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Spatiotemporal distribution and pollution assessment of trace metals in the Buriganga River, Bangladesh

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ABSTRACT

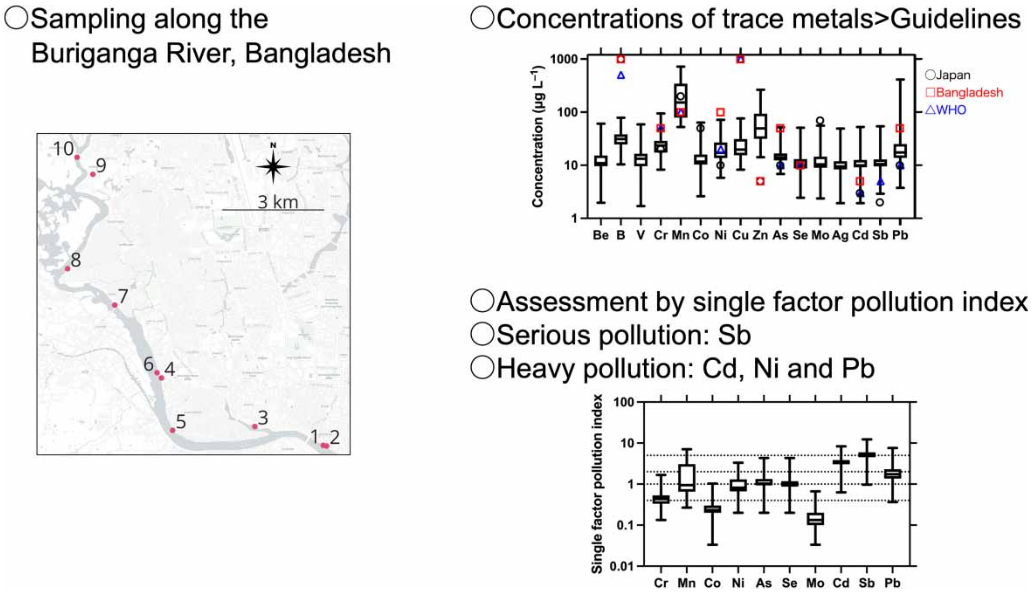
The Buriganga River plays a key role in the socioeconomic structure of Dhaka, the capital of Bangladesh. However, this river is severely polluted and is considered one of the most polluted in the world. Therefore, this study aimed to assess the concentrations of various metals in the Buriganga River. A study was conducted from August 2019 to February 2020 to determine the concentrations of 16 metals in water samples ($n = 210$) collected from 10 distinct sites in the Buriganga River. The mean values for the concentrations of Cr, Mn, Ni, Zn, As, Se, Cd, Sb, and Pb in river water were above the guideline values prescribed by the WHO, Japan, and Bangladesh. Moreover, the fraction ratios of Be, Cr, Co, Ni, Cu, Zn, Se, Mo, Ag, Cd, Sb, and Pb were high (>0.85); consequently, these metals could accumulate at high concentrations in river sediments. Assessment using the single-factor pollution index allowed the classification of the pollution level as 'serious pollution' for Sb and 'heavy pollution' for Cd, Ni, and Pb. The trace metal concentrations in this river imply that crops cultivated along the river using river water may also be contaminated with trace metals.

Key words: ICP-AES, ICP-MS, pollution index, river pollution, trace metals

HIGHLIGHTS

- The highest concentrations of Cr, Mn, Co, Ni, Zn, As, Se, Mo, Cd, Sb, and Pb were above the guideline values.
- High fraction ratio values of Be, Cr, Co, Ni, Cu, Zn, Se, Mo, Ag, Cd, Sb, and Pb indicated that these metals were distributed on suspended solids.
- A single-factor pollution index showed that the Buriganga River was heavily polluted by Sb, Cd, Ni, and Pb.
- Mn showed relatively high concentrations in the dry season.

GRAPHICAL ABSTRACT



INTRODUCTION

Pollution of river environments by trace metals, which are highly toxic and persistent, is attracting global attention (Varol 2011; Yuan *et al.* 2011; Fu *et al.* 2014; Islam *et al.* 2015). Trace metals are a primary environmental concern because they are non-biodegradable and toxic and have a cumulative effect (Briffa *et al.* 2020). Trace metals deposited in the sediment can also be re-mixed into the water column via suspension, desorption, oxidation, or reduction processes, posing an increased risk to aquatic organisms, animals, and humans (Dong *et al.* 2012; Zhao *et al.* 2013). With the intensification of industrialization in many countries, numerous reports on trace metal contamination of river environments owing to industrial effluent discharge have emerged, raising concerns regarding the adverse effects of trace metal exposure on human health and ecosystems (Li & Zhang 2010; Islam *et al.* 2014b). The International Agency for Research on Cancer categorizes the metals As, Pb, and Ni as group 1 ('carcinogenic to humans'), 2B ('possibly carcinogenic to humans'), and 3 ('not classifiable as to its carcinogenicity to humans'), respectively (World Health Organization 2023). Furthermore, adverse health effects, such as hypertension (As), diabetes (Cd), reproductive toxicity (Pb, Ni), dopamine oxidation (Mn), metabolic function effects (Cu), and respiratory problems (Zn), have been reported in humans who accidentally ingest trace metals (Uddin & Jeong 2021).

The Buriganga River is considered one of the most polluted rivers in the world. Anthropogenic activities have been reported as the primary source of trace metal contamination in this river, which flows southwest of Dhaka City, the capital of Bangladesh (Islam *et al.* 2014a; Nargis *et al.* 2021), with huge volumes of toxic waste discharged from thousands of industrial units and sewage lines being the primary pollutants (Islam *et al.* 2006; Kawser Ahmed *et al.* 2016). Hospital waste, boat and launch repairs, galvanizing steel, motor repairs, rechargeable batteries, foundry products, metal ceramics, textiles, dyeing, and leather industries, municipal sewage sludge, fertilizers, pesticides, insecticides, and oil emissions are also pollutant sources (Islam *et al.* 2015; Raknuzzaman *et al.* 2016). These deleterious effluents, including chromium, lead, sulfur, ammonium, salt, and other materials, severely pollute the Buriganga River (Hoque *et al.* 2016) and can seriously impact human health. The residents of Dhaka use this river daily as a water transportation route and for ablutions. Furthermore, oral exposure of humans to contaminants from ingesting crops cultivated using the river water is of particular concern, as the Buriganga River is utilized as a source of water for agricultural purposes (Islam *et al.* 2014a; Bhuyan *et al.* 2017). Therefore, a comprehensive study of the contamination levels of trace metals in this river is important for protecting the health of the residents of Dhaka.

This study aimed to assess the risk posed by trace metal contamination in the Buriganga River, Bangladesh. We collected water samples at 10 sites along the Buriganga River from August 2019 to February 2020. Trace metal concentrations, fraction ratios between dissolved and particulate phases, and single-factor pollution index values were determined to evaluate the level of metal contamination in the Buriganga River.

METHODS

Chemicals

Standard solutions for Be, B, V, Cr, Mn, Ni, Cu, Zn, As, Se, Y, Mo, Ag, Cd, In, Sb, Pb, and Bi were obtained from FUJIFILM Wako Pure Chemical Industries (Osaka, Japan). Y, In, and Bi were used as internal standards. Inductively coupled plasma (ICP) analysis grade nitric acid, ultra-trace analysis grade HF, and poisonous grade $HClO_4$ were purchased from FUJIFILM Wako Pure Chemical Ind. Ultrapure water (18.2 MΩ cm) was used to dilute nitric acid.

Sampling sites: Buriganga River

Water samples ($n = 210$) were collected from 10 sampling sites on the Buriganga River (Figure 1), Dhaka, Bangladesh from August 2019 to February 2020. These sites were investigated to evaluate the pollution status of the Briganga River (e.g., Nargis *et al.* 2021). Three samples were collected at a depth of approximately 10–20 cm from the water surface using precleaned polyethylene bottles. The water temperature and pH were measured by using a multi water quality analyzer (U-51, HORIBA, Ltd, Kyoto, Japan) at the time of sampling and are listed in Supplementary Table S1.

The water samples were filtered using a quartz fiber filter to separate the suspended solids (SS) to determine the fraction of dissolved metals and metals adsorbed on SS. The filtered water samples and filters were transported from Dhaka to the University of Shizuoka, Japan, for instrumental analysis.

Pretreatment

The samples were treated using a previously reported modification (Islam *et al.* 2014b; Bhuyan *et al.* 2017). Briefly, the filtered water samples (10 mL), nitric acid (60%, 75 μL), and internal standards (100 μL) were added to precleaned polyethylene centrifuge tubes and stored at 4 °C until the analysis. Ultrapure water was treated in the same manner as the filtered water samples to evaluate their blank levels.

A quarter of the filter was cut using Teflon scissors and weighed. The cut filter, nitric acid (60%, 2.5 mL), and HF solution (50%, 2.5 mL) were mixed in a Teflon beaker and heated at 200 °C for 30 min. Subsequently, $HClO_4$ (60%, 3.0 mL) was added to the Teflon beaker, and the mixture was heated at 200 °C for 30 min. The mixture was heated, and the solution was almost depleted. Nitric acid (0.1 mol L^{-1}, 5 mL) was then added to the Teflon beakers, and the contents were heated at 120 °C to dissolve the remaining residue. The nitric acid solution in the beaker was then transferred to precleaned polyethylene centrifuge tubes containing an internal standard (100 μL). The volume of the solution was fixed with nitric acid (0.1 mol L^{-1}) to 10 mL. Precleaned filters were treated in the same manner as the filter samples to evaluate the blank level.

Instrumental analysis

The metal concentrations in the pretreated water and filter samples were measured using an ICP atomic emission spectrometer (AES) (5110 Agilent Technologies, Santa Clara, CA) and ICP mass spectrometer (ICP-MS) (Varian 820-MS, Agilent). Samples with concentrations lower than the ICP-AES calibration range were analyzed using ICP-MS.

Quality assurance and quality control

No metals were detected in the blank samples. The calibration curves for each metal were linear over the entire range of standard concentrations (1, 10, 25, 50, 100, and 500 μg L^{-1} for ICP-AES; 0.05, 0.1, 0.5, 1, 5, and 10 μg L^{-1} for ICP-MS; coefficient of determination >0.99). The metal concentrations in the samples were determined to be in the ranges of the calibration curves.

Calculation of the single-factor pollution index

The single-factor pollution index was calculated to evaluate pollution levels in the study area (Ajibare *et al.* 2002; Yan *et al.* 2015). The single-factor pollution index was primarily developed for risk assessment in soil environments (Teng *et al.* 2022). Recently, this index has also been used for water quality assessment (Ajibare *et al.* 2002, Yan *et al.* 2015).

$$P_i = \frac{C_i}{S_i}, \tag{1}$$

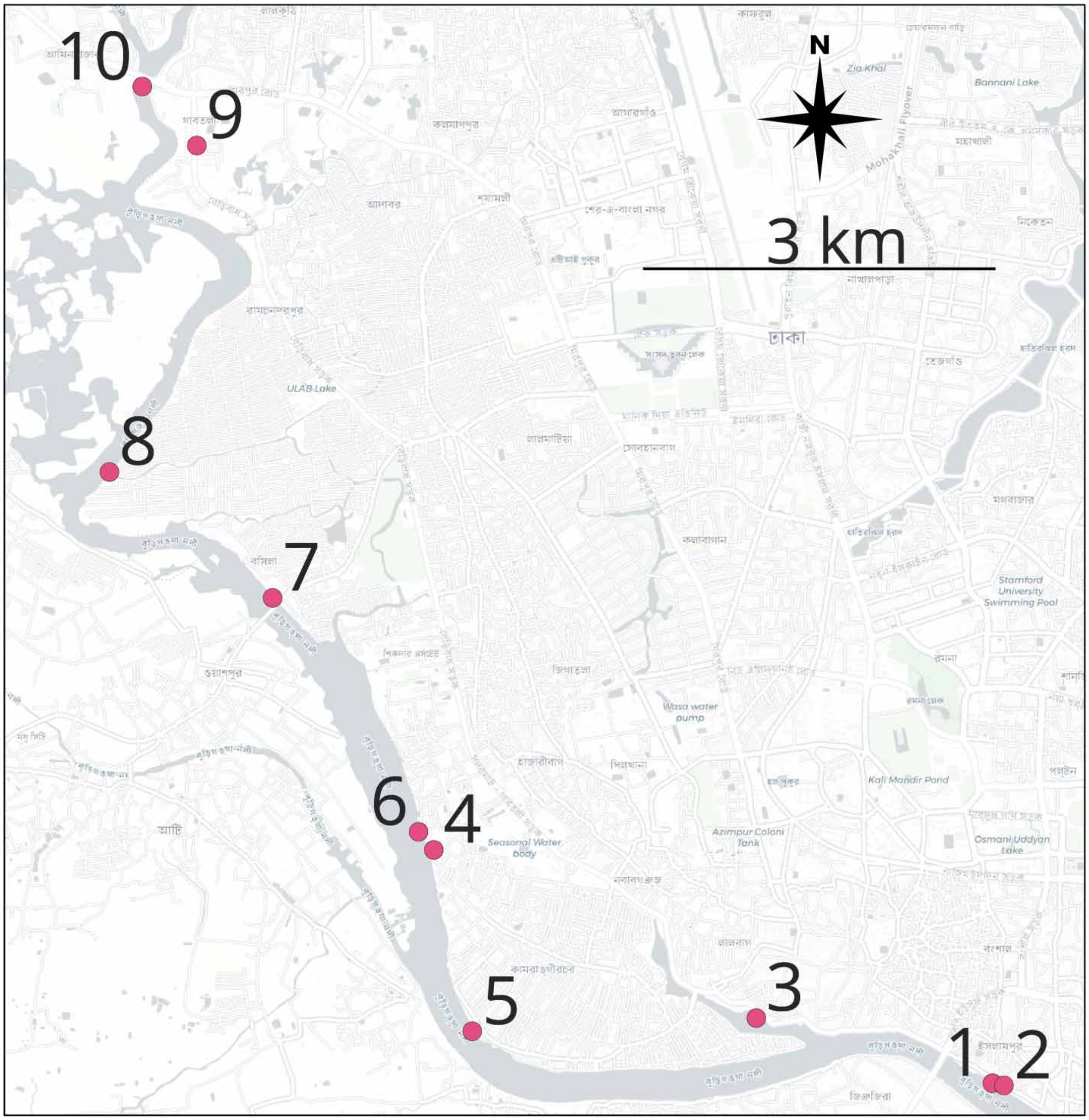


Figure 1 | Sampling sites along the Buriganga River, Dhaka, Bangladesh.

where P_i (a.u.) is the single-factor pollution index of metal i, C_i (μg L^{-1}) is the total concentration of metal i, and S_i (μg L^{-1}) is the permissible limit for metal i in the aquatic environment. The C_i values were defined as the sum of the dissolved metal concentrations and the concentrations of metals adsorbed on the SS. The median values of the three standard values from Bangladesh, the WHO, and Japan were used as S_i values here (Table 1) (Bangladesh 2019; Hayami *et al.* 2022). The P_i values were classified into five grades: <0.4 = no pollution, 0.4–1 = slight pollution, 1–2 = medium pollution, 2–5 = heavy pollution, and >5 = serious pollution (Li & Zhang 2010).

Temporal distributions and spatial distributions of P_i values were investigated by Tukey's multiple comparison test. Prism 9 (GraphPad Software, Boston, MA) was used for the analysis.

Table 1 | Metal concentrations ($\mu g\ L^{-1}$) in water samples ($n = 210$) of the Buriganaga River in the present study, fractions, single-factor pollution index values, guideline values, and reported concentrations

		Be	B	V	Cr	Mn	Co	Ni	Cu	Zn	As	Se	Mo	Ag	Cd	Sb	Pb
Concentration in the present study ($\mu g\ L^{-1}$)	Maximum	60.5	113	60.6	97.5	968	64	87.7	77.4	363	54.8	51.7	56.4	49.1	52.7	54.7	412
	Mean	14.1	33.2	14.0	27.7	238	15.3	22.2	26.0	65.1	16.5	12.6	13.4	11.8	12.7	13.1	29.9
	Std.	9.97	13.2	8.55	17.6	199	10.7	14.1	15.9	48.2	8.52	8.26	9.18	7.78	8.36	8.33	51.4
Fraction (a.u.)	Mean	0.94	0.2	0.68	0.9	0.79	0.93	0.91	0.88	0.88	0.75	0.93	0.91	0.95	0.94	0.9	0.98
	Std.	0.18	0.16	0.32	0.18	0.24	0.19	0.2	0.18	0.2	0.18	0.18	0.21	0.17	0.18	0.19	0.11
Single-factor pollution index (a.u.)	Mean	n.d.	<0.01	n.d.	0.49	2.1	0.29	1.0	<0.01	<0.01	1.3	1.2	0.17	n.d.	3.8	5.7	2.1
	Std.	n.d.	<0.01	n.d.	0.26	2.0	0.18	0.61	<0.01	<0.01	0.74	0.68	0.12	n.d.	1.7	2.5	1.4
Bangladesh standard[1] ($\mu g\ L^{-1}$)		–	1,000	–	50	100	–	100	1,000	5	50	10	–	–	5	–	50
WHO guideline[2] ($\mu g\ L^{-1}$)		–	500	–	50	100	–	20	1,000	–	10	10	–	–	3	5	10
Japan standard[3] ($\mu g\ L^{-1}$)		n.d.	1,000	n.d.	20	200	50	10	1,000	5	10	10	70	n.d.	3	2	10
Reported values in the Buriganga River ($\mu g\ L^{-1}$)	Maximum	n.d.	n.d.	n.d.	180[4]; 60,000[5]	1,560[6]; 310[4]	19[6]; 400[4]	400[4]	81[6]; 990[4]	427[6]; 900[4]	220[4]; 7,000[5]	n.d.	n.d.	n.d.	90[4]	n.d.	210[4]; 22,000[5]
	Minimum	n.d.	n.d.	n.d.	12[4]; 40,000[5]	740[6]; 6[4]	5[6]; 90[4]	90[4]	25[6]; 100[4]	167[6]; 110[4]	5[4]; 4,000[5]	n.d.	n.d.	n.d.	30[4]	n.d.	100[4] 15,000[5]
Reported values in other Bangladesh rivers ($\mu g\ L^{-1}$)	Maximum				126[7]; 20[8]; 112[9]	500[8]	9[8]	71[7]; 300[8]	119[7]; 27[8]	40[8]	92[7]; 53[9]				22[7]; 18[8];18.3[9]		64[7]; 27.5[9]
	Minimum				33[7]; 46[9]			9.3[7]	23[7]		10[7]; 15[9]				1[7]; 2.5[9]		8[7]; 5.3[9]
Reported values of Japanese industrial[10] ($\mu g\ L^{-1}$)	Maximum	n.d.	n.d.	n.d.	600	n.d.	n.d.	n.d.	n.d.	n.d.	1,000	100	n.d.	n.d.	100	n.d.	1,700
	Minimum	n.d.	n.d.	n.d.	0	n.d.	n.d.	n.d.	n.d.	n.d.	0	0	n.d.	n.d.	0	n.d.	0

n.d., not determined; std., standard deviation.

Data taken from [1]Republic of Bangladesh Government of the People's (2019); [2]World Health Organization (2023); [3]Hayami *et al.*, (2022); [4]Bhuiyan *et al.* (2014); [5]Islam *et al.* (2014a); [6]Khan *et al.* (2014), [7]Islam *et al.* (2015); [8]Bhuyan *et al.* (2017); [9]Ali *et al.* (2016), [10]Ministry of the Environment Japan (2022).

RESULTS AND DISCUSSIONS

Trace metal concentration

The maximum trace metal concentrations found in this study were comparable to those reported in the previous studies, implying that the trace metal pollution status of the Buriganga River had not improved over the last decade (Table 1) (Mohiuddin *et al.* 2011; Bhuiyan *et al.* 2014; Islam *et al.* 2014a; Khan *et al.* 2014; Islam *et al.* 2015; Ministry of the Environment 2022). The trace metal concentrations in the Buriganga River reported in previous studies and in the present study were 100 times higher for Cr, 7 times higher for As, 9 times higher for Cd, and 13 times higher for Pb, than those reported in factory effluents in Japan, indicating a severe level of pollution (Table 1). As the trace metal concentrations measured in this study were comparable to those reported in previous studies on the Buriganga River and industrial wastewater in Japan, the risk of trace metal pollution was high, as discussed below (Table 1).

The Mn, Co, Zn, Cd, and Pb concentrations recorded in the present study were higher than those reported for other Bangladeshi rivers, such as the Korotoa, Meghna, and Karnaphuli Rivers (Table 1) (Islam *et al.* 2015; Ali *et al.* 2016; Bhuyan *et al.* 2017). Thus, the Buriganga River showed a relatively high level of pollution compared to that of other rivers, and the pollution status requires continuous investigation and monitoring. Conversely, large areas of Bangladesh may be contaminated by As and Cr, because the concentrations of As and Cr in the Buriganga River were similar to the concentrations of these metals in other rivers, and the concentrations observed exceeded the levels indicated in the guidelines (Islam *et al.* 2015; Ali *et al.* 2016; Bhuyan *et al.* 2017).

The mean concentrations (μg L^{-1}) of Be, B, V, Cr, Mn, Co, Ni, Cu, Zn, As, Se, Mo, Ag, Cd, Sb, and Pb were 14.1 ± 9.97, 33.2 ± 13.2, 14.0 ± 8.55, 27.7 ± 17.6, 238 ± 199, 15.3 ± 10.7, 22.2 ± 14.1, 26.0 ± 15.9, 65.1 ± 48.2, 16.5 ± 8.52, 12.6 ± 8.26, 13.4 ± 9.18, 11.8 ± 7.78, 12.7 ± 8.36, 13.1 ± 8.33, and 29.9 ± 51.4, respectively. The temperature ranged from 20.3 to 23.9 °C, and pH ranged from 5.4 to 9.1 (Supplementary Table S1). The observed concentrations were lower than the trace metal concentrations in the Buriganga River reported in previous studies but higher than the guideline values (Table 1), indicating that the degree of trace metal contamination in the Buriganga River is fairly unfavorable.

Fractional ratios

Figure 2 and Table 1 show the fractional ratios of dissolved metals to metals adsorbed on the SS. These metals should accumulate at high concentrations in the river sediments because the fraction ratios of Be, Cr, Co, Ni, Cu, Zn, Se, Mo, Ag, Cd, Sb, and Pb were high (>0.85). The fraction ratios of B, V, Mn, and As ranged from 0.22 (B) to 0.78 (Mn). Metals form different molecules depending on their valence, and the fraction ratio changes depending on the type of molecule (Skyllberg 2008). Metals with high fraction ratios, similar to those measured in this study, might exist as highly hydrophobic molecules in the river.

Comparing guideline values and observed values

Figure 3 and Table 1 compare the total metal concentrations measured in this study with the guideline values of the WHO, Japan, and Bangladesh. The mean values for Zn, Se, Cd, and Sb concentrations were higher than the guideline values, suggesting a high degree of contamination for these metals. Although the mean values of Cr, Mn, Co As, and Pb were

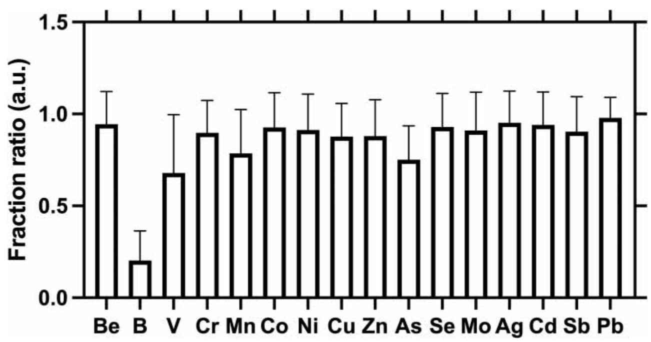


Figure 2 | Fraction ratios of metals. The number of samples was 210, and error shows one standard deviation.

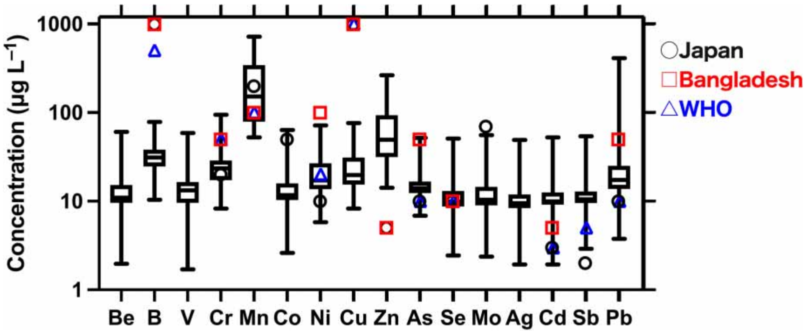


Figure 3 | Total concentration of metals collected from the Buriganga River. The number of samples was 210, and error shows one standard deviation. Circles, squares, and triangles show guideline values of Japan, Bangladesh, and the WHO, respectively.

lower than the guideline value, the maximum values recorded exceeded the guideline values, contributing to the pollution of the river environment. The mean and standard deviation of the total metal concentrations are shown in Table 1.

Assessing metal pollution

Figure 4 and Table 1 show the P_i values for each metal. Because the mean P_i value of Sb was 5.7, the pollution level for this metal was classified as 'serious pollution.' The International Agency for Research on Cancer categorizes antimony trioxide as group B2 ('possibly carcinogenic to humans'). Additionally, antimony trioxide is genotoxic; therefore, the morphology of Sb should be analyzed to accurately assess Sb pollution in the river.

Cd, Ni, and Pb were classified as 'heavily polluted' (Figure 4 and Table 1). Cd is highly toxic; for example, Cd-contaminated mine drainage discharged into the Jinzu River, which flows through Toyama Prefecture in Japan, caused itai-itai disease in individuals who consumed rice cultivated using contaminated river water (Aoshima 2016). A previous study found that Cd adsorbs on SS migrating from the river to the paddy fields (Faroon *et al.* 2012). Indeed, the rice paddy soils were found to be contaminated with high concentrations of Cd (1–10 mg Cd kg^{-1}) (Faroon *et al.* 2012). In light of the threat posed by Cd exposure to humans, future studies should examine Cd concentration in rice cultivated using water from the Buriganga River. Humans may also be exposed to Ni and Pb through the same pathway as Cd, given their high partitioning ratios to SS (Figure 2).

The P_i values and concentrations of Mn increased during the dry season (November–February), whereas the concentrations and P_i values of other elements did not increase (Figure 5 and Supplementary Fig. S1). Tukey's multiple comparison test revealed that the Mn concentrations in the dry season were significantly higher than those in the rainy season (Supplementary Table S2 and Figure 5). This may be a result of the dilution effect of rainwater on the temporal distribution of Mn in the Buriganga River (Whitehead *et al.* 2019). A consistent amount of Mn might be discharged into rivers throughout the year via industrial wastewater, and the Mn concentration in rivers probably increases during periods when the

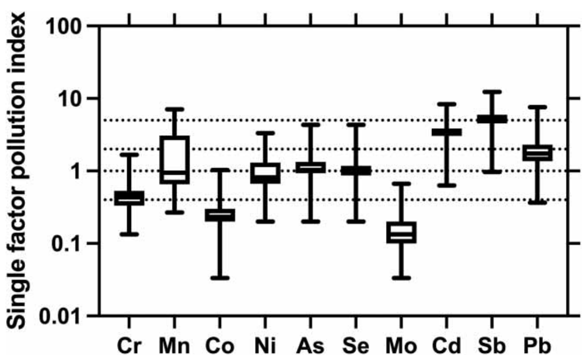


Figure 4 | Single-factor pollution index (P) of metals collected from the Buriganga River. The number of samples was 210, and error shows one standard deviation. Dotted lines show $P = 0.4$, 1, 1, and 5.

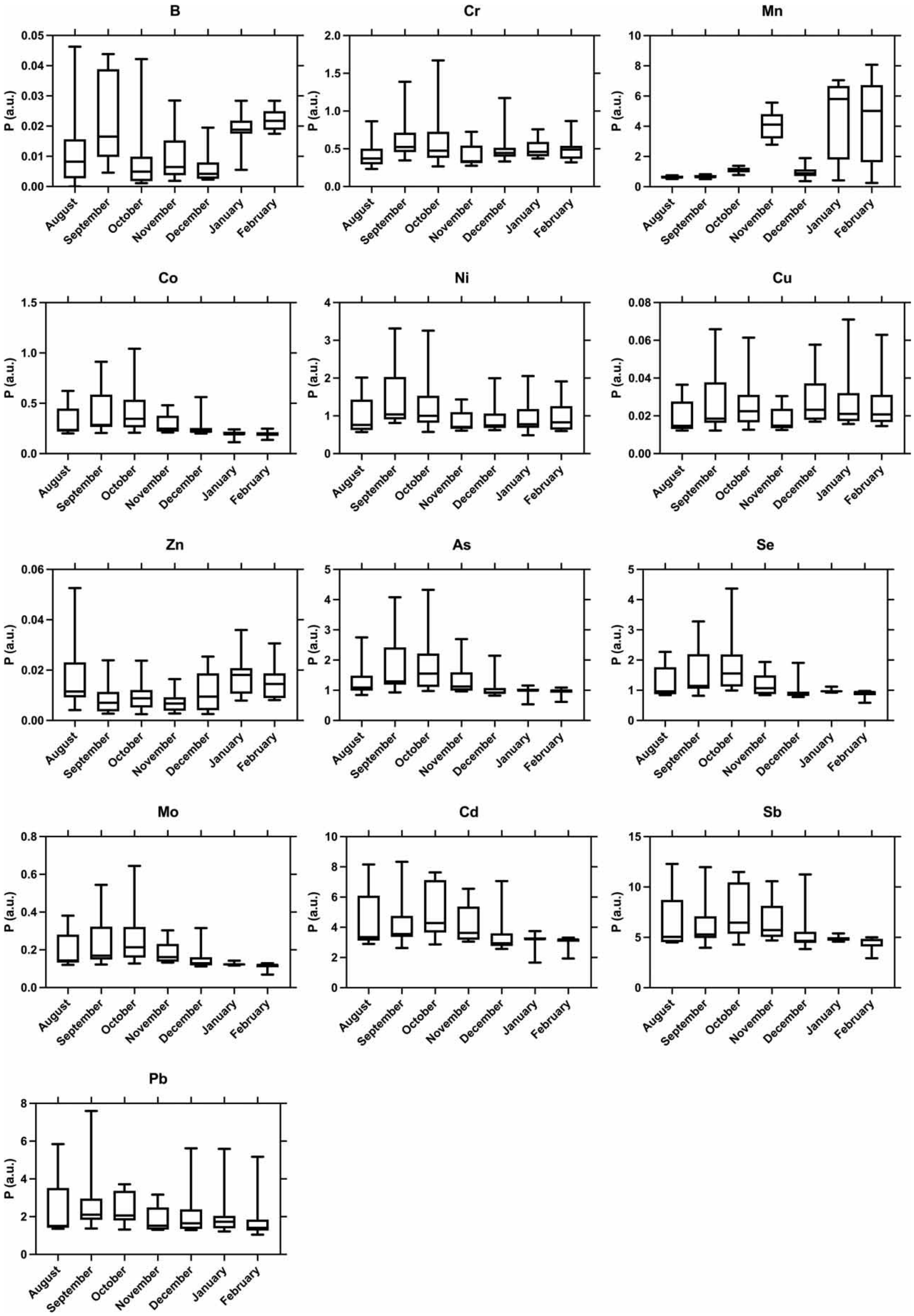


Figure 5 | Temporal distributions of the single-factor pollution index (*P*) of metals.

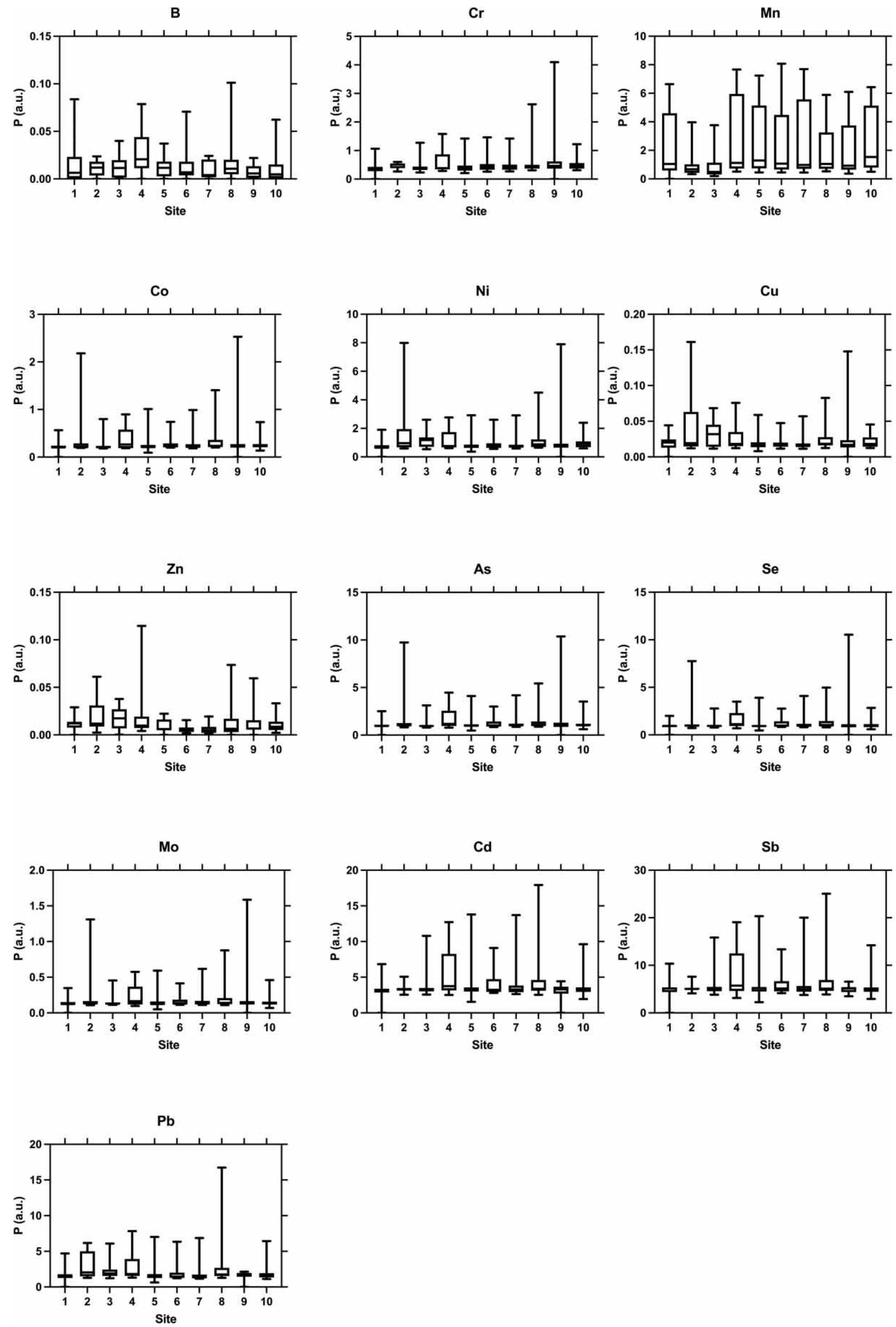


Figure 6 | Spatial distributions of the single-factor pollution index (*P*) of metals.

volume of river water is low. No seasonal changes were observed in the P_i values, except those for Mn. Consistent amounts of metals, except Mn, may leach from soils and waste and be carried into the river environment.

The spatial distribution of the P_i values was not observed (Figure 6 and Supplementary Table S3), as significant differences in the concentrations of different metals among sites were not detected, except in the case of B, Cu, and Zn (Supplementary Table S3). The lack of spatial distribution implies that the metals may not originate from the sampling area. Therefore, sources may be located upstream of the sampling points on the Buriganga River.

CONCLUSION

This study revealed that the Buriganga River was contaminated by anthropogenic activities because the Sb pollution was 'serious' in the Buriganga River, and Cd, Ni, and Pb pollution were 'heavily polluted.' Be, Cr, Co, Ni, Cu, Zn, Se, Mo, Ag, Cd, Sb, and Pb should accumulate in the river sediments because of their high particle-water fraction ratio. Additionally, these metals may accumulate on crops cultivated using contaminated river water. The Mn concentration was diluted by rainwater during the rainy season, whereas the concentrations of other elements did not show seasonal trends. The source of the elements may be distributed among the sources of the Buriganga River.

ACKNOWLEDGEMENTS

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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