matrix correction and reducing lithogenic effects: $0 < I_{geo} < 1$: from uncontaminated to moderately contaminated; $1 < I_{geo} < 2$: moderately contaminated; $3 < I_{geo} < 5$: from heavily to extremely contaminated; and $5 < I_{geo}$: extremely contaminated.

Contamination factor

Contamination factor Cf_{metal} , the ratio of the concentration of individual metal with its background (Hakanson 1980), was calculated as:

$$Cf_{\text{metal}} = \frac{C_{\text{metal}}}{C_{\text{background}}} \quad (2)$$

where C_{metal} is the concentration of the metal at a site and $C_{\text{background}}$ is the background concentration of the metal: $1 < CF < 3$, moderate contamination; $3 < CF < 6$, considerable contamination; and > 6 defines very high contamination.

Pollution load index

Pollution load index was obtained as below:

$$PLI = (CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)^{1/n} \quad (3)$$

In Eq. 3, n is the number of study metals and $CF_1 \dots CF_n$ are the contamination factors of individual metal at a given site. The pollution load index (PLI) provides an assessment of overall toxicity status: 1 indicates baseline levels and above 1 indicates progressive alteration.

Enrichment factor

Enrichment factor is used to assess anthropogenic and geogenic effects. To quantify the enrichment factor (E_f), Fe was used as a conservative metal (Delgado et al. 2010; Varol and Şen 2012).

$$E_f = \frac{(C_m/C_{Fe})_{\text{sample}}}{(C_m/C_{Fe})_{\text{background}}} \quad (4)$$

where $(C_m/C_{Fe})_{\text{sample}}$ is the concentration of individual metal and that of Fe in the sample, and $(C_m/C_{Fe})_{\text{background}}$ is the reference ratio in the background (E_f 1.5–3.0: minor enrichment; 3.0–5.0: moderate; 5.0–10.0: severe; and > 10: very severe enrichment).

Ecological risks and potential ecological risks

The ecological risks and the potential ecological risks are used to assess ecological risk of metal in sediment (Hakanson 1980):

$$E_r^i = T_r^i \times \left(\frac{C_i}{C_o} \right) \quad (5)$$

$$\text{PERI} = \sum_{i=1}^n T_r^i \left(\frac{C_i}{C_o} \right) \quad (6)$$

where C_i is the concentration of metal i in sediment, C_o is the background concentration of metal, and T_r defines biological toxicity factor for individual metal. T_r was used following Suresh et al. (2012) as Cu = Pb = Ni = 5, Zn = 1, Cr = 2, and Cd = 30 (ecological risk (E_r) < 40: low risk; 40–80: moderate risk; 80–160 considerable; 160–320: high; > 320 very high risk. PERI < 150: low risk; 150–300: moderate; 300–600: considerable; and > 600: high-risk condition).

The modified degree of contamination and the modified pollution index

The limitations of single metal indices lead to the development of multi-metal indices. Two most widely used such indices, developed by Hakanson (1980) and Nemerow (1991), include the modified degree of contamination (mC_d) and the pollution index (PI). Brady et al. (2015) developed a modified pollution index (MPI) considering enrichment factor. These indices are calculated as below:

$$mC_d = \sum_{i=1}^n \frac{Cf_i}{n} \quad (7)$$

$$PI = \sqrt{\frac{(Cf_{\text{average}})^2 + (Cf_{\text{max}})^2}{2}} \quad (8)$$

$$MPI = \sqrt{\frac{(Ef_{\text{average}})^2 + (Ef_{\text{max}})^2}{2}} \quad (9)$$

where Cf_{max} , Cf_{average} , Ef_{average} , and Ef_{max} represent maximum contamination factor, average contamination factor, average enrichment factor, and maximum enrichment factor respectively. The mC_d 1.5 < mC_d < 2.0 indicates slightly polluted, 2 < mC_d < 4 moderately polluted, 4 < mC_d < 8 from moderately to heavily polluted, 8 < mC_d < 16 severely polluted, 16 < mC_d < 32 heavily polluted, and mC_d > 32 extremely polluted condition. The PI 0.7 < PI < 1 indicates slightly polluted, 1.0 < PI < 2.0 moderately polluted, 2.0 < PI < 3.0 severely polluted, and PI > 3.0 heavily polluted condition. The MPI 1.0 < MPI < 2.0 indicates slightly polluted, 2.0 < MPI < 3.0 moderately polluted, 3.0 < MPI < 5.0 from moderately to heavily polluted, 5.0 < MPI < 10.0 severely polluted, and MPI > 10 heavily polluted condition.

Statistical analysis

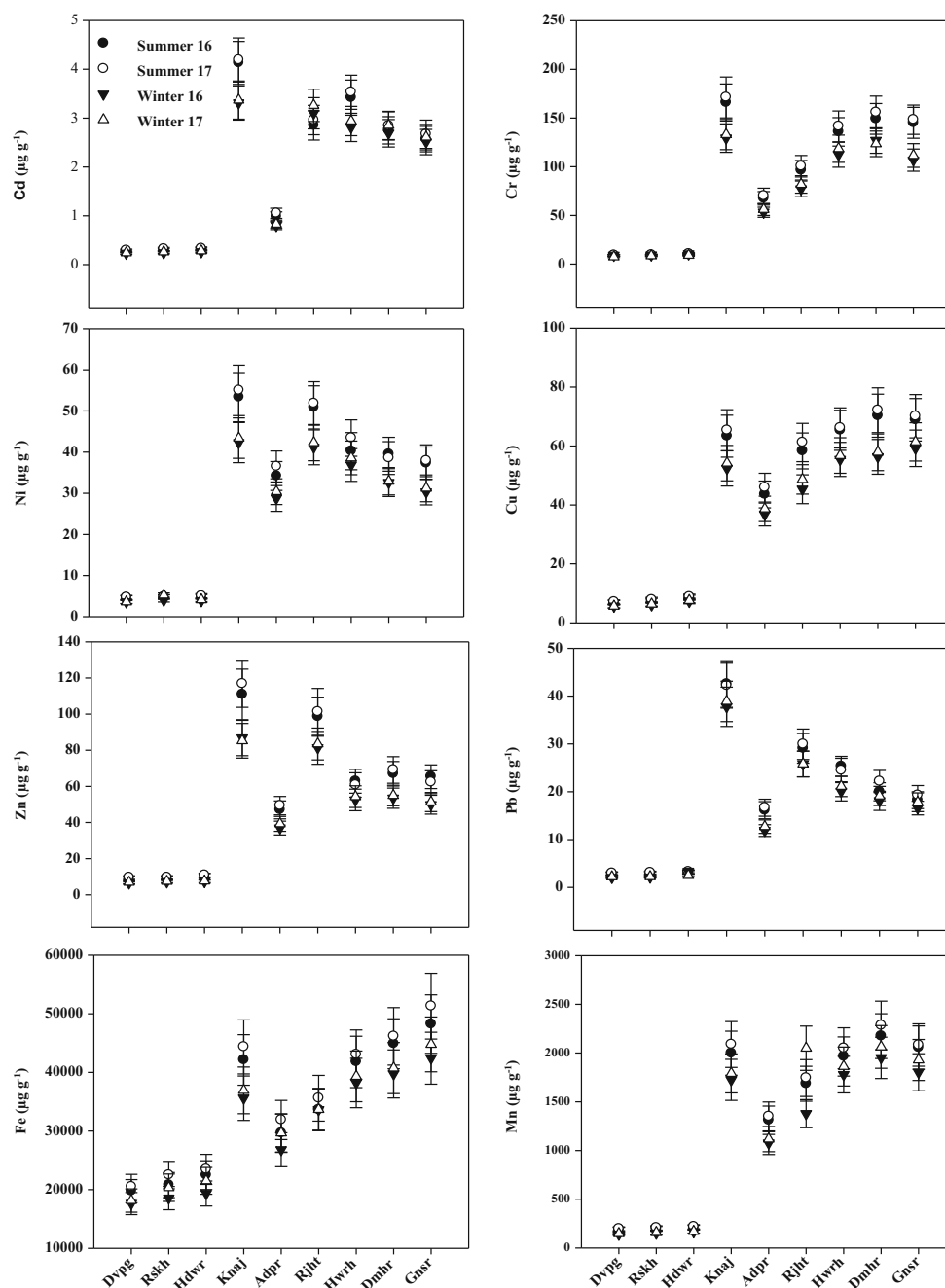
Principal component analysis (PCA) was performed to extract significant PCs and associated loadings. Correlation analysis was used to test the significant relationship between variables. Analysis of variance (ANOVA) was performed to test the significant effects of spatiotemporal determinants. Analyses were done on Excel, Sigma plot, SPSS package (version 16), and PAST (version 16).

Results

Spatiotemporal variations in heavy metals

The overall trend in the concentration of study metals in sediment did appear as Fe > Mn > Cr > Zn > Cu > Ni > Pb > Cd (Fig. 2). Except for Cr, a similar trend was observed for metals in water (Fig. 3). The concentration of Cr in sediment ranged from 7.12 to 9.13 $\mu\text{g g}^{-1}$ at Devprayag and 108.67 to 153.00 $\mu\text{g g}^{-1}$ at Ganga Sagar and was found to be the highest at Kannauj. The concentrations were lower at upper stretch sites and spatial variation was significant (ANOVA; $p < 0.05$). The concentration of Cd ranged from 0.210 to 0.29 $\mu\text{g g}^{-1}$ at Devprayag and 2.47 to 2.73 $\mu\text{g g}^{-1}$ at Ganga Sagar and was recorded highest at Kannauj. The concentration of Cu, Ni, and Pb ranged from 2.1 to 2.89, 3.54 to 4.67, and 2.05 to 3.02 $\mu\text{g g}^{-1}$ at Devprayag, and 56.87 to 68.90, 28.69 to 37.87, and 16.45 to 32.14 $\mu\text{g g}^{-1}$ at Ganga Sagar respectively. The concentrations of Fe and Mn were found highest among the study metals. On a spatial scale, Fe and Mn were found the highest at Ganga Sagar.

Fig. 2 Heavy metal concentration in the bed sediments (0–10 cm) of the Ganga River. Values are mean ($n = 12$) \pm SE

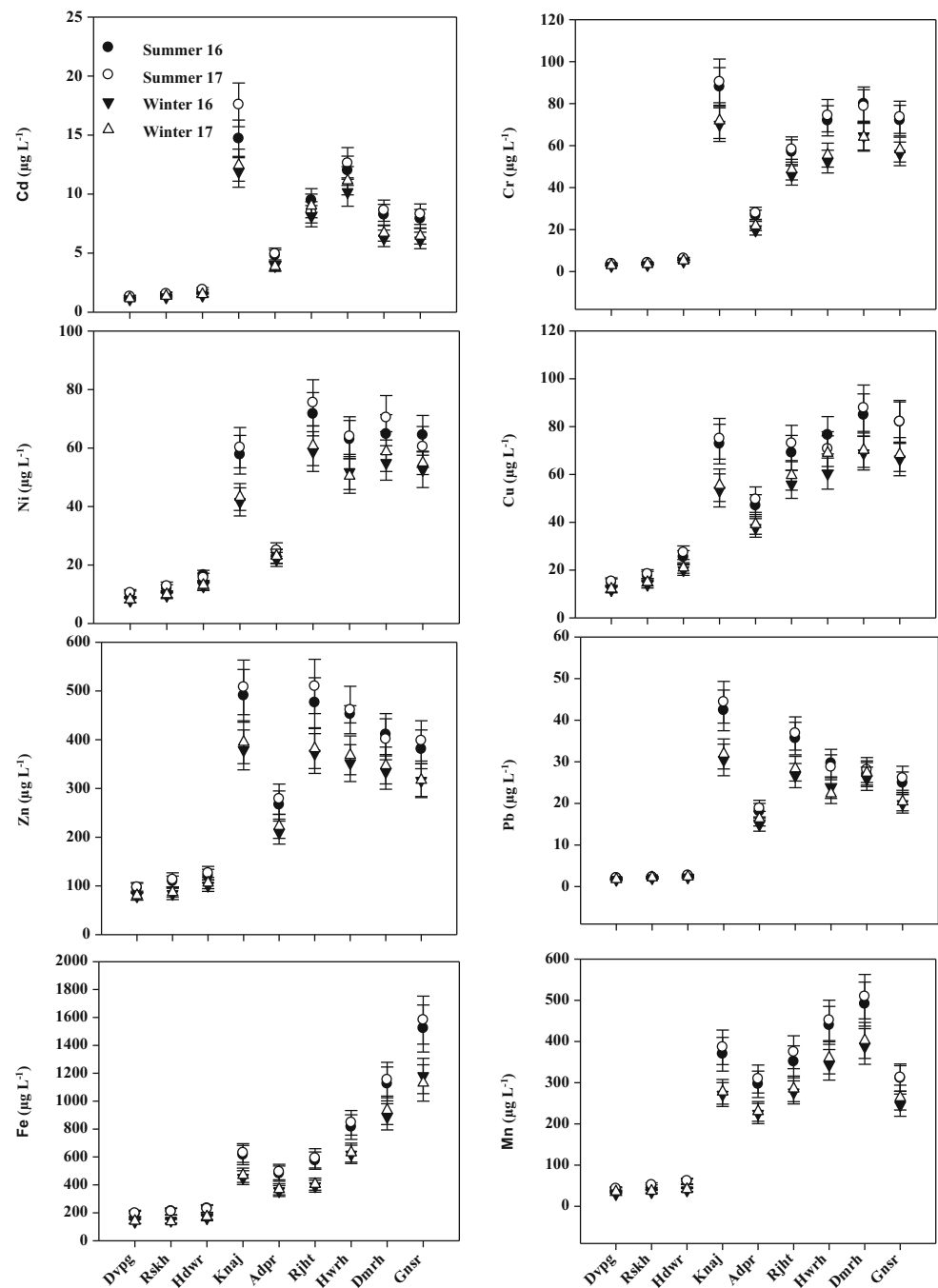


Water quality-permissible limits

About 44.44% samples of Cr and 33.33% of Cd showed concentrations below their permissible limits of 50 and $3.0 \mu\text{g L}^{-1}$ respectively (BIS 2012). About 54.63% samples of Cu showed exceedance over its permissible limit of $50 \mu\text{g L}^{-1}$. The concentration of Pb in water ranged from 1.64 to $44.34 \mu\text{g L}^{-1}$, being highest at Kannauj, and ~66.66% of the observations did exceed its permissible limit of $10 \mu\text{g L}^{-1}$. The concentration of Zn in water did not exceed its permissible limit of

$5000 \mu\text{g L}^{-1}$. About 44.44% of Mn and 33.33% of Ni samples contained these metals above their respective permissible limits. The concentration of Fe (136.89 – $1580.78 \mu\text{g L}^{-1}$) was found higher among the study metals and was recorded highest at Ganga Sagar. About 66.66% of the samples contained Fe above its permissible limit of $300 \mu\text{g L}^{-1}$. Generally, all the metals were found below their respective permissible limits at upper reach sites. The concentration of Pb, Cd, and Ni were above their respective permissible limits in middle and lower reaches.

Fig. 3 Heavy metal concentration in water in the Ganga River. Values are mean ($n = 12$) \pm SE



Sediment quality guidelines

About 99.1% and 88.9% samples of Zn and Pb were below their threshold effective concentration (TEC; MacDonald et al. 2000), (Table 1). We did not find sediment Cd, Ni, Zn, and Cu exceeding their respective PECs. Almost 100% samples from Devprayag, Rishikesh, and Haridwar contained metals below TEC. About 33.3% samples of Cr were below TEC, 32.4%

between TEC and probable effective concentration (PEC), and 34.2% were above PEC. Almost 100% samples from Kannauj and about 91.11% from Howrah contained Cr above PEC indicating likely toxicity to benthic organisms at the Kannauj and Howrah sites. About 42.6% of Cd samples were below TEC while 57.4% were between TEC and PEC. For Ni, 33.3% of the samples were below TEC, 48.1.1% between TEC and PEC, and 18.5% above PEC.

Table 1 Distribution of heavy metals in bed sediment of the Ganga River as per sediment quality guidelines

		Cr	Cd	Pb	Ni	Cu	Zn	Fe	Mn
Sediment quality guidelines	TEC	43.4	.99	35.8	22.7	31.6	121	–	–
	PEC	111	4.98	128	48.6	149	459	–	–
In this study	Min	7.12	.21	2.1	3.54	2.1	6.3	17,389	139
	Max	155.0	3.6	36.5	53.1	73.98	104.3	49,568	2167
	Mean	64.54	1.6	13.84	25	35.57	41.97	31,878	1182
Dvpg	< TEC	12	12	12	12	12	12	–	–
	TEC-PEC	0	0	0	0	0	0	–	–
	> PEC	0	0	0	0	0	0	–	–
Rskh	< TEC	12	12	12	12	12	12	–	–
	TEC-PEC	0	0	0	0	0	0	–	–
	> PEC	0	0	0	0	0	0	–	–
Hdwr	< TEC	12	12	12	12	12	12	–	–
	TEC-PEC	0	0	0	0	0	0	–	–
	> PEC	0	0	0	0	0	0	–	–
Knaj	< TEC	0	0	0	0	0	11		
	TEC-PEC	0	12	12	0	12	1		
	> PEC	12	0	0	12	0	0		
Adpr	< TEC	0	10	12	0	0	12	–	–
	TEC-PEC	12	2	0	9	12	0	–	–
	> PEC	0	0	0	3	0	0	–	–
Rjht	< TEC	0	0	12	0	0	12	–	–
	TEC-PEC	12	12	0	7	12	0	–	–
	> PEC	0	0	0	5	0	0	–	–
Hwrh	< TEC	0	0	12	0	0	12	–	–
	TEC-PEC	1	12	0	12	12	0	–	–
	> PEC	11	0	0	0	0	0	–	–
Dmrh	< TEC	0	0	12	0	0	12	–	–
	TEC-PEC	4	12	0	12	12	0	–	–
	> PEC	8	0	0	0	0	0	–	–
Gnsr	< TEC	0	0	12	0	0	12	–	–
	TEC-PEC	6	12	0	12	12	0	–	–
	> PEC	6	0	0	0	0	0	–	–
Total	< TEC	33.3%	42.6%	88.9%	33.3%	33.3%	99.1%	–	–
	TEC-PEC	32.4%	57.4%	11.1%	48.1%	66.6%	0.92%	–	–
	> PEC	34.2%	0.00%	0.00%	18.5%	0.00%	0.00%	–	–

TEC, threshold effective concentration; PEC, probable effective concentration

Geo-accumulation index

At an upper stretch, the I_{geo} values of the study metals were below unity indicating Devprayag, Rishikesh, and Haridwar to represent unpolluted river sites (Table 2). Among metals, the I_{geo} for Cd > 5.39 at Kannauj indicates the extremely polluted condition. The I_{geo} for Cr and Cd did appear in heavily contamination range except for Adalpura where the values were in moderate levels. Zn showed moderate contamination at Adalpura; moderately to heavily contaminated range at Rajghat, Howrah,

Diamond Harbour, and Ganga Sagar; and heavily contaminated range at Kannauj. Mn showed moderate to heavy contamination range at Kannauj, Adalpura, Rajghat, Howrah, and Ganga Sagar. I_{geo} for Pb did appear in moderate contamination range at Adalpura, Diamond Harbour, and Ganga Sagar while in heavy contamination range at Kannauj, Rajghat, and Howrah. Ni showed moderate contamination except at Kannauj where it did appear in heavy contamination range. Similarly, Cu showed moderate contamination levels except at Rajghat where it did appear in heavy contamination range.

Table 2 Geo-accumulation index (I_{geo}) and contamination factor (CF) for the Ganga River bed sediment

Sites	Geo-accumulation index							
	Cr	Cd	Cu	Ni	Pb	Zn	Mn	Fe
Dvpg	−0.59	−0.57	−0.58	−0.57	−0.59	−0.57	−0.57	−0.57
Rskh	−0.41	−0.51	−0.43	−0.47	−0.58	−0.51	−0.54	−0.43
Hdwr	−0.32	−0.39	−0.23	−0.41	−0.52	−0.47	−0.47	−0.39
Knaj	3.72	5.93	2.64	3.72	3.68	3.04	2.91	0.46
Adpr	2.31	1.44	2.01	2.43	2.36	1.84	2.26	0.04
Rjht	2.85	3.19	3.37	2.95	3.31	2.92	2.76	0.26
Hwrh	3.38	3.34	2.56	2.73	3.03	2.10	2.92	0.50
Dmhr	3.44	3.15	2.59	2.58	2.83	2.34	3.07	0.58
Gnsr	3.29	3.05	2.70	2.50	2.67	2.25	2.96	0.79
Contamination factor								
Dvpg	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00
Rskh	1.14	1.04	1.10	1.16	1.03	1.04	1.05	1.08
Hdwr	1.21	1.14	1.27	1.09	1.21	1.12	1.13	1.14
Knaj	20.08	17.86	9.39	13.20	16.87	12.42	11.30	2.07
Adpr	7.56	4.09	6.17	8.03	5.69	5.36	7.21	1.55
Rjht	11.02	14.04	7.14	11.48	10.97	11.32	10.19	1.79
Hwrh	15.96	15.52	9.07	9.82	9.02	7.15	11.38	2.13
Dmhr	16.64	13.57	9.26	8.86	7.87	7.59	12.60	2.25
Gnsr	15.02	12.40	9.80	8.40	7.12	7.10	11.70	2.45

Contamination factor and pollution load index

Based on CF, sites such as Kannauj, Rajghat, Howrah, Diamond Harbour, and Ganga Sagar can be categorized under the highly contaminated category ($CF > 6$) (Table 2). Upper stretch did show less or no contamination. A similar interpretation can be made also from PLI data (Table 3). Among the metals, Cr (20.08), Cd (17.86), and Pb (16.87) showed the highest degree of contamination at Kannauj; Cr, Cu, Cd, Ni, Pb, and Zn showed a high degree of contamination at middle

Table 3 Pollution load index (PLI), pollution index (PI), modified pollution index (MPI), and modified degree of contamination (mCd) for bed of the Ganga River

Sites	PLI	PI	MPI	mCd
Dvpg	1.00	1.00	1.00	1.000
Rskh	1.08	1.11	1.04	1.070
Hdwr	1.16	1.17	1.05	1.130
Knaj	11.02	17.32	10.75	14.04
Adpr	5.70	9.84	9.05	5.70
Rjht	8.60	14.97	10.34	9.80
Hwrh	8.75	18.81	9.45	10.06
Dmhr	8.60	19.31	9.34	9.80
Gnsr	8.28	17.52	9.10	9.24

and lower stretch sites. Fe showed a moderate degree of contamination at middle and lower stretches. Haridwar and all the middle and lower reach sites have PLI values above 1.

Enrichment factor

The EF followed a trend as $Cu > Cr > Cd > Mn > Pb > Ni > Zn$ (Table 4). Among the metals, the Cu showed highest EF. The upper stretch sites (Devprayag, Rishikesh, and Haridwar) showed little enrichment while Kannauj, Rajghat, Howrah and Diamond Harbour showed great enrichment. The Cr and Cd showed severe enrichment in middle and lower reaches. The almost similar condition did appear with Cu except at Kannauj where this metal showed a moderate enrichment. The EF for Ni, Pb, and Zn indicated moderate enrichment at Adalpura, Rajghat, Diamond Harbour, and Ganga Sagar while severe enrichment at Kannauj and Howrah. The Mn also showed severe enrichment at Kannauj, Rajghat, Howrah, and Diamond Harbour. Among the study sites, Kannauj, Rajghat, and Howrah showed greater enrichment with respect to heavy metals.

Ecological risks and potential ecological risks

The ecological risk was assessed in terms of E_r and PERI. As expected, PERI at Devprayag, Rishikesh, and Haridwar indicated low risk (Table 4); Adalpura, Diamond Harbour, and Ganga Sagar did appear under moderate risk while Kannauj, Rajghat, and Howrah under high-risk category. The PERI was found highest at Kannauj (784) where Cd could impose very high ecological risk ($E_r = 535$). Also, the E_r for Cd indicates very high risk at Rajghat, Howrah, Diamond Harbour, and Ganga Sagar. The E_r of Cr and Zn did appear under low risk irrespective of site. The Cu showed considerable risk at Kannauj, Rajghat, Howrah, Diamond Harbour, and Ganga Sagar while Pb showed moderate risk at Rajghat, Howrah, and Diamond Harbour but considerable risk at Kannauj. The Ni showed moderate risk at middle and lower reaches of the river.

Pollution index and the modified degree of contamination

Based on the pollution index (PI), Devprayag did appear unpolluted, Rishikesh and Haridwar moderately polluted, and the rest heavily polluted sites. Based on the mCd , Devprayag, Rishikesh, and Haridwar (Table 3) can be categorized as unpolluted; Kannauj, Rajghat, Howrah, Diamond Harbour, and Ganga Sagar under heavily polluted; and Adalpura as the moderately polluted site. We also tested the modified degree of pollution (MPI) to category sites with respect to the degree of pollution. The MPI revealed that Devprayag and Rishikesh are unpolluted while Kannauj,

Table 4 Enrichment factor (EF), ecological risk (Er), and potential ecological risk (PERI) for the Ganga River bed sediment

Sites	Enrichment factor								
	Cr	Cd	Cu	Ni	Pb	Zn	Mn	Fe	
Dvpg	1.00	1.00	1.00	1.00	1.00	1.00	1.00	1.00	
Rskh	1.05	1.01	1.05	1.04	0.92	1.01	1.01	1.00	
Hdwr	1.06	1.02	1.15	0.95	1.07	1.02	1.02	1.00	
Knaj	8.97	8.17	4.40	6.10	7.84	5.70	5.39	1.00	
Adpr	4.88	2.63	11.81	4.76	3.23	3.33	4.67	1.00	
Rjht	6.14	7.27	12.72	6.66	6.15	6.19	5.70	1.00	
Hwrh	7.48	7.81	11.81	4.57	4.30	3.33	5.35	1.00	
Dmhr	7.38	6.00	11.81	3.95	3.53	3.33	5.60	1.00	
Gnsr	6.12	5.00	11.81	3.42	2.92	2.85	4.78	1.00	
	Ecological risk								
									PERI*
Dvpg	2.00	30.00	5.00	5.00	5.00	1.00	—	—	8.02
Rskh	2.28	31.42	5.100	5.80	5.18	1.04	—	—	8.47
Hdwr	2.43	34.28	6.39	5.49	6.09	1.12	—	—	9.11
Knaj	40.16	535.0	46.95	66.60	84.35	12.40	—	—	784.0
Adpr	15.1	122.85	30.88	40.18	28.45	5.36	—	—	242.82
Rjht	22.04	420.0	38.87	57.44	54.86	11.32	—	—	604.0
Hwrh	31.93	465.71	45.35	49.1	45.11	7.15	—	—	644.0
Dmhr	32.29	407.14	46.23	44.1	39.39	7.59	—	—	577.0
Gnsr	30.04	372.85	49.04	42.0	35.63	7.10	—	—	536.0

*Potential ecological risk

Adalpur, Rajghat, Howrah, Diamond Harbour, and Ganga Sagar are heavily polluted sites.

Principal component analysis and source identification

The principal component analysis identified three significant PCs for water and sediments (Table S3). The PC1 which explained 35.6% of the variance showed strong loadings with Cu, Ni, Zn, and Pb for sediment and with 32.78% of variance showed strong loadings with Cu, Ni, Pb, Zn, and moderate loading with Cd for water. The PC2 which explained 26.8% for the variance for sediment showed strong loadings on Fe and Mn, and 28.85% of the variance for water showed strong loading on Fe, Mn, and moderate loadings of Ni. The PC3 which explained 16.60% of the variance for sediment showed strong loadings of Cr and Cd and moderate loadings of Pb, and 18.61% of the variance with water showed strong loadings with Cr and Cd. Correlation analysis showed strong correlation between Cr and Cd ($r = 0.88$; $p < 0.01$ for sediment, $r = 0.81$; $p < 0.01$ for water) and between Fe and Mn ($r = 0.82$; $p < 0.01$ for sediment; $r = 0.79$; $p < 0.01$ for water) (Table S4 and S5). Similarly, Zn, Cu, Pb, and Ni were significantly correlated with each other. Also, we found strong correlations ($r = 0.76$ to 0.92 ; $p < 0.01$) between metals in water and sediments.

Discussion

The Ganga River Basin is being rapidly transformed through rising population density, widespread land cover change, urbanization, industrialization, and other human activities that add a large number of pollutants including heavy metals to the Ganga River. Our results agree with previous studies demonstrating the co-occurrence of multiple metals in the Ganga River with anthropogenic factors predominant. We found that the middle and lower reaches are moderately to badly polluted. Our results are also in congruence with river threat assessment conducted by the Central Pollution Control Board of India. Although uncertainties and variations are inevitable owing to insufficient information on source synergies, yet our study provides greater levels of spatial details essential for the rejuvenation planning and management. The studies so far, to our knowledge, available in these lines of research, have generated fragmentary database: for instance, limited to short river stretch/tributaries (Sarin et al. 1979; Ajmal et al. 1984; Khwaja et al. 2001; Aktar et al. 2010) or limited to month/season/year on temporal scale (Sarkar et al. 2007; Prasad et al. 1989; Garg et al. 1992) or to a domain only, either water or sediment (Subramanian et al. 1987; Singh et al. 2003; Kar et al. 2008). This study considers, for the first time, to generate systematic database along with 2320-km-long stretch considering both the domains (water and sediment) and all the three segments (upper, middle, and lower). In this context, our study

provides a first detailed watershed-scale database on heavy metal concentration in water and sediment, the magnitude of contamination, and likely ecological risk to aquatic organisms in the Ganga River. The database provided here will serve to identify critical areas at risk and to diagnose the chief factors giving rise to the risk. These results highlight basin-scale patterns and comparison with other studies on Ganga and world rivers and argue for replacing fragmentary approaches to the Ganga River rejuvenation with integrative strategies to alleviate multiple sources of pollution.

Distribution and permissible limits

The basin-scale historic comparison of heavy metal distribution in the Ganga River is constrained by the scale dependence (Prasad et al. 1989; Subramanian et al. 1987). For instance, Sarin et al. (1979), Ajmal et al. (1984), and Prasad et al. (1989) have generated local-scale database while Subramanian et al. (1987) collected samples between October and December only. This study compares data on a large spatial scale. As expected, concentrations were high during summer low flow and between-season variations were significant. The concentrations of Cr and Cd were lower in the Ganga River than those reported for many other rivers of the world (Yang et al. 2009; Omwene et al. 2018; Table S1). High concentrations of Cd at the Howrah and Rajghat sites seemed to be associated with high traffic density and industrial release (Sun et al. 2014; Mummullage 2015). The Kolkata-Howrah represents one of the largest urban agglomerations of India with the highest number of vehicle per hour. The concentration of Cu was highest at Diamond Harbour. Boat ramps seemed to be the additional contributor of Cu at this site (Brady et al. 2015). The concentrations of Ni, Pb, and Zn were highest at the Kannauj site, whereas Fe and Mn were highest at Ganga Sagar due possibly to the influence of the sea. The Ganga Sagar site is located downstream of the Hooghly estuary which receives pollutants from a variety of urban industrial sources. Sites such as Kannauj, Rajghat, and Howrah showed strong urban influence.

Several hundred industrial units discharge effluents into the Ganga River; most of these are located in the middle and lower segments (Dwivedi et al. 2018). The middle segment, particularly Kannauj to Kanpur, receives a large amount of Cr, Cu, Pb, Mn, and Zn from tannery effluents (Deepali and Gangwar 2010; Katiyar 2011; Dixit et al. 2015). The concentrations of metals such as Cu, Pb, Ni, and Zn observed in this study were lower than those reported for the Gomati, a major tributary of the Ganga River (Singh et al. 1997). The significant spatial variations in the metal concentration may be due to the variation in anthropogenic sources, lithological inputs, hydrological effects, geological features, and vegetation cover (Islam et al. 2018). Studies conducted in the main channel show a high level of heavy metals with the mixed influence

of polluting sources (Ansari et al. 1999; Singh et al. 2003; Chaturvedi and Pandey 2006; Pandey and Singh 2017). Industrial effluents, municipal sewage, and agricultural releases are the most common anthropogenic source of the study metals (Islam et al. 2015; Ali et al. 2016; Yadav and Pandey 2017). Industrial sectors including tannery, battery, electroplating and heavy duty (Kannauj, Kanpur), engineering (Allahabad), and locomotives and carpet industries (Varanasi) strongly influence the middle segment. Over 359 industries drain effluents to Yamuna tributary (CPCB 2016) situated ~ 120 km upstream to Varanasi. In addition to direct urban-industrial input to the main channel, a number of tributaries add a huge amount of heavy metals in the middle segment. Tributaries of the Ganga River accounts for about 60% of the water of the main channel and drain their pollutants into the main stem (CPCB 2013). Some of the stations considered in this study showed marked influences of tributary confluences. For instance, the Alaknanda is situated upstream to Devprayag, Ramganga strongly influences the Kannauj site, while Assi and Varuna strongly influence the Varanasi site. The higher concentration of Fe in the lower reach of the river reflects the tidal influence of the Bay of Bengal.

We performed principal component analysis (PCA) to extract a small number of latent factors (principal components, PCs) to explore the similarities in distribution and to analyze the relationships among variables (Ma et al. 2016). The PCA identified three PCs: the first with 35.6% of the variance showed strong loadings on Cu, Ni, Zn, and Pb for sediment and with 32.78% of the variance showed strong loadings with Cu, Ni, Pb, Zn, and moderate loading with Cd for water indicating urban-industrial inputs (Omwene et al. 2018). The second factor which explained 26.8% of the variance for sediments showed strong loadings of Fe and Mn, and 28.85% of the variance for water showed strong loading on Fe, Mn, and moderate loadings of Ni, relating lithogenic contribution (Cox and Preda 2005; Brady et al. 2014a; Saleem et al. 2015). The third PC, which explained 16.60% of the variance for sediment showed strong loadings of Cr and Cd and moderate loadings of Pb, and 18.61% of the variance with water showed strong loadings with Cr and Cd. These factors again link anthropogenic sources, for instance, vehicular emission and diesel boats combined with other sources at the Howrah site. Further, we found a strong correlation between Cr and Cd and between Fe and Mn (Table S4 and S5). Similarly, Zn, Cu, Pb, and Ni were significantly correlated with each other. Also, we found strong correlations between metals in water and sediments indicating commonalities in sources and sedimentation-re-suspension relationships.

We assessed metal concentration with respect to permissible limits prescribed by the Bureau of Indian Standards (BIS 2012). The concentrations in over 55% samples of Cr and over 64% of Cd did exceed their permissible limit of $50 \mu\text{g L}^{-1}$ and $3.0 \mu\text{g L}^{-1}$ respectively. Similarly, over 54% samples of Cu

did exceed its permissible limit of $50 \mu\text{g L}^{-1}$. This metal, largely released from agrochemical industries, agricultural fields, and urban sewage (Islam et al. 2015), was high in the middle and lower river reaches. None of the samples of Zn was found with a concentration above its permissible limits of $5000 \mu\text{g L}^{-1}$. The main sources of Zn include agrochemical industries, agricultural fertilizers, and pesticides. Our data show that the concentrations of study metals in water did exceed their respective WHO (2003) permissible limits in the lower and middle reaches. This merits attention because the Ganga water is used for drinking purpose by a large population.

Also, we used sediment quality guidelines to assess whether a metal in sediment can impose a threat to aquatic life. About 99.1% and 88.9% samples of Zn and Pb were below their threshold effective concentration (TEC; MacDonald et al. 2000). This indicates that, at most of the study sites, Zn and Pb might not be able to impose toxic effects. These results indicate that Cd, Cr, and Ni could impose toxic effects on benthic organisms particularly at Kannauj, Rajghat, Howrah, and Diamond Harbour. The Kannauj site, with 100% samples of Cr and Ni above PEC and 100% samples of Cd, Pb, and Cu between TEC and PEC, experiences the highest risk. This has concern for food chain-associated health risk and from a biogeochemical perspective, and demonstrates why the ecological risk assessment and monitoring programs are needed. Overall, the riverine sediment is heavily polluted in the middle and lower reaches and the problem is acute near the tributaries and close to the cities. These results support the need for cutting the threat at the source and to develop efficient remediation strategies to reduce pollution level and contamination of the downstream fluvial system.

Contamination and risk assessment

The I_{geo} of metals was calculated based on the average concentrations obtained from a 2-year data. The I_{geo} ascertains the background metal enrichment. In this study, the I_{geo} classifies all sediment as unpolluted to the heavily polluted category. These observations are very similar to those based on the enrichment factor used to assess anthropogenic contributions. Thus, the I_{geo} results indicate the recent input of Cr, Cd, Cu, Ni, and Pb in the sediments of the Ganga River entering from anthropogenic sources in a major way.

Higher levels of contamination in the middle and lower stretch, particularly at urban sites, indicate anthropogenic-driven enrichment (Varol 2011; Omwene et al. 2018). The upper stretch has very less degree of contamination because of little or no industrial activity. The middle and lower reaches receive pollutants from urban sewage, industrial effluents, agricultural runoff, and atmospheric deposition (Pandey and Yadav 2017; Dwivedi et al. 2018). The sites situated in the middle and lower reaches showed moderate to high degree of contamination. The high contamination of Cr can be linked with the tanneries and

shipping-related activities (Mummullage 2015; Brady et al. 2014b). High contamination factor (CF) of Cd at Kannauj, Rajghat, Howrah, and Diamond Harbour indicated strong urban influence. Pollution load index (PLI) ranged from 1.0 to 11.02 and was found to be the highest at the Kannauj site. Based on the PLI, Devprayag and Rishikesh did appear under the unpolluted category, Haridwar under less polluted category, and the middle and lower reach sites under the polluted category (> 1). These location-specific changes in PLI provide valuable information on pollution status and causal relationships relevant to policymakers (Suresh et al. 2012).

The enrichment factor (EF) of the metal between 0.05 and 1.50 indicates the crustal origin and > 1.5 indicates anthropogenic effects (Zhang and Liu 2002). The overall data on EF indicate a strong influence of the anthropogenic activities (Islam et al. 2015; Islam et al. 2018) in the middle and lower reaches of the river. The upper reach sites (Devprayag and Rishikesh), situated in the pristine forests and mountainous region with little anthropogenic influence, experience little or no enrichment. The high enrichment at Kannauj, Rajghat, and Howrah could be linked with urban-industrial release and tributary influences (CPCB 2013). Further, the antifouling paints from ferry and shipyard can enhance metals such as Cu at Rajghat and Howrah (Brady et al. 2014b). Among the metals, the Fe showed very little enrichment indicating that this metal is largely derived from lithogenic sources.

As expected, the potential ecological risk (PERI) at Devprayag, Rishikesh, and Haridwar indicated low risk (Table 4). Adalpura, Diamond Harbour, and Ganga Sagar did appear under moderate risk while Kannauj, Rajghat, and Howrah under high-risk category. The PERI was found to be the highest at Kannauj (784) where Cd could impose very high ecological risk ($E_r = 535$). Also, the E_r for Cd indicates very high risk at Rajghat, Howrah, Diamond Harbour, and Ganga Sagar. The E_r of Cr and Zn did appear under low risk irrespective of site. The Cu showed considerable risk at Kannauj, Rajghat, Howrah, Diamond Harbour, and Ganga Sagar while Pb showed moderate risk at Rajghat, Howrah, and Diamond Harbour but considerable risk at Kannauj. The Ni showed moderate risk at the middle and lower reaches of the river. Overall, based on the ecological risk associated with potential metals, the severity of pollution did appear in the order as $\text{Cd} > \text{Ni} > \text{Pb} > \text{Cu} > \text{Cr} > \text{Zn}$. The high ecological risk associated with Cd is due to its highly toxic response factor. With respect to spatial distribution, sites with the high potential ecological risk associated with Cd were located near tributary (Kannauj), large cities (Rajghat and Howrah), and ports (Howrah, Diamond Harbour). Urban-industrial discharge, leaching from agricultural lands together with tributary influences, all enhance the pollutants' concentration and ecological risk in the middle and lower stretch of the river especially during low flow (Yadav and Pandey 2017; Dwivedi et al. 2018).

Co-occurrence of metals often constrains toxicity linkages. Multi-metal-based indices can assess the impact of multiple contaminations at a site and overcome some of the limitations. Both the multi-element indices computed here (pollution index, PI; and modified pollution index, MPI) indicate that the Ganga River sediment especially, in the middle and the lower reaches, is polluted with heavy metals. The PI has an advantage over the other indices, as even the low trigger values will ultimately lead to classifying sediment in risk category which most likely warrants further examination to identify the sources of contamination. The MPI is a potentially better index for the sediment quality assessment because it uses enrichment factor which takes into consideration diverse sediment behavior in a complex system (Duodu et al. 2016).

Ganga River pollution relates the world rivers

To understand the differences in the causal relationships in the international scenario of metal pollution, we compared our results with other major rivers of the world. The concentration of Cr was lower in the Ganga River than those reported for the Danube River, Europe (Woitke et al. 2003), the Yangtze River, China (Yang et al. 2009), the Nile River (Rifaat 2005), and the Borate Basin, Turkey (Omwene et al. 2018; Table S1). However, the Cr concentration in the Ganga River was higher than those reported for the Rimac River, Peru (Mendez 2005), the Yellow River, China (Liu et al. 2009), and the Liaohe River, China (Ke et al. 2017). The higher concentration of Cr in the Yangtze River ($205 \mu\text{g g}^{-1}$) was associated with high-intensity anthropogenic flushing (Yang et al. 2009). Relatively higher concentration of Cr in the Borate Basin ($103\text{--}3710 \mu\text{g g}^{-1}$) has been linked to intense chrome deposits in the area (Omwene et al. 2018) while there are no chrome deposits along the Ganga River. The Cd concentrations recorded here were lower than those reported for the Danube River, Europe (Woitke et al. 2003), the Rimac River, Peru (Mendez 2005), and the Borate Basin, Turkey (Omwene et al. 2018), but higher than those recorded for the Liaohe River, China (Ke et al. 2017), and the Brisbane River, Australia (Duodu et al. 2016). The upper limits of Cd and Pb in the Borate Basin (46.40 and $276 \mu\text{g g}^{-1}$, respectively) were > tenfold and > sixfold higher than the upper limits reported in the Ganga River. The reason seemed to be associated with excessive chrome deposits in the Borate Basin. Similarly, the upper limit of Ni in the Borate Basin exceeds over 22-fold higher than those recorded in the Ganga River. The higher concentrations of Cd in the study river ($0.21\text{--}4.28 \mu\text{g g}^{-1}$) compared to the Liaohe River (0.4 to $2.74 \mu\text{g g}^{-1}$; Ke et al. 2017) and the Brisbane River ($0.6\text{--}0.9 \mu\text{g g}^{-1}$; Duodu et al. 2016) has been linked to extensive input of this metal from the tributaries and cities. The values for Cu, Pb, and Ni recorded here are comparable to those reported for the Krotova River, Bangladesh (Islam et al. 2015). The concentrations of Cu, Pb,

Ni, and Zn observed in this study were lower than those reported for the Gomati River, India (Singh et al. 1997), the Danube River, Europe (Woitke et al. 2003), and the Tigris River, Turkey (Varol and Şen 2012). The higher concentration in the Gomati has been linked to large flushing of urban sewage from densely populated Lucknow metropolis. In their study, Singh et al. (1997) have shown that the addition of sewage from the 4 major drain network was responsible for the high level of these metals in the 15 km stretch of the river. This shows that city sewage is an important contributor to the metal concentration in the urban rivers. The higher ranges of Cr, Cd, Cu, Ni, and Zn in the Danube River have been assigned to be due to the tributaries under strong anthropogenic control. Under the influence of copper mine (Varol and Şen 2012), the Cu concentration in the Tigris River was over 38-fold higher ($2860.25 \mu\text{g g}^{-1}$) compared to the upper limit ($73.98 \mu\text{g g}^{-1}$) recorded in this study. The concentration of Pb was relatively higher in the Ganga River than those reported for the Liaohe River, China (Ke et al. 2017). The obvious reason was the intense anthropogenic activities along the middle and lower reaches including treated and untreated effluents, tanneries, vehicular emission, and paint industries (Aktar et al. 2010; Bhuiyan et al. 2011; Dixit et al. 2015).

Furthermore, we compared the water-metal concentrations measured here with other study results. The concentrations of study metals in water were lower than those reported for the Krotova River, Bangladesh (Islam et al. 2015), and the San Pedro, River Mexico (Gutiérrez et al. 2008), but higher than those recorded for the Amur River, Russia (Levshina 2018) (Table S2). Furthermore, relative to our results, the Cr in water was higher in Sabarnati River, India (Kumar et al. 2013), and Nairobi River, Kenya (Njuguna et al. 2017). The upper limit of Cd found in the Khoshk River water ($180.0 \mu\text{g L}^{-1}$; Salati and Moore 2010) was over tenfold higher than those reported here. However, the concentration of Cd in the Ganga River water did appear higher than those reported for the Tagus River, Spain (Nevado et al. 2009), and the Nairobi River, Kenya (Njuguna et al. 2017), but comparable to the Burgiana River, Bangladesh (Ahmad et al. 2010). The concentration of Zn in the Ganga River water (78.89 to $507.4 \mu\text{g L}^{-1}$) was lower than those reported for the Khoshk River, Iran (Salati and Moore 2010), and the Nairobi River, Kenya (Njuguna et al. 2017). The concentrations of Cu and Ni recorded here were higher than those reported for Nairobi River, Kenya (Njuguna et al. 2017). The concentration of Pb recorded here (1.64 to $44.2 \mu\text{g L}^{-1}$) was lower than those reported for the Burgiana River ($58.17\text{--}71.12 \mu\text{g L}^{-1}$; Ahmad et al. 2010), the Khoshk River ($20\text{--}130 \mu\text{g L}^{-1}$; Salati and Moore 2010), and the Nairobi River ($0.0\text{--}158.0 \mu\text{g L}^{-1}$; Njuguna et al. 2017) but was comparable to the Tagus River ($2.95\text{--}48.80 \mu\text{g L}^{-1}$; Nevado et al. 2009). The Fe and Mn in the Ganga River water were higher than those in the Khoshk River, Iran (Salati and Moore 2010), and lower

than those in the Nairobi River, Kenya (Njuguna et al. 2017). Variation in human perturbation has been recognized as a single major contributor to the differences in the overall metal concentration.

Conclusions

This basin-scale study showed significant spatial variations in the concentration of heavy metals in water and bed sediments of the Ganga River. The concentrations of Cr and Cd in water and of Mn in sediment were higher in the Ganga River in comparison to many other rivers of the world. In the middle and lower reaches, Cr, Cd, Pb, Ni, Cu, and Fe in water have exceeded their respective permissible limits of drinking water. About 34.3% samples of Cr and ~ 18.0% of Ni have reached their respective PEC; 100% samples of these metals have exceeded PEC at Kannauj. Although headword river stretch seemed very less contaminated, the middle and lower reaches showed moderately to heavily contaminated range. Kannauj, Rajghat, and Howrah did appear as the most polluted sites. Among the study metals, the highest I_{geo} was found for Cd, whereas the highest degree of contamination was recorded for Cr at Kannauj. The PEC combined with PERI and sediment quality guidelines indicated high risk to sediment dwellers at Kannauj, Rajghat, and Howrah. The conditions did appear more severe during low flow, and the recent decision by the Government of India (The Gazette of India 2018, Extraordinary, Part II, Section 3) regarding regulation of river flow may lead, at least partly, to overcome the threat. The PCA identified three PCs for water and sediment data separating anthropogenic (Cr, Cd, Cu, Ni, Pb, and Zn) and geogenic (Fe and Mn) sources. The study provides a wealth of data on heavy metals for inter-segment comparison, source apportionment, and assessment of risk to aquatic life and human consumers, and designing an action plan to rejuvenate the Ganga River.

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