



# Spatial distribution of heavy metals in surface sediments from the Ganges River basin, Bangladesh

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## Abstract

The spatial distribution and source of heavy metals (Ba, Cr, Ni, Pb, V, As, Zn, and Cu) in surface sediments of the Ganges River basin, Bangladesh, were investigated using the geo-accumulation index (Igeo), contamination factor (CF), and pollution load index (PLI). The sequences of mean concentrations of the studied heavy metals are in decreasing order as follows: Ba > V > Cr > Zn > Pb > Ni > As > Cu > Hg. Sediments in the Ganges River basin are characterized by low to moderate Igeo values for Ni, Zn, Pb, and V, while high values for Ba, As, Cu, and Cr, indicating low to moderate/high contamination of the river sediments. The mean CF load in the sediments shows that Ba is highly enriched, having moderate enrichment of As, Cu, and Cr, and relatively low enrichment of Pb, V, Zn, and Ni for the lower river basin reflecting high to moderate degree of metal pollution of the sediments. The PLI values range from 0.11 to 5.89 (average  $0.88 \pm 1.28$ ) and 0.41 to 8.79 (average  $2.13 \pm 1.81$ ) for the upper and lower Ganges River basin, respectively, which suggests that the enhanced metal pollution in the latter site is due to the threat of progressive industrialization. The principal component analysis (PCA) and hierarchical clustering (CA) records imply that the sediment samples are primarily polluted by As, Cu, Cr, and Ni resulting from anthropogenic sources. The mean Igeo and PLI values further reflecting high level of metal contamination in the lower Ganges River basin and relatively low level of contamination in the upper basin.

**Keywords** Heavy metal · Geo-accumulation index · Pollution load index · Hierarchical cluster analysis · Ganges River basin · Bangladesh

## Introduction

Heavy metal pollution of sediments is a major environmental concern in developing countries (e.g., Bangladesh, India, China, Malaysia etc.) in the last few decades owing to rapid urbanization and industrialization (Selvaraj et al. 2004; Hossain et al. 2015; Khan et al. 2017; Duncan et al. 2018). Accumulation of heavy metals in the modern sediments has

become a worldwide environmental problem due to their exposure to earth surface environments as well as sink to the aquatic regimes. It is a well-known recognized fact that sediments play a significant reservoir for the accumulation of toxic heavy metals released from rocks, agricultural materials, and industries (Huang et al. 2014; Hossain et al. 2015). However, heavy metals in most river sediments enter through the pathways by surface runoff, leachate from landfills, and direct discharge of industrial waste (Duncan et al. 2018).

The Ganges River originates in the Gangotri glacier of western Kumaun Himalayas with an elevation of ~ 7000 m (Sarin et al. 1989) and meets with the Ramganga, Gomoti, Karnali, Gandak, Bagmati, and Kosi Rivers within the Indian part making Ganges River systems, and after entering Bangladesh, it further joined with the Brahmaputra and Meghna rivers in its lower reaches (Fig. 1). The Ganges River runs 2515 km southeastward through India to Bangladesh, and draining a surface area of nearly  $1.05 \times 10^6$  km<sup>2</sup> (Meybeck and Ragu 2012). This river flows across different rock types in the Himalayas,

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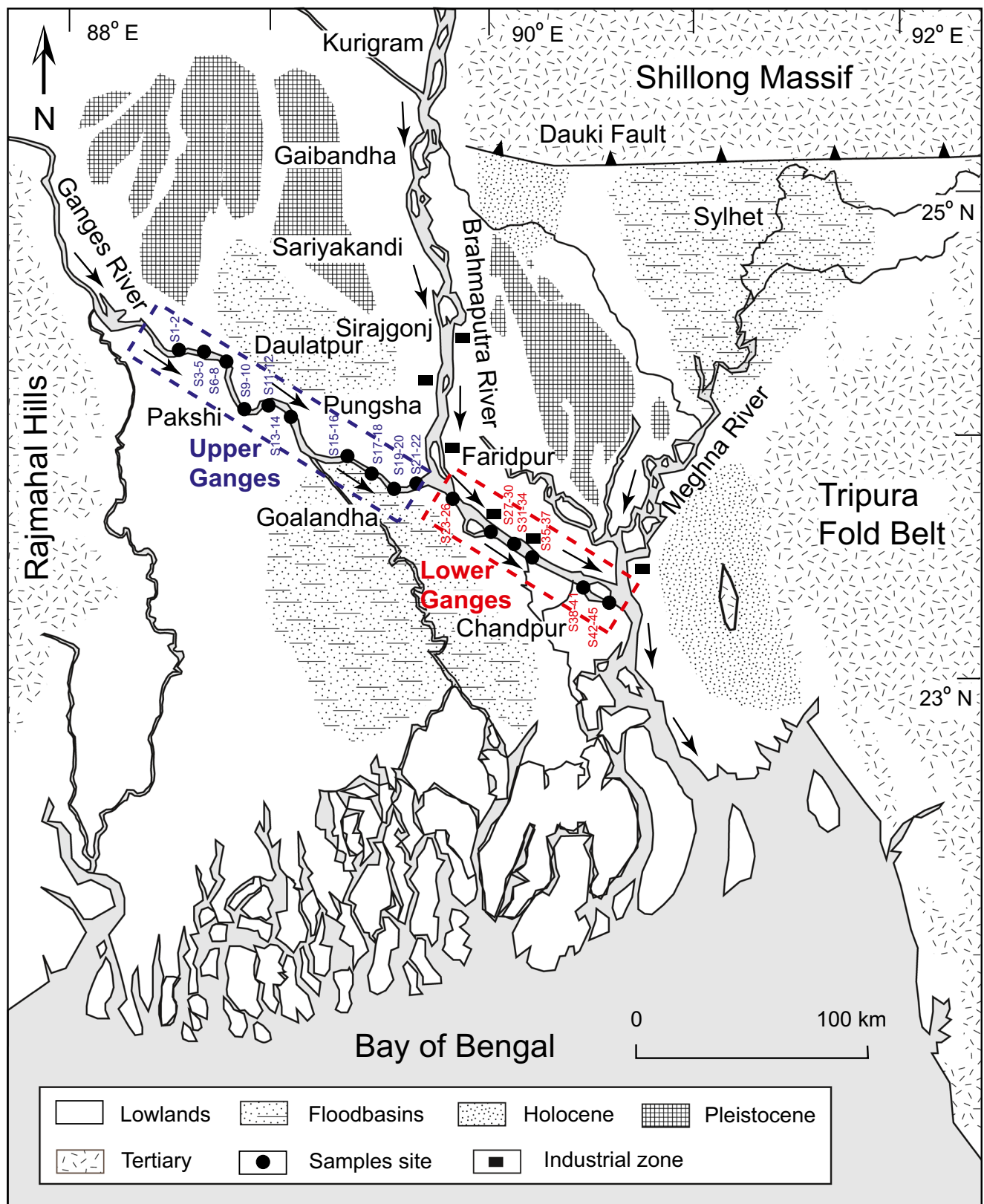


Fig. 1 Map of the Ganges River basin and surrounding areas (Hossain 2019), showing location of the sampling sites Figure 1 please confirm if captured correctly.ok

such as medium to high grade schists, metasediments, granitic gneisses, leucogranite, and carbonates (Manaka

et al. 2017). This basin is one of the most densely populated regions in the globe, with an average density of 250

person's  $\text{km}^{-2}$  (Paul 2017). The average depth of the Ganges River is about 17 m, and the maximum depth is 33 m. The Ganges River basin is a vast terraced floodplains located in the northwestern vicinity of the Bengal Basin in Bangladesh and West Bengal of India (Fig. 1). The mean annual water discharge of the Ganges River in Bangladesh is approximately  $1.14 \times 10^4 \text{ m}^3 \text{ s}^{-1}$ , with flood levels occurring at  $5.5 \times 10^4 \text{ m}^3 \text{ s}^{-1}$  (Webster et al. 2010; Manaka et al. 2015). Sediment discharge of the Ganges River is about  $330\text{--}550 \times 10^6 \text{ t year}^{-1}$  (Manaka et al. 2015). This basinal area in Bangladesh is sub-divided into upper basin and lower basin parts. The upper part of it comprises between Rajshahi and Faridpur districts, whereas the lower part is placed between joining of two large rivers namely the Brahmaputra and Meghna Rivers (Fig. 1). Many industries are located in its lower reaches which directly discharge industrial effluents to the river as well as floodplain areas. Heavy metal pollution is a serious environmental concern in the world because it cannot be degraded by natural systems and persist in the water, soil, and sediment (Paul 2017). Heavy metals such as As, Cd, Cu, Fe, Zn, Hg, Ni, Pb, and many others are micronutrients and are often considered as indicators of anthropogenic influence in aquatic environments (Selvaraj et al. 2004; Birkefeld et al. 2005; Uria et al. 2008; Bhuiyan et al. 2010; Hossain et al. 2015; Khan et al. 2017; Duncan et al. 2018), although these elements are naturally derived from various rock types (Hossain et al. 2017). These heavy metals are widely used in manufacturing industries to produce alloys, chemical and petrochemical products, food and beverage, and many other purposes (Li et al. 2009; Islam et al. 2015; Khan et al. 2017; Paul 2017). Thus, the contamination of sediments through heavy metals from industrial effluents has become a possible risk to soil ecosystems, surface environments, and human health (Hossain et al. 2015; Duncan et al. 2018). In general, the abundances of heavy metals in the soils/sediments vary from trace levels to as high as  $100,000 \text{ mg kg}^{-1}$  depending on the type of element and its location (Blaylock and Huang 2000; Asrari 2014; Hossain et al. 2015).

A study on heavy metals in the Bengal Basin of Bangladesh is limited (e.g., Bhuiyan et al. 2010; Khan et al. 2017), only those have been concentrated on agricultural soils in coal mine area and surface sediments of the Bay of Bengal, Bangladesh. This study has never been conducted in the major rivers sediments in Bangladesh to date. The present study focuses on the spatial distribution of heavy metals in the upper and lower reaches of the Ganges River basin in Bangladesh. Furthermore, the geostatistical analysis, geo-accumulation index, and pollution load index of heavy metals in the Ganges River basin, Bangladesh, have been applied to infer potential sources and degree of pollution risk in the ecosystem.

## Materials and methods

### Sampling and sample preparation

Forty-five surface sediment samples (ca.  $\sim 100 \text{ g}$ ) were collected from the lower Ganges River, Bangladesh. Sediment samples were taken from sixteen sites of the river during low tidal period in the winter season, of which 22 from the upper reaches, and 23 from the downstream where the river join with the Brahmaputra main channel. Two to four replicate samples were collected from each sampling point in the river basin. The Ganges River sediments are characterized by medium to fine sands, with mean grain size values of  $1.84\phi$  and  $2.24\phi$  for the upper and lower reaches, respectively. The sample locations are shown in Fig. 1. The samples were packed in airtight polythene bags immediately after collection, and were dried at room temperature. The dried samples were then pulverized using mortar and pestle prior to geochemical analysis in the laboratory.

### Laboratory procedures

The concentrations of Ba, Cr, Ni, Pb, V, As, Zn, and Cu were determined by Rigaku RIX 2000 X-Ray fluorescence spectrometer (XRF) at Shimane University. Loss on ignition (LOI) was estimated from net wet loss after ignition of  $1020 \text{ }^\circ\text{C}$  at muffle furnace for over 2 h. Glass fusion beads were prepared from ignited powdered samples fused with 80% lithium tetraborate and 20% lithium metaborate flux, with a flux to sample ratio of 1:2. Sample preparation technique and calibration is used by Kimura and Yamada (1996).

### Statistical analyses

Spatial dissemination and pollution of surface sediments were examined by statistical correlation, principal component analysis (PCA), hierarchical cluster analysis (CA), contamination factors (CF), geo-accumulation index (Igeo), and pollution load index (PLI). The experimental data were treated statistically using the SPSS software package version 17.0 for Windows. Pearson's correlation matrix was applied to examine the relationships among the heavy metals and support the result obtained from multivariate analysis.

### Index of geo-accumulation (Igeo)

The Igeo index is widely used to evaluate sediment quality and/or the degree of contamination in geologic materials. This index is calculated from the following equation (Müller 1969):

$$I_{\text{geo}} = \log_2 \frac{C_n}{1.5B_n}$$

where  $C_n$  is a measured heavy metal concentration of a given sediment samples and  $B_n$  is the geochemical background. Factor (1.5) is the background matrix correction factor due to lithological differences. The Igeo index consists of seven grades or classes (Müller 1969). Class 0 (practically uncontaminated):  $I_{geo} \leq 0$ ; Class 1 (uncontaminated to moderately contaminated):  $0 < I_{geo} < 1$ ; Class 2 (moderately contaminated):  $1 < I_{geo} < 2$ ; Class 3 (moderately to heavily contaminated):  $2 < I_{geo} < 3$ ; Class 4 (heavily contaminated):  $3 < I_{geo} < 4$ ; Class 5 (heavily to extremely contaminated):  $4 < I_{geo} < 5$ ; Class 6 (extremely contaminated):  $I_{geo} > 5$ . However, Class 6 is an open class and comprises all values of the Igeo index greater than Grade 5 (Müller 1969).

### Contamination factor (CF)

The CF (Müller 1969) is widely used to calculate the PLI value. The CF ratio value is calculated by dividing the concentration of each metal in the sediments by the baseline/background value taken from fresh materials (Müller 1969):

$$CF = \frac{C_{\text{metal}}}{C_{\text{backgroundvalue}}}$$

The CF index consists of four major categories to express metal contamination factor (Müller 1969; Khan et al. 2017):  $CF < 1$  refers to the low contamination factor;  $1 \leq CF < 3$  refers to the moderate contamination factor;  $3 \leq CF < 6$  refers to the very high contamination factor.

### Pollution load index (PLI)

The PLI is commonly used to assess the pollution level of heavy metal in the natural environment (Tomlinson et al. 1980; Bhuiyan et al. 2010; Bentum et al. 2011; Hossain et al. 2015).

The PLI value is estimated using the equation:

$$PLI = \sqrt[n]{(CF_1 \times CF_2 \times CF_3 \times \dots \times CF_n)}$$

where  $C_{\text{metal}}$  = metal concentration taken from sediment sample,  $C_{\text{background value}}$  = geochemical background/baseline value of the metal, and  $n$  = total number of metals. The  $PLI > 1$  imply heavy metal pollution and  $PLI < 1$  show no pollution (Hakanson 1980; Tomlinson et al. 1980).

## Results

Distributions of heavy metals (Ba, Cr, Ni, Pb, V, As, Zn, and Cu) in the Ganges River sediments are presented in Fig. 2. Concentrations of heavy metals in the surface sediments decreased in the order of  $Ba > V > Cr > Zn > Pb > Ni > As > Cu$

$> Hg$ . Mean concentration values of Ba, Cr, Ni, Pb, V, As, Zn, and Cu are  $298.21 \pm 48.96$  ppm,  $37.24 \pm 19.03$  ppm,  $11.57 \pm 4.59$  ppm,  $16.74 \pm 3.04$  ppm,  $49.83 \pm 22.30$  ppm,  $4.05 \pm 0.71$  ppm,  $32.42 \pm 7.14$  ppm, and  $2.97 \pm 2.38$  ppm, respectively. Spatial distributions of heavy metals in the lower Ganges River basin display high average enrichment of Ba ( $308.06 \pm 53.24$  ppm), V ( $63.65 \pm 21.34$  ppm), Cr ( $49.68 \pm 17.08$  ppm), Zn ( $36.22 \pm 6.19$  ppm), Pb ( $15.97 \pm 3.59$  ppm), Ni ( $13.92 \pm 3.86$  ppm), As ( $4.08 \pm 0.73$  ppm), and Cu ( $3.48 \pm 2.38$  ppm) relative to those of the upper basin (Ba,  $287.92 \pm 41.62$  ppm; V,  $35.38 \pm 11.52$  ppm; Zn,  $28.44 \pm 5.76$  ppm; Cr,  $24.23 \pm 10.21$  ppm; Pb,  $17.55 \pm 2.03$  ppm; Ni,  $9.12 \pm 3.97$  ppm; As,  $4.01 \pm 0.67$  ppm; and Cu,  $2.45 \pm 2.28$  ppm). This variation depends on varied source materials input into the studied river basin either from lithogenic or anthropogenic effluents as discussed in the "Discussion" section.

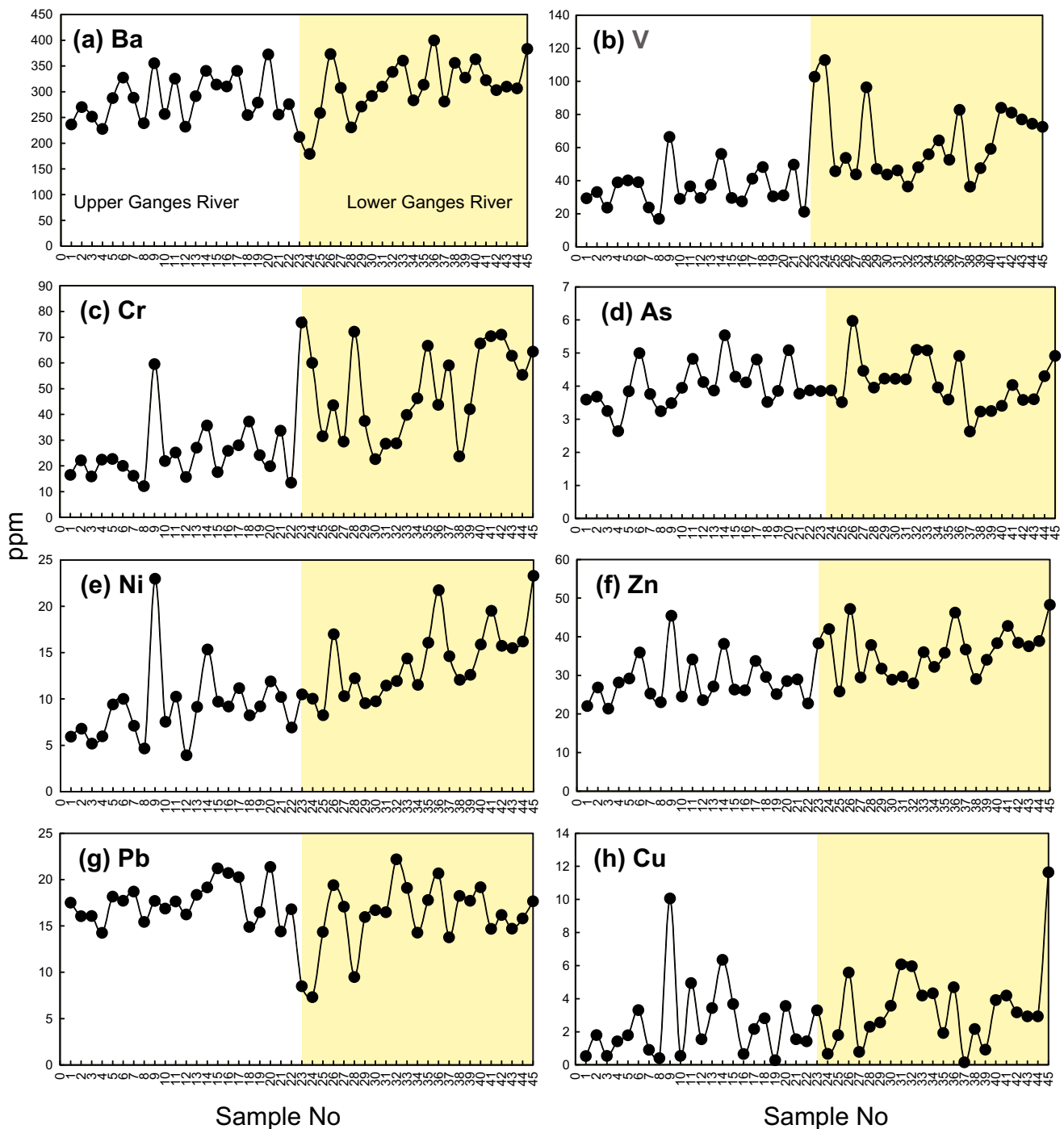
The statistical Igeo values in the studied river sediments are listed in Table 1. The mean Igeo values of Ba ( $20.54 \pm 3.55$ ), Cr ( $1.32 \pm 0.46$ ), As ( $1.82 \pm 0.33$ ), and Cu ( $1.54 \pm 1.06$ ) are relatively higher in the lower Ganges River basin, whereas little lower Igeo values are seen in the upper basin ( $19.19 \pm 2.77$ ,  $0.65 \pm 0.27$ ,  $1.78 \pm 0.30$ , and  $1.09 \pm 1.01$ , respectively). Average Igeo values of Pb, Zn, and Ni for both suits are nearly identical (Table 1). The estimated CF and PLI values are listed in Table 2. The CF values in the lower Ganges River basin vary from 17.93 to 40.01 ( $30.81 \pm 5.32$ ), 0.90 to 3.03 ( $1.99 \pm 0.68$ ), 0.37 to 1.11 ( $0.80 \pm 0.18$ ), 0.61 to 1.88 ( $1.06 \pm 0.36$ ), 1.75 to 3.98 ( $2.72 \pm 0.49$ ), and 0.11 to 7.76 ( $2.32 \pm 1.59$ ) in Ba, Cr, Pb, V, As, and Cu, respectively. Mean CF values in the upper Ganges River basin decreased in the order of  $Ba > As > Cu > Cr > V > Pb > Zn > Ni$ . However, Zn and Pb show very low ( $< 1$ ) CF values in both the lower and upper river basins, respectively (Table 2). The calculated PLI values in the lower and upper basins range from 0.41 to 8.79 ( $2.13 \pm 1.81$ ) and 0.06 to 5.89 ( $0.88 \pm 1.28$ ), respectively with high maximum values present in the sample S-45 (Table 2).

## Discussion

### Spatial distribution of heavy metals

The distribution and accumulation of heavy metals in river sediments are largely influenced by several factors such as geological and geomorphological settings, hydrological condition, and anthropogenic inputs (Yuan et al. 2014). Spatial variation of heavy metals in the upper and lower reaches of the Ganges River shows high enrichment of Ba, V, and Cr in the latter site, while Pb, As, and Cu contents are nearly identical overall (Fig. 2). Average concentration of As is similar for both sites, but Cr and V contents are twice higher in the lower basin. Mafic rocks also contained high abundances of Cr and V (Hossain et al. 2017). Some tributaries of the Brahmaputra River cross over the Himalaya-Tibetan Plateau, where mafic rocks are





**Fig. 2** Distributions of heavy metal contents (ppm) in modern sediments from the Ganges River basin, Bangladesh

extensively exposed (Hren et al. 2007). The enrichment of these elements in the sediments indicates mixing of mafic and anthropogenic source materials from riverine or industrial effluents by the Brahmaputra River. The Brahmaputra River transports sediments from the northeastern syntaxis of the Himalaya and Assam alluvial plains (Hossain et al. 2010), and many industries are located nearby to the downward stream of the Brahmaputra basin. Average concentration of Cr ( $37 \pm 19$  ppm) and V ( $50 \pm 22$  ppm) in the Ganges upper reaches

sediments is close to the average Brahmaputra River sediments (Bhuiyan et al. 2011). Thus, a high content of some metals in the lower Ganges basin is the impact of anthropogenic inputs (Hossain et al. 2015; Duncan et al. 2018). The mean averages of heavy metal concentrations in the investigated sediment samples decreased as the following order: Ba > V > Cr > Zn > Pb > Ni > As > Cu > Hg. However, the mean concentrations of heavy metals in the upper and lower reaches of the Ganges River basin decreased as the following order: Ba > V > Zn >

**Table 1** Geo-accumulation index (Igeo) values of heavy metals in modern sediments from the Ganges River basin, Bangladesh.

Sa/Nr	Ba	Cr	Ni	Pb	V	As	Zn	Cu
Upper Ganges River basin								
S-01	15.78	0.44	0.007	0.58	0.33	1.60	0.21	0.24
S-02	18.02	0.59	0.008	0.54	0.37	1.64	0.25	0.80
S-03	16.78	0.42	0.006	0.54	0.26	1.45	0.20	0.24
S-04	15.20	0.60	0.007	0.48	0.43	1.17	0.26	0.63
S-05	19.18	0.60	0.011	0.61	0.45	1.71	0.27	0.80
S-06	21.84	0.53	0.012	0.59	0.43	2.22	0.34	1.47
S-07	19.22	0.43	0.009	0.62	0.26	1.67	0.24	0.41
S-08	15.93	0.32	0.006	0.51	0.19	1.44	0.22	0.19
S-09	23.69	1.59	0.028	0.59	0.74	1.55	0.43	4.47
S-10	17.13	0.58	0.009	0.56	0.32	1.76	0.23	0.24
S-11	21.69	0.67	0.012	0.59	0.41	2.15	0.32	2.20
S-12	15.49	0.42	0.005	0.54	0.33	1.83	0.22	0.69
S-13	19.42	0.72	0.011	0.61	0.42	1.72	0.25	1.53
S-14	22.72	0.95	0.019	0.64	0.62	2.46	0.36	2.82
S-15	20.94	0.47	0.012	0.71	0.33	1.91	0.25	1.64
S-16	20.68	0.69	0.011	0.69	0.30	1.83	0.25	0.30
S-17	22.70	0.75	0.014	0.68	0.46	2.14	0.32	0.97
S-18	16.98	0.99	0.010	0.50	0.54	1.57	0.28	1.25
S-19	18.61	0.65	0.011	0.55	0.34	1.72	0.24	0.13
S-20	24.84	0.53	0.014	0.71	0.35	2.26	0.27	1.59
S-21	17.05	0.90	0.012	0.48	0.55	1.68	0.27	0.69
S-22	18.40	0.36	0.008	0.56	0.23	1.72	0.21	0.64
Mean ( $n = 22$ )	19.19 $\pm$ 2.77	0.65 $\pm$ 0.27	0.01 $\pm$ 0.01	0.58 $\pm$ 0.07	0.39 $\pm$ 0.13	1.78 $\pm$ 0.30	0.27 $\pm$ 0.05	1.09 $\pm$ 1.01
Lower Ganges River basin								
S-23	14.16	2.02	0.013	0.28	1.14	1.71	0.36	1.47
S-24	11.95	1.60	0.012	0.24	1.25	1.72	0.39	0.30
S-25	17.25	0.84	0.010	0.48	0.51	1.56	0.24	0.80
S-26	24.88	1.16	0.021	0.65	0.60	2.66	0.44	2.49
S-27	20.51	0.78	0.012	0.57	0.49	1.99	0.28	0.35
S-28	15.40	1.92	0.015	0.32	1.07	1.76	0.36	1.03
S-29	18.09	1.00	0.012	0.53	0.52	1.88	0.30	1.14
S-30	19.46	0.60	0.012	0.56	0.48	1.88	0.27	1.59
S-31	20.67	0.76	0.014	0.55	0.51	1.87	0.28	2.70
S-32	22.57	0.77	0.014	0.74	0.40	2.27	0.26	2.65

Table 1 (continued)

Sa/Nr	Ba	Cr	Ni	Pb	V	As	Zn	Cu
S-33	24.04	1.06	0.017	0.64	0.53	2.26	0.34	1.86
S-34	18.90	1.23	0.014	0.48	0.62	1.76	0.30	1.93
S-35	20.91	1.78	0.019	0.59	0.71	1.60	0.34	0.86
S-36	26.67	1.17	0.026	0.69	0.58	2.18	0.43	2.09
S-37	18.73	1.58	0.018	0.46	0.92	1.17	0.34	0.07
S-38	23.74	0.63	0.015	0.61	0.40	1.44	0.27	0.96
S-39	21.83	1.12	0.015	0.59	0.53	1.45	0.32	0.41
S-40	24.22	1.80	0.019	0.64	0.66	1.51	0.36	1.74
S-41	21.51	1.88	0.024	0.49	0.93	1.80	0.40	1.87
S-42	20.21	1.89	0.019	0.54	0.90	1.60	0.36	1.41
S-43	20.67	1.67	0.019	0.49	0.86	1.60	0.35	1.31
S-44	20.43	1.48	0.020	0.53	0.83	1.91	0.37	1.31
S-45	25.57	1.72	0.028	0.59	0.81	2.19	0.45	5.18
Mean (n = 23)	20.54 ± 3.55	1.32 ± 0.46	0.02 ± 0.01	0.53 ± 0.12	0.71 ± 0.24	1.82 ± 0.33	0.34 ± 0.06	1.54 ± 1.06

Cr > Pb > Ni > As > Cu and Ba > V > Cr > Zn > Pb > Ni > As > Cu, respectively. Barium is normally allied with clay fractions due to the progressive chemical weathering of felsic source rocks (Hossain et al. 2010, 2018). High relative abundance of Ba in the lower Ganges River sediments signifying industrial discharge and also clay mineral control their distribution in the basin (Hossain et al. 2015). Chromium shows marked positive correlation with V ( $r = 0.91$ ) for upper Ganges River basin (Fig. 3), indicating a comparable source. However, moderate positive correlation ( $r = 0.85$ ) is observed between Cr and V for the lower Ganges basin (Fig. 3), which suggests that association of these elements is also controlled by similar source detritus.

**Pollution assessment of heavy metals**

The Igeo is commonly applied to infer the degree of metal pollution in terrigenous sediments (Müller 1969; Huang et al. 2014; Hossain et al. 2015). The calculated mean Igeo values for both lower and upper Ganges basins are in decreasing order as Ba > As > Cu > Cr > Pb > V > Zn > Ni (Table 2). The elevated (> 1) mean Igeo values for both investigated areas are observed in Ba (19.88), As (1.80), and Cu (1.32) indicating moderate to high contamination by the investigated metals (Müller 1969; Hossain et al. 2015). Accordingly, mean Igeo values are equal/close to 1 for Cr (0.99), Pb (0.56), and V (0.55) suggesting that the river sediments are probably low to moderately contaminated by these metals. For the rest of the metals, Zn (0.30) and Ni (0.01) also have Igeo values nearly zero, which reflects mainly of uncontamination (Müller 1969).

The two approaches such as CF and PLI are extensively used to assess the degree of heavy metal contamination in the sediments (Tomlinson et al. 1980; Bhuiyan et al. 2010; Bentum et al. 2011; Huang et al. 2014; Hossain et al. 2015; Khan et al. 2017; Duncan et al. 2018). The CF values in the lower Ganges basin are comparatively higher than those of the upper Ganges basin, and CF value of Ba varied from 22.88 to 37.25 and 17.93 to 40.01, respectively (Table 2). The increasing order of mean CF in the studied samples is Ba > As > Cu > Cr > Pb > V > Zn > Ni. The CF load is relatively very high in all the samples for Ba, and high to moderate intensities for As, Cu, and Cr, however, low level of CF loads for Pb, V, Zn, and Ni. These features indicate that the examined sediment samples are experienced in highly to moderately polluted with metals of Ba, As, Cu, and Cr. Additionally, the PLI is also used to estimate effective pollution in the sediments or soils (Tomlinson et al. 1980; Harikumar et al. 2009; Hossain et al. 2015; Khan et al. 2017; Duncan et al. 2018). The PLI values in the upper Ganges River basin range from 0.11 to 5.89 (average  $0.88 \pm 1.28$ ) and 0.41 to 8.79 (average  $2.13 \pm 1.81$ ) for the lower basin (Table 2). The above observation suggests that majority of the samples in the lower Ganges basin are polluted by anthropogenic effluents due to the threat of increased industrialization.

**Table 2** Contamination factors (CFs) and pollution load indices (PLIs) of heavy metals in modern sediments from the Ganges River basin, Bangladesh

SaNr	Contamination factors (CFs)										PLIs
	Ba	Cr	Ni	Pb	V	As	Zn	Cu			
<b>Upper Ganges River basin</b>											
S-01	23.67	0.66	0.01	0.88	0.49	2.40	0.31	0.36	0.14		
S-02	27.03	0.89	0.01	0.80	0.55	2.46	0.38	1.20	0.38		
S-03	25.17	0.63	0.01	0.80	0.39	2.17	0.30	0.36	0.11		
S-04	22.80	0.90	0.01	0.71	0.65	1.76	0.40	0.95	0.26		
S-05	28.77	0.91	0.02	0.91	0.67	2.57	0.41	1.20	0.58		
S-06	32.77	0.80	0.02	0.89	0.65	3.33	0.51	2.21	1.01		
S-07	28.83	0.65	0.01	0.94	0.40	2.51	0.36	0.61	0.22		
S-08	23.90	0.49	0.01	0.77	0.28	2.16	0.32	0.28	0.06		
S-09	35.54	2.38	0.04	0.89	1.11	2.33	0.64	6.71	5.89		
S-10	25.69	0.87	0.01	0.84	0.48	2.64	0.34	0.36	0.20		
S-11	32.53	1.01	0.02	0.88	0.61	3.22	0.48	3.30	1.29		
S-12	23.23	0.63	0.01	0.81	0.49	2.75	0.33	1.03	0.20		
S-13	29.13	1.08	0.02	0.92	0.62	2.58	0.38	2.30	0.83		
S-14	34.08	1.43	0.03	0.96	0.93	3.69	0.54	4.24	3.19		
S-15	31.40	0.70	0.02	1.06	0.49	2.86	0.37	2.46	0.73		
S-16	31.03	1.03	0.02	1.04	0.46	2.74	0.37	0.45	0.34		
S-17	34.05	1.12	0.02	1.01	0.69	3.21	0.47	1.45	1.09		
S-18	25.46	1.49	0.01	0.74	0.80	2.35	0.42	1.88	0.79		
S-19	27.91	0.97	0.02	0.82	0.51	2.57	0.35	0.19	0.18		
S-20	37.25	0.80	0.02	1.07	0.52	3.39	0.40	2.38	1.07		
S-21	25.57	1.35	0.02	0.72	0.83	2.52	0.41	1.03	0.64		
S-22	27.61	0.54	0.01	0.84	0.35	2.58	0.32	0.95	0.21		
Mean ( <i>n</i> = 22)	28.79 ± 4.16	0.97 ± 0.41	0.02 ± 0.01	0.88 ± 0.10	0.59 ± 0.19	2.67 ± 0.45	0.40 ± 0.08	1.63 ± 1.52	0.88 ± 1.28		
<b>Lower Ganges River basin</b>											
S-23	21.24	3.03	0.02	0.42	1.71	2.57	0.54	2.21	1.65		
S-24	17.93	2.40	0.02	0.37	1.88	2.58	0.59	0.45	0.61		
S-25	25.88	1.26	0.01	0.72	0.76	2.35	0.36	1.20	0.52		
S-26	37.33	1.74	0.03	0.97	0.89	3.98	0.66	3.73	4.15		
S-27	30.76	1.18	0.02	0.85	0.73	2.98	0.41	0.53	0.53		
S-28	23.09	2.89	0.02	0.47	1.61	2.64	0.53	1.54	1.57		
S-29	27.13	1.50	0.02	0.80	0.78	2.82	0.45	1.71	0.98		
S-30	29.18	0.90	0.02	0.84	0.73	2.82	0.41	2.38	0.88		
S-31	31.00	1.14	0.02	0.82	0.77	2.81	0.42	4.05	1.49		
S-32	33.85	1.15	0.02	1.11	0.61	3.40	0.39	3.98	1.74		
S-33	36.07	1.59	0.03	0.95	0.80	3.39	0.51	2.80	2.35		
S-34	28.35	1.85	0.02	0.71	0.93	2.64	0.45	2.89	1.59		
S-35	31.37	2.67	0.03	0.89	1.07	2.40	0.50	1.29	1.91		
S-36	40.01	1.75	0.04	1.03	0.88	3.28	0.65	3.13	4.09		
S-37	28.10	2.36	0.03	0.69	1.38	1.75	0.52	0.11	0.41		
S-38	35.61	0.95	0.02	0.91	0.61	2.16	0.41	1.45	0.72		
S-39	32.74	1.68	0.02	0.89	0.79	2.17	0.48	0.61	0.75		
S-40	36.32	2.70	0.03	0.96	0.99	2.27	0.54	2.62	2.93		
S-41	32.26	2.81	0.04	0.73	1.40	2.69	0.60	2.80	3.88		
S-42	30.32	2.84	0.03	0.81	1.35	2.39	0.54	2.12	2.72		



Table 2 (continued)

Sa/Nr	Contamination factors (CFs)										PLIs
	Ba	Cr	Ni	Pb	V	As	Zn	Cu			
S-43	31.00	2.51	0.03	0.74	1.28	2.41	0.53	1.96	2.27		
S-44	30.65	2.22	0.03	0.79	1.24	2.87	0.55	1.96	2.45		
S-45	38.35	2.58	0.04	0.88	1.21	3.28	0.68	7.76	8.79		
Mean (n = 23)	30.81 ± 5.32	1.99 ± 0.68	0.03 ± 0.01	0.80 ± 0.18	1.06 ± 0.36	2.72 ± 0.49	0.51 ± 0.09	2.32 ± 1.59	2.13 ± 1.81		

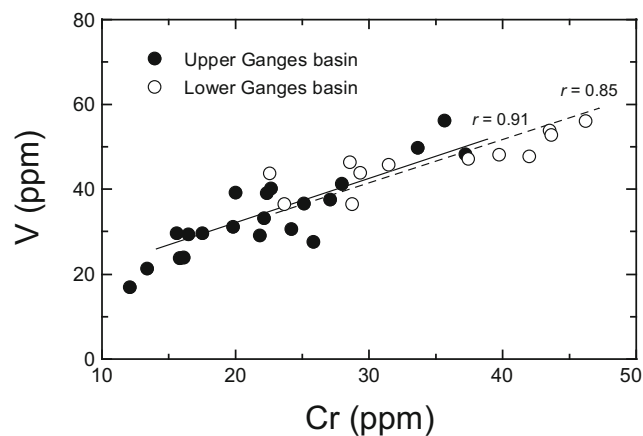


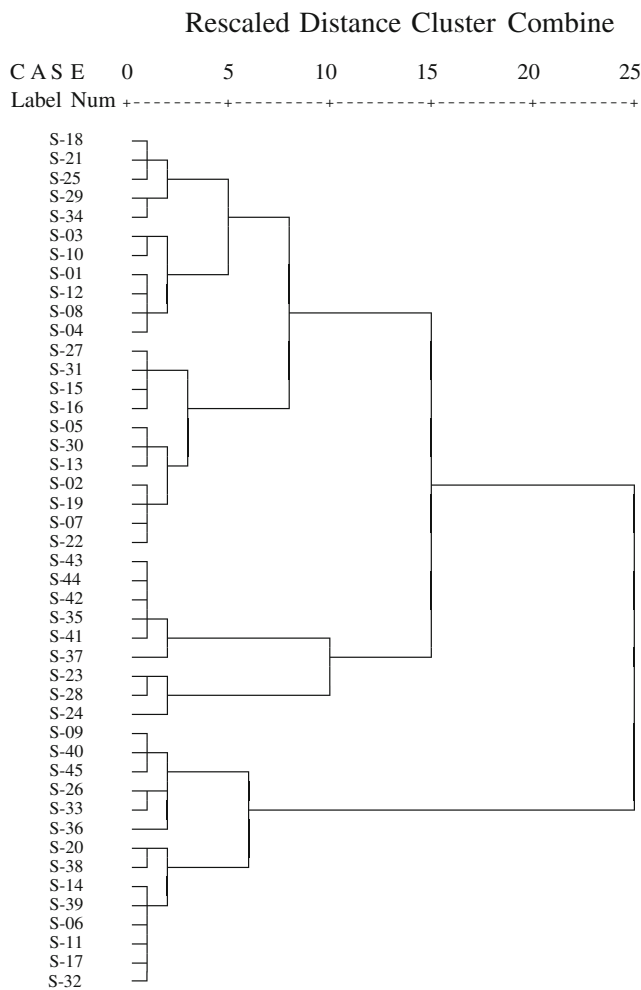
Fig. 3 Chromium-vanadium variation diagram for the Ganges River basin sediments. Solid lines (fitted by eye) are indicative detrital trends in the studied sediment samples

### Potential sources of heavy metals

#### Inter-element relationship and hierarchical clustering

The inter-element relationships have been used to evaluate metal sources and pathways for mobilization of elements (Zhou et al. 2008). The hierarchical clustering was performed in identical data spread on Ward’s method using the squared Euclidean distance as a comparable measure (Gotelli and Ellison 2004; Zhou et al. 2008; Bhuiyan et al. 2010; Li et al. 2013; Hossain et al. 2015; Islam et al. 2015). The CA is frequently used to measure metal variables in order to illustrate a spatial sampling approach (Li et al. 2013; Hossain et al. 2015; Duncan et al. 2018). Thus, CA has been applied in random sample sites for the Ganges River basin, and the statistical data reveals four major clusters from the dendrogram with significant linkage distance. The dendrogram shows clustering of investigated sediment samples, subsequently grouped into four statistically significant clusters, such as cluster 1, cluster 2, cluster 3, and cluster 4 (Fig. 4). These clusters display variable degree of contaminations resulting from anthropogenic sources. Major population of the upper Ganges River sediment samples belongs to the clusters 1 and 2, which relates to the reasonably lesser pollution sections and/or characteristically uncontaminated. However, sediment samples from the lower Ganges River basin belonged into the clusters 3 and 4 (Fig. 4), and these sediments contained relatively high Ba, V, Cr, Zn, and Ni (Fig. 2) reflecting moderate to high contamination and the toxic metals were primarily derived from rapidly growing large-scale industrial effluents (Hossain et al. 2015). Many industries (e.g., metal, ceramic, dyeing, textile, poultry feed, and food and beverage) are present nearby the lower Ganges River basin and thus released these toxic substances that directly contaminate the river sediments.

Pearson correlation coefficient of the studied heavy metals and *p* values (probability of no correlation) for statistical



**Fig. 4** Dendrogram showing clustering of sampling sites according to Ward's method using squared Euclidean distance

hypothesis testing are shown in Table 3. The Cu content shows a statistically significant positive correlation with Ni ( $r = 0.71, p < 0.01$ ), Zn ( $r = 0.62, p < 0.01$ ), and Ba ( $r = 0.61, p < 0.01$ ), and weak positive correlation with Cr ( $r = 0.34, p < 0.05$ ). However, Zn has a strong positive

correlation with Ni ( $r = 0.88, p < 0.01$ ), Cr ( $r = 0.79, p < 0.01$ ), V ( $r = 0.76, p < 0.01$ ), and Ba ( $r = 0.47, p < 0.01$ ), and weak positive correlation with As ( $r = 0.34, p < 0.05$ ). The correlation between As and Ba is statistically significant Ba ( $r = 0.54, p < 0.01$ ), whereas V alone shows strong positive correlation with Cr ( $r = 0.92, p < 0.01$ ) and Ni ( $r = 0.57, p < 0.01$ ) (Table 3). Similarly, Ni shows a significant negative correlation with Cr ( $r = 0.71, p < 0.01$ ) and Ba ( $r = 0.70, p < 0.01$ ), while correlation between V and Pb is negative ( $r = -0.63, p < 0.01$ ). The overall positive correlation between the examined heavy metals indicates that they were primarily derived from similar source materials (Hossain et al. 2015). Barium, V, and Cr are primarily associated with phyllosilicate clay minerals, and a positive correlation among them suggests comparable source detritus input into the basin (Hossain et al. 2010). In addition, V, Cr, Zn, and Ni are found in industrial effluents; therefore, high concentrations of these elements relative to background values (Singh et al. 2003) in the lower Ganges River basin are ascribed to be an anthropogenic source. The correlation coefficient values between Pb and V ( $r = -0.63, p < 0.01$ ) and Cr ( $r = -0.44, p < 0.01$ ) for the investigated sediments indicate diverse source materials input to the surface sediments.

**Principal component analysis**

The PCA with varimax normalization is commonly used as a tool to estimate anthropogenic or natural source identification in sediments or soils (Rubio et al. 2000; Gotelli and Ellison 2004; Zhou et al. 2008; Chabukdhara and Nema 2013; Yuan et al. 2014; Hossain et al. 2015). A total of four principal components (PCs) with eigenvalues ( $> 1$ ) were extracted which elucidate 96.98% of the total variance (Table 4). The possible scores and loadings of the first three PCs for the analyzed heavy metals are displayed in Fig. 5. The varimax-rotated PC1 is marked positively loaded on Cr, V, Zn, Ni, and weak positive loading on Cu, Ba, and As, which elucidate 50.19% of

**Table 3** Pearson correlation matrix for heavy metal concentration in modern sediments from the Ganges River basin, Bangladesh

	Ba	Cr	Ni	Pb	V	As	Zn	Cu
Br	1							
Cr	0.111	1						
Ni	<i>0.701**</i>	<i>0.712**</i>	1					
Pb	<i>0.751**</i>	<i>-0.435**</i>	0.166	1				
V	-0.096	<i>0.919**</i>	<i>0.568**</i>	<i>-0.633**</i>	1			
As	<i>0.535**</i>	-0.069	<i>0.304*</i>	<i>0.439**</i>	-0.047	1		
Zn	<i>0.466**</i>	<i>0.792**</i>	<i>0.884**</i>	-0.129	<i>0.755**</i>	<i>0.335*</i>	1	
Cu	<i>0.611**</i>	<i>0.343*</i>	<i>0.707**</i>	0.264	0.25	<i>0.495**</i>	<i>0.620**</i>	1

Values in italics represent correlation with significance

\*\* Correlation is significant at the 0.01 level (2-tailed)

\* Correlation is significant at the 0.05 level (2-tailed)

**Table 4** Varimax-rotated principal component analysis for heavy metals in modern sediments from the Ganges River basin, Bangladesh

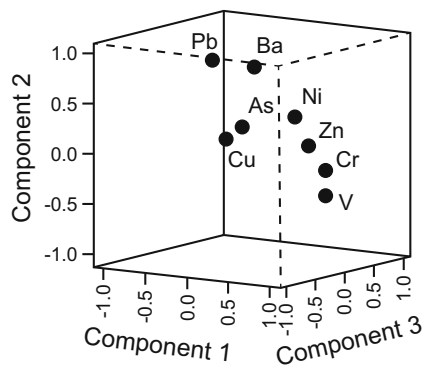
Parameter	Principal component			
	PC1	PC2	PC3	PC4
Ba	0.209	0.887	0.283	0.246
Cr	0.955	-0.096	0.11	-0.11
Ni	0.775	0.474	0.377	0.08
Pb	-0.379	0.878	0.098	0.198
V	0.92	-0.35	0.066	0.01
As	0.028	0.274	0.203	0.937
Zn	0.878	0.161	0.296	0.235
Cu	0.303	0.269	0.878	0.248
Variance (%)	50.19	33.836	8.192	4.761
CV	50.19	84.027	92.219	96.979

Extraction method: principal component analysis

Rotation method: Varimax with Kaiser normalization

a. Rotation converged in 5 iterations

the total variance. In contrast, Pb alone shows negative loading in PC1 section. However, high positive loading of Ba and Pb is observed on PC2, and moderate to weak loading of Ni, As, and Cu on PC2 accounting for 84.03% of the total variance representing the potential impact from industrial discharges in addition to agricultural effects (Li et al. 2013; Hossain et al. 2015). Accordingly, PC3 displays strong positive loading of Cu and moderate loading of Ni, Zn, and Ba which accounts for 33.84% of the total variance, signifying mixed sources (Hossain et al. 2015). Arsenic has distinctly positive loading in PC4 and negative loading of Cr accounting for 8.19% of the total variance. The positive loading of Zn, Cu, and As on PCs is ascribed to be derived from anthropogenic source mostly from fertilizer (e.g., phosphate, zinc sulfate, zinc oxide, copper sulfate) used in agricultural soils (Zarcinas et al. 2004; Bandyopadhyay et al. 2014; Hossain et al. 2015; Jayasumana et al. 2015).



**Fig. 5** Plot of three principal components for studied heavy metals (factor loadings, factor 1 vs. factor 2 vs. factor 3; rotation, varimax normalized; extraction, principal components) in the Ganges River basin, Bangladesh

## Conclusions

We examined spatial distribution of selected heavy metals in sediments from upper and lower reaches of the Ganges River basin in Bangladesh. Our main conclusions are as follows:

1. Mean concentrations of heavy metals in the investigated sediments are as follows: Ba > V > Cr > Zn > Pb > Ni > As > Cu > Hg.
2. High Igeo values are observed in most samples of the lower Ganges basin than those of upstream, representing low to moderate/high contamination of the river sediments.
3. The mean CF loads in the sediments are in decreasing order as follows: Ba > As > Cu > Cr > Pb > V > Zn > Ni, and moderate to high enrichment of Ba, As, Cu, Cr, and Ni in the lower Ganges River basin indicating moderate to high level of metal pollution owing to the input of industrial effluents.
4. Mean PLI loads are high in the lower basin sediments ( $2.13 \pm 1.81$ ) than those of the upper basin sediments ( $0.88 \pm 1.28$ ) inferring that progressive metal pollution in the lower basin site is due to the admixture of anthropogenic source materials.
5. The PCA and CA records suggest that the sediments are mainly polluted by Cu, Cr, As, and Ni with subsequent influx from anthropogenic effluents.

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