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Analysis of Heavy Metal Pollution in the Water of the Halda River: A Distinctive Breeding Habitat for Carp Fish in Bangladesh

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ABSTRACT

Background: The present study aimed to assess the heavy metal concentrations in water from different sampling sites of the Halda River, the only natural breeding ground of Indian major carps.

Objectives: We have analysed the contamination level and seasonal variations of heavy metals (viz. Fe, Cu, Zn, Cr, Cd and Pb). The favourability of the breeding environment was assessed by different contamination indices.

Methods: Samples were collected from six sampling sites for four different seasons, and the metal concentrations were determined for water samples using Flame atomic absorption spectrophotometer.

Results: The obtained order of mean concentrations of heavy metals for four seasons was winter > spring > monsoon > pre-monsoon, and the assessed mean concentrations (mg/L) of heavy metals follow the trend: Fe (9.129) > Pb (0.033) > Zn (0.024) > Cr (0.017) > Cu (0.012) > Cd (0.002). The concentration of Fe was found above the permissible limit set by the World Health Organization (WHO) (Guidelines for Water Quality) and ECR (1997), and the concentrations for Pb were found above the USEPA (2006) limit in all four seasons. Contamination indices (heavy metal pollution Index [HPI] and metal index [MI]) indicate that during winter and monsoon, the river water metal condition is critical (HPI > 100) and the sites are moderately as well as slightly (1< MI <2) affected by the studied heavy metals. Contamination indices, such as contamination factors (C_f), degree of contamination (C_{deg}) and the pollution load index (PLI), demonstrate the fact that Fe and Pb remarkably contaminated the studied sampling sites. The Pearson correlation matrix and the principal component analysis (PCA) revealed that the studied metals are connected by their sources, which can be geological or anthropogenic.

Conclusion: Urgent initiatives need to be taken, and effective strategies must be implemented to control the different sources responsible for the entry of heavy metals into the Halda River water.

1 | Introduction

As per section 20 of the Bangladesh Water Act (2013)—'water pollution means direct or indirect harmful changes of physical, chemical and organic properties of water'. The introduction of effluviums containing different harmful chemicals and substances from unplanned urbanization and sewage systems, ineffective sanitary waste disposal systems and untreated or poorly treated domestic sewage are responsible for these changes in water in Bangladesh (Khuda 2020). These chemicals can be some naturally occurring metals having an atomic number greater than 20, and whose elemental density is greater than 5 g cm^{-3}, known as heavy metals (Ali and Khan 2018). Heavy metals can be essential (which are necessary for a living) or can be potentially toxic to biota depending on their dose and duration of exposure. Mn, Fe, Cu, Zn and so on are the essential heavy metals, and Cr, Ni, Cu, Zn, Cd, Pb, Hg, As and so on are biologically non-essential heavy metals, which can be hazardous for the aquatic ecosystem as well as aquatic lives (Ramirez 2013; Jovi´, Onjia, and Stankovi´c 2012; Khan et al. 2011). Volcanic activity, erosion of bedrock and weathering of minerals from soil (Bhardwaj, Gupta, and Garg 2017) are some natural sources (Zhang et al. 2009), and spinning mills, power plants, brickfields, industries, cultivable lands, domestic wastages, different water vehicles and so on are some anthropogenic sources (Bhuyan and Bakar 2017) of these toxic elements. The aquatic biota, including zooplankton, phytoplankton, crustaceans, micro-organisms, macro-algae and fish species, is being eradicated day by day (Islam et al. 2018) due to the non-biodegradable nature of heavy metals which are emitted into the river water from various natural and anthropogenic sources (Saha and Paul 2016). Like many other water resources, rivers like Halda, which is a river of a total length of 81 km (Akter and Ali 2012), one of the major tributaries of Karnaphuli River (Haque 1990), also receive a huge number of untreated discharges from many types of sources as this river passes through the industrial zone of Chittagong (Islam, Azadi, et al. 2020). This river is renowned as 'Bangabandhu Fisheries Heritage' (Islam 2021) and has gained national and international attention for its unique natural ecosystem (Azadi 1979). In recent years, the released untreated effluents from its nearby sources have increased the concentration of heavy metals in the aquatic condition of the Halda River (Bhuyan and Bakar 2017), which can cause a drastic economic loss as this river contributes to the economy of Bangladesh by being a natural breeding ground for pure Indian major carps (Rui, Katla, Mrigal and Kalibaush) (Tsai et al. 1981; Kabir et al. 2013). Considering the biological and environmental threat of these increasingly high concentrations of heavy metals in river water, it is necessary to assess the present status of heavy metals in Halda River water to preserve ecological sustainability as well as decrease the deleterious effects happening for this serious contamination (Islam, Das, et al. 2020). Therefore, the main focus of this current study is to determine the mean concentration of different heavy metals with their seasonal variations and also the favourability assessment for the carp breeding environment.

2 | Materials and Methods

2.1 | Site Description

The current study was conducted in the Halda River, which is one of the major tributaries of the Karnaphuli River located in the southeastern region of Chittagong, Bangladesh. Sample collection was started from downstream to upstream (Kalurghat–Garduara) covering around 20 km, and six sampling sites were selected (Figure 1), among which three of them were Halda—Karnaphuli Confluence (22° 24.382′ N and 91° 53.436′ E), Chayar Char (22° 25.756′ N and 91° 53.250′ E) and Khandakia Khal (22° 25.832′ N and 91° 52.507′ E), which receive direct discharges and untreated effluents from industries. Because this river serves as a unique natural breeding ground, three breeding zones were selected as sampling sites named Horekrishno-Mohajoner Tek (22° 27.905′ N and 91° 51.400′ E), Napiter Ghona (22° 28.130′ N and 91° 52.412′ E) and West Binajuri (22° 29′59″ N and 91° 84′56″ E) to assess the favourability for breeding environment. The renowned Kalurghat Industrial Area as well as different industries, mills and garments are located near the Halda–Karnaphuli confluence. Chayar Char is only 2 km away from the Halda–Karnaphuli confluence, where several brickfields are present on the riverbank. Krishnakhali canal, which carries wastages from different sources, is also present in this region. Khandakia Khal, adjacent to the Madari Khal and Modunaghat, is mainly responsible for carrying contaminants like Industrial and sewage discharges from several industries, mills, factories, tannery and paper mills. Engine-operated water vehicles, more than 30,000 ha of croplands (The New Nation 2009), present on the riverbank, tea gardens, a local sanitation system for tobacco farming and setting up a rubber dam on the rivers upstream and so on, are the causes of disrupting the water quality of the breeding zone (Horekrishno-Mohajoner Tek, Napiter Ghona and West Binajuri) (Bhuyan and Bakar 2017). Due to receiving a huge amount of untreated effluents from different sources, heavy metal contamination in Halda river water has become a matter of major concern nowadays. Taking into consideration the increased pollution, which is not only impacting the health of the river but also our national economy, motivated us to conduct this study.

2.2 | Sample Collection

In this study, a total of 24 water samples were collected in four phases starting from 2021 to 2022: first in September 2021 (monsoon season); second in January 2022 (winter season); third in March 2022 (spring season); and fourth in June 2022 (pre-monsoon season). Each sample was collected from about 5 cm below the surface. Before collection, the polyethylene sampling bottles were pre-conditioned with 5% nitric acid and later rinsed thoroughly with distilled de-ionized water. Every sampling bottle was marked with an identification code containing information about their collection sites and seasons. Then the pre-cleaned polyethylene sampling bottles were dipped into 5 cm below, and about 0.5 L/500 mL of the water samples were collected from each sampling site.

2.3 | Preservation and Processing of the Samples

Samples were brought to the laboratory immediately after collection and acidified with 1 mL HNO_3 (70%) to keep the metal state stable in solution. After that, by using Whatman No. 42 filter paper, samples were filtrated. A blank was prepared, using de-ionized water and the same acid ratio as the real samples but without the collected water sample. The standard solution was also prepared for each studied element. The prepared acid blank and the standards were analysed by Flame atomic absorption spectrophotometer (FAAS). After filtration, the real samples were also analysed, and the determination of metals was carried out, using a FAAS present in the Atomic Energy Centre (BAEC), Chattogram, for determining the concentration of Fe, Zn, Cu, Pb, Cd and Cr.

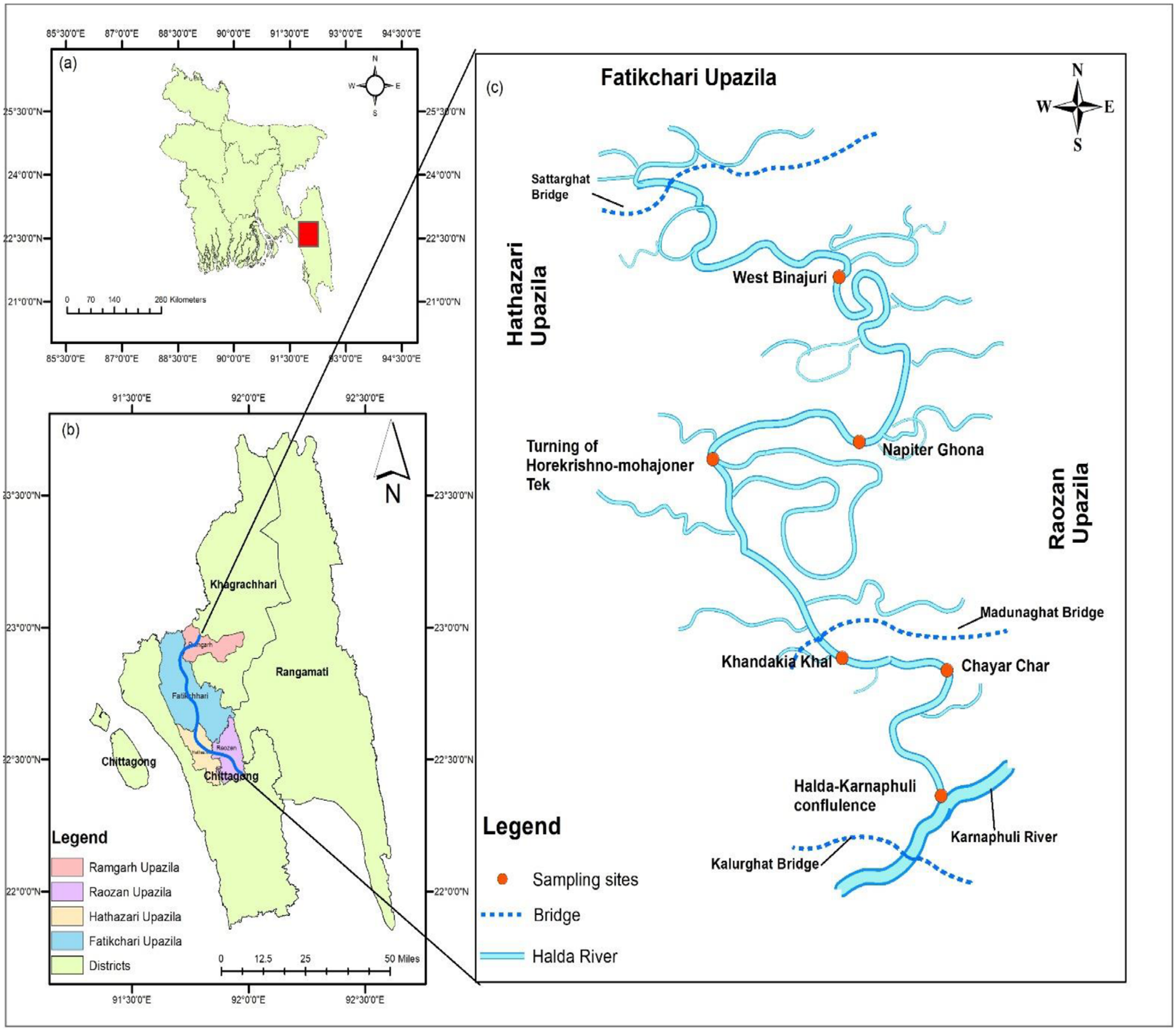

FIGURE 1 | Location of Halda River: (a) location of Halda River in Bangladesh; (b) location in upazilas of Chattogram division; (c) different sampling sites of Halda River in Chattogram district (Mapping software ArcGIS version 10.8).

Spectral lines and the instrumental detection limit for the elements measured using Atomic Absorption Spectrophotometer

Elements	Cr	Cu	Cd	Fe	Zn	Pb
Wavelength (nm)	357.9	324.8	228.8	248.3	213.9	283.3
Detection limit (mg/L)	0.02	0.01	0.001	0.02	0.005	0.05

2.4 | Heavy Metal Pollution Index (HPI)

HPI is a powerful technique for the assessment of water quality based on heavy metal concentration. It is a method of rating that shows the composite influence of individual heavy metals on the overall quality of water. Water quality and its suitability for drinking purposes can also be examined by determining its quality index (Mohan, Nithila, and Reddy 1996; Prasad and Kumari 2008; Prasad and Mondal 2008).

The heavy metal pollution index is calculated by the following equation:

$$\mathrm{HPI} = \sum_{i=1}^{n} (Q_i W_i) / \sum_{i=1}^{n} W_i,$$

where Q_i is the sub-index of the ith parameter, W_i is the unit weight of the ith parameter; and n is the number of parameters considered.

The weightage of ith parameter is

$$W_i = k/S_i$$

where W_i is the unit weightage, S_i is the recommended standard for ith parameter ($i = 1$–6), k is the constant of proportionality and the individual quality rating is given by the expression

$Q_i = 100\ V_i/S_i$; where Q_i is the sub-index of the ith parameter, V_i is the monitored value of the ith parameter in mg/L and S_i is the standard or permissible limit for the ith parameter (Appiah-Opong et al. 2021). In this study, the S_i value was obtained from the World Health Organization (WHO) standard permissible value for drinking water. The HPI value <100 indicates low ecological risk, HPI = 100 indicates heavy metal pollution on the threshold risk and HPI >100 indicates high heavy metal pollution (critical pollution index) (Prasad and Bose 2001).

2.5 | Metal Index (MI)

The MI was calculated using the following formula:

$$\mathrm{MI} = \sum_{i=1}^{n} \frac{C_i}{(\mathrm{MAC})_i};$$

where C_i represents each element's mean concentration, and MAC is the maximum allowable concentration of the same metal (Tamasi and Cini 2004). According to Caeiro et al. (2005), the MI values were divided into six groups: MI value: <0.3 = very pure, MI value: 0.3–1 = pure, MI value: 1–2 = slightly affected, MI value: 2–4 = moderately affected, MI value: 4–6 = strongly affected and MI value: >6 = seriously affected.

2.6 | Contamination Level Assessment of the Breeding Environment

2.6.1 | Contamination Factor and Degree of Contamination

In our present work, metal contamination level was assessed as described by Pal (2012). The degree of contamination ($C_{\deg}$) is the sum of contamination factors (C_f) for all the elements that represent metal contamination of the sample (Hakanson 1980). The contamination factor and the degree of contamination by the six heavy metals in Halda River water were determined by the following equation:

$$C_f = C_i/C_n$$

where C_f is the contamination factor of a particular heavy metal, C_i is the mean concentration of metal and C_n is the concentration of a reference value for individual metal. In this study, C_n is the reference value taken from Alam et al. (2022) for Cr, Cu, Cd and Zn, and reference values for Fe and Pb were taken from Islam, Das, et al. (2020):

$$C_{\deg} = \sum C_f$$

where $C_{\deg}$ is the degree of contamination, which is determined by the sum of $C_{f.}$

The contamination factors (C_f) and the degree of contamination ($C_{\deg}$) can be categorized as $C_f < 1$ = low contamination; $1 \leq C_f < 3$ = moderate contamination; $3 \leq C_f < 6$ = considerable contamination; $6 \leq C_f$ = very high contamination and $C_{\deg} < 6$ = low degree of contamination; $6 \leq C_{\deg} < 12$ = moderate degree of contamination; $12 \leq C_{\deg} < 24$ = considerable degree of contamination; $24 \leq C_{\deg}$ = very high degree of contamination, respectively (Suryawanshi et al. 2016).

2.6.2 | Pollution Load Index (PLI)

To determine the effect of metal contamination in different sampling stations, the PLI was calculated (El-sammak and Aboul-Kassim 1999). The PLI for each sampling site has been estimated by the multiplications of the nth root of the studied heavy metals (Tomlinson et al. 1980):

$$\mathrm{PLI} = \left(C_f 1 \times C_f 2 \times C_f 3 \times \ldots C_f n\right)^{1/n}$$

where C_f is the contamination factor and n is the number of samples (Bentum et al. 2011). The PLI > 1 indicates metal pollution, whereas PLI < 1 indicates no heavy metal pollution (Harikumar et al. 2009).

2.7 | Statistical Analysis

Correlation matrix and multivariate analysis (principal component analysis [PCA]) among the variables were done after the determination of mean concentrations and standard deviations. To determine the association among the variables, Pearson's correlation matrix ($n - 1$) was used, and the analysis was done using Microsoft Excel, 2016. In general, PCA converts the dimension of large sets of variables to a small set of variables (Dey et al. 2021). To find out the standard features of variations in the data set and evaluate the probable sources of heavy metals, PCA was performed. In this study, factors with an eigenvalue >0.5 were extracted for the PCA (Bhuyan and Bakar 2017). The data were processed using Microsoft Excel and IBM SPSS Statistics software (version 29).

3 | Results and Discussion

3.1 | Seasonal Variation of Metal Concentration in Water Samples

The mean concentrations of heavy metals found in the water samples collected in different seasons from the Halda River are shown in Table 1. The mean concentrations of heavy metals for different seasons follow the order: Fe > Pb > Zn > Cr > Cu > Cd, where the mean concentration of Fe was highest (Figure 2) in all four seasons and Cd was below the detection limit for the spring and pre-monsoon, which implies no toxicity (Table 1). The concentration of Pb in all four seasons crosses the allowable limit set by both WHO (Guidelines for Water Quality) and USEPA (2006). Fe concentrations were found above the permissible limit set by WHO (Guidelines for Water Quality) and ECR (1997) in all four seasons. Apart from that, the concentrations for Cu were found above the USEPA (2006) limit in winter (Table 1). Metal concentrations for Cr, Cd and Zn were found within the WHO, USEPA and ECR critical limits.

In case of winter, most of the metals were found in the highest concentration except Zn and Pb, which were highest in monsoon. Table 1 shows that, in the Halda–Karnaphuli confluence and

TABLE 1 | Seasonal variation of heavy metals concentration (mg/L) in water samples of Halda River and their comparison with International standards.

Sites	Seasons	Cr	Cu	Cd	Fe	Zn	Pb
Confluence	Monsoon	0.004	0.013	BDL	13.9	0.048	0.045
Chayar Char		0.009	0.010	BDL	19.37	0.050	0.045
Khandakia Khal		BDL	BDL	BDL	4.187	0.021	0.036
Horekrishno-Mohajoner Tek		BDL	BDL	BDL	2.925	0.015	0.036
Napiter Ghona		BDL	0.003	0.001	4.182	0.021	0.036
West Binajuri		BDL	0.003	0.001	4.205	0.026	0.036
	Mean ± SD	0.007 ± 0.004	0.007 ± 0.005	0.001 ± 0	8.128 ± 6.830	0.030 ± 0.015	0.039 ± 0.005
Confluence	Winter	0.041	0.047	0.001	43.45	0.095	0.063
Chayar Char		BDL	BDL	0.002	5.720	0.013	0.036
Khandakia Khal		BDL	0.003	0.001	7.271	0.021	0.036
Horekrishno-Mohajoner Tek		BDL	BDL	0.002	3.594	0.009	0.027
Napiter Ghona		BDL	0.003	0.002	5.908	0.012	0.027
West Binajuri		BDL	0.014	0.001	4.481	0.023	0.036
	Mean ± SD	0.041	0.017 ± 0.021	0.002 ± 0.001	11.737 ± 15.587	0.029 ± 0.033	0.037 ± 0.013
Confluence	Spring	0.009	0.007	BDL	5.700	0.015	0.036
Chayar Char		0.014	0.010	BDL	6.072	0.017	0.027
Khandakia Khal		0.004	0.003	BDL	1.487	0.003	0.027
Horekrishno-Mohajoner Tek		0.004	0.007	BDL	2.983	0.005	0.027
Napiter Ghona		0.041	0.034	BDL	33.2	0.072	0.045
West Binajuri		0.004	0.007	BDL	2.625	0.006	0.027
	Mean ± SD	0.013 ± 0.014	0.011 ± 0.011	BDL	8.678 ± 12.147	0.019 ± 0.026	0.032 ± 0.007
Confluence	Pre-monsoon	0.023	0.027	BDL	19.53	0.046	0.036
Chayar Char		0.004	0.010	BDL	5.683	0.015	0.027
Khandakia Khal		0.009	0.010	BDL	8.85	0.023	0.027
Horekrishno-Mohajoner Tek		0.004	0.010	BDL	3.449	0.013	0.018
Napiter Ghona		0.004	0.007	BDL	3.715	0.007	0.018
West Binajuri		0.009	0.0135	BDL	6.614	0.015	0.027
	Mean ± SD	0.008 ± 0.007	0.013 ± 0.007	BDL	7.973 ± 6	0.019 ± 0.014	0.025 ± 0.007
Guidelines for Water Quality 3rd edition by WHO		0.050	2	0.003	0.300	3.00	0.010
USEPA (2006)		0.1	0.013	0.0025	—	0.12	0.0025
ECR (1997)		0.05	1	0.005	0.3–1	5	0.05

Abbreviations: BDL = Below Detection Limit; WHO = World Health Organization.

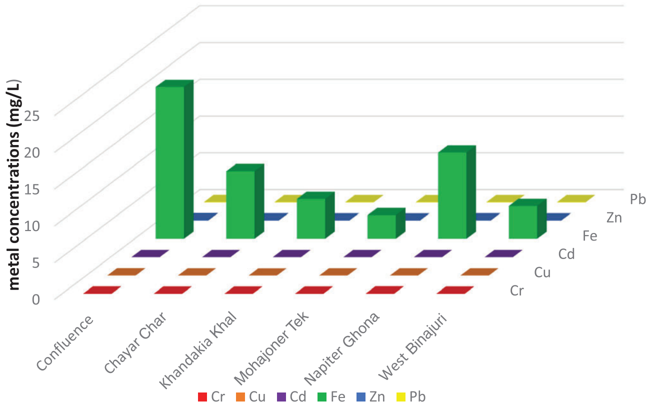

FIGURE 2 | Graphical representation of the studied metal concentration in the sampling sites.

Napiter Ghona, Cr concentration is maximum during winter and spring, respectively. Cu, Fe, Zn and Pb concentrations are highest in the Halda–Karnaphuli confluence during winter, whereas Cd reaches its maxima in Chayar Char, Horekrishno-Mohajoner Tek and Napiter Ghona during the same season. The lowest metal concentrations were found for Fe as well as Pb, but Cd could not be detected in spring and pre-monsoon. From all the sampling sites, the Horekrishno-Mohajoner Tek possesses the lowest metal concentrations during monsoon and winter (Table 1). The order of metal concentrations found in the four studied seasons is as follows: winter > spring > monsoon > pre-monsoon (Figure 3). The concentration variation of different heavy metals in river water depends on different sources, the velocity of the river flow and human activities, such as settlements, farms, car wash and mining industries that occur near the river water (Chaves et al. 2016). During winter, the heavy metal concentration becomes high due to reduced water volume, which can result in a drastic change in metal concentration even if a small amount of waste is discharged in the river water (Islam et al. 2015). High precipitation dilutes the river water, and also, by the increased flow, turbulence is created, which is responsible for carrying away metals, resulting in low metal concentration. The beginning of the dry season (winter) leads to a rise in metal concentration (Shanbehzadeh et al. 2014) for the low flow state of the river (Dey, Das, and Manchur 2015), which again decreases at the ending of the dry season (spring). According to Islam et al. (2017), industrial waste (53%), sewage contamination (20%), tobacco farming (13%), rubber dams (8%) and sand extraction (6%) are the reasons for the increase of the contamination of Halda River water.

In the present study, the overall mean concentration of Cr was recorded 0.017 mg/L, and the highest concentration (0.041 mg/L) was found for both Confluence and Napiter Ghona during winter and spring, which is within the permissible limits set by different international guidelines like WHO, USEPA and ECR (Table 1). In 2022, Alam et al. (2022) recorded 0.116 mg/L, and Bhuyan and Bakar (2017) found 0.06 mg/L Cr in Halda River water. Khatun et al. (2021) found 0.091 mg/L Cr in the Karnaphuli River. Cr was assessed by Ali et al. (2016) at 0.08 mg/L in the Karnaphuli River, where 0.069 mg/L was recorded in summer and 0.087 mg/L in winter. In the Tembi River, upstream of 0.19 mg/L Cr was recorded by Shanbehzadeh et al. (2014). Cr compounds are generally used in leather tanning, dyes, pigments, industrial welding, wood preservations and so on (Barnhart 1997). The non-essential element Cr concentration in the Halda River may be related to the tanneries and shipping-related activities near the Halda River (Brady et al. 2014).

The highest Cu concentration was found in Halda–Karnaphuli confluence (0.047) mg/L in winter, which is below the permissible limits set by WHO and ECR but above the limit set by USEPA (2006) (Table 1). Cu of 0.038 mg/L was recorded in the Karnaphuli River by Khatun et al. (2021) and 0.10 mg/L in the Halda River water by Bhuyan and Bakar (2017). Didar-Ul Islam and Azam (2015) found 0.082, 0.031 and 0.215 mg/L Cu concentration in the Buriganga River water during pre-monsoon, monsoon and post-monsoon, respectively. Shanbehzadeh et al. (2014) reported 0.47 mg/L in upstream of the Tembi River. The amount of Cu was recorded as 0.017 mg/L in the Turag River by Zakir, Sharmin, and Shikazono (2006). Bhuyan and Bakar (2017) found the highest Cu in the Halda River water during post-monsoon, and similarly, the present study recorded the highest Cu concentration during winter (post-monsoon) (Table 1). Agricultural fields, agrochemical industries and urban sewage present near the Halda River are the sources of Cu, which may be responsible for the introduction of Cu in the Halda River water (Islam et al. 2015).

During winter, the maximum amount of Cd was recorded at 0.002 mg/L in Horekrishno-Mohajoner Tek, Chayar Char and in Napiter Ghona, which is below the tolerable limit set by WHO, USEPA and ECR (Table 1). On the other hand, Cd concentration is below the detection limit found for both spring and pre-monsoon, indicating no Cd toxicity in these seasons. In Halda River water, 0.038 mg/L Cd was assessed by Alam et al. (2022). The Cd concentration in Karnaphuli River was determined by Khatun et al. (2021), which was found below the detection limit. Bhuyan and Bakar (2017) found 0.03 mg/L in the Halda River water, Ali et al. (2016) recorded 0.008 mg/L Cd in Karnaphuli River water, Hasan et al. (2015) recorded 0.003 mg/L Cd in the Meghna River and 0.009 mg/L Cd in the Buriganga River water was determined by Ahmed et al. (2010). Metal industry, coal combustion and waste disposal are the major sources of the toxic element cadmium (Rzętała 2016). Cd is one of the most toxic metals, which is related to agricultural pesticides and fertilizers. So, the presence of the metal industry and agricultural activities near the Halda Riverbank may be the reason for Cd's existence in the river water (Green-Ruiz and Paez-Osuna 2001; Maanan et al. 2014).

The mean concentration of Fe was found, 9.129 mg/L (Table 2) and the concentrations of Fe found in Halda River water cross different international guidelines and permissible limits in all the four seasons (Table 1). In the water of Karnaphuli River, Fe concentration was found at 3.340 mg/L, studied by Khatun et al. (2021), and in the Meghna River, 1.0224 mg/L was recorded by Hasan et al. (2015). Islam, Azim, and Islam (2008) mentioned Fe concentration ranged between 0.00126 and 0.00831 mg/L in Sitalakkhya River water. The findings of Fe concentration in the present study were 8.128, 11.737, 8.678 and 7.973 mg/L during monsoon, winter, spring and pre-monsoon, respectively, but in the Buriganga River, Fe concentrations were 0.74, 1.79 and 1.34 mg/L in monsoon, post-monsoon and pre-monsoon, respectively, studied by Didar-Ul Islam and Azam (2015). High concentrations of Fe in the river water could be associated with effluent discharge from metal alloy industries (Dey et al. 2021).

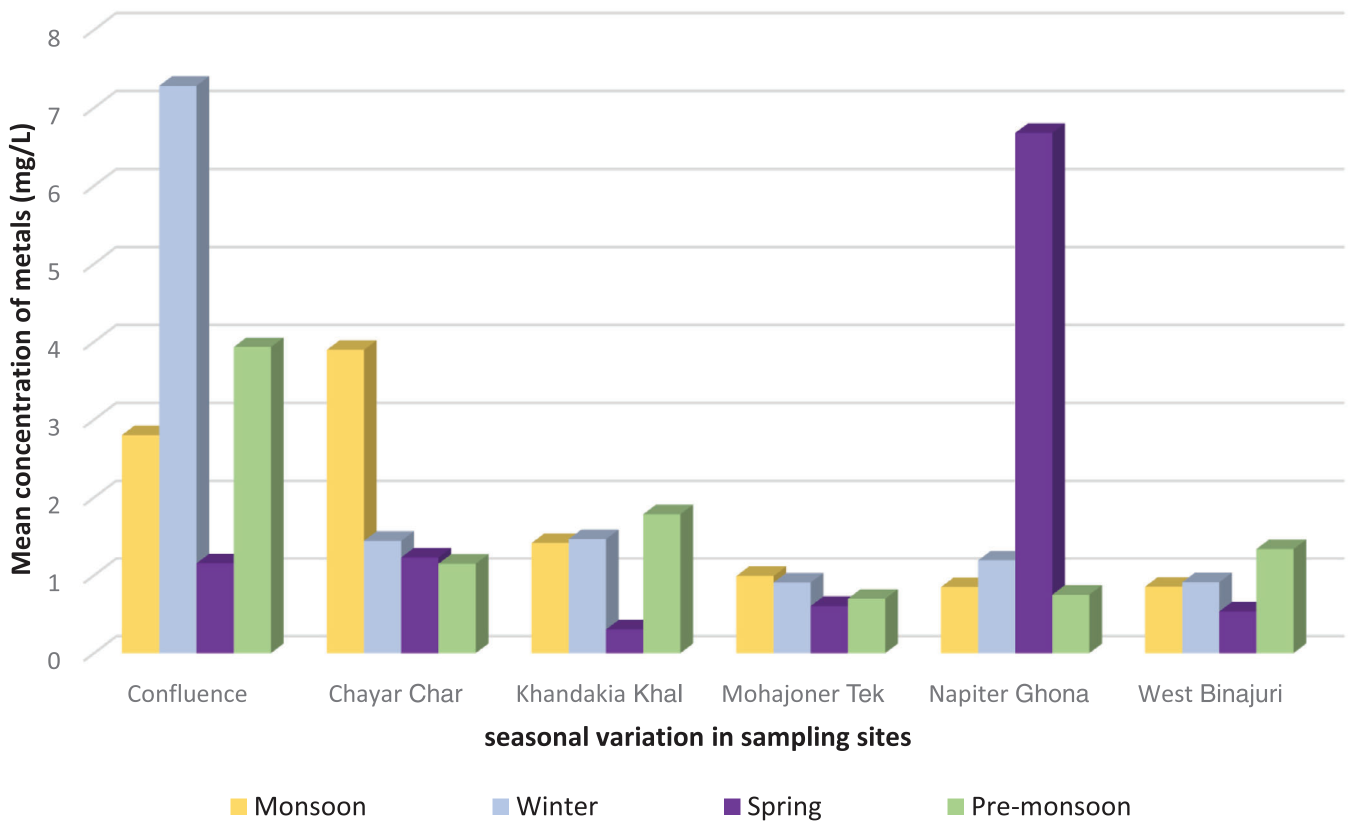

FIGURE 3 | Graph showing mean concentrations of heavy metals in the sampling sites during four seasons.

TABLE 2 | Comparison of heavy metal concentrations (mg/L) of Halda River water with other national and international rivers.

River	Cr	Cu	Cd	Fe	Zn	Pb	References
Halda River	**0.017**	**0.012**	**0.002**	**9.129**	**0.024**	**0.033**	**Present study**
Halda River	0.116	0.029	0.038	1.832	0.212	—	Alam et al. (2022)
Halda River	0.06	0.10	0.03	—	0.35	0.07	Bhuyan et al. (2017)
Brahmaputra River	0.01	0	0.01	—	0.02	0.02	Islam, Rahman, and Khan (2016)
Balu River	0.02	0.010	0.020	—	0.08	0.032	Islam, Rahman, and Khan (2016)
Buriganga River	0.114	—	0.059	—	0.332	0.112	Bhuiyan et al. (2015)
Dhaleshwari River	0.44	0.15	0.006	—	—	0.05	Ahmed et al. (2009)
Dhaleshwari River	0.13	0	0	—	—	0.20	Ahmed et al. (2012)
Karnaphuli River	0.25	0.05	0.01	—	0.28	0.14	Islam et al. (2013)
Karnaphuli River	0.09	0.038	BDL	3.340	BDL	BDL	Khatun et al. (2021)
Meghna River	0.035	—	0.003	—	0.036	BDL	Hasan et al. (2015)
Sitalakkhya River	0.08	0.04	0.003	—	0.72	0.05	Islam et al. (2014)
Turag River	0.030	0.017	—	0.23	0.082	0.014	Zakir, Sharmin, and Shikazono (2006)
Tembi River	0.19	0.47	—	—	0.20	—	Shanbehzadeh et al. (2014)

The concentration of Zn ranged between 0.003 and 0.095 mg/L (Table 1), and the overall mean concentration was found to be 0.024 mg/L, where the maximum mean concentration of Zn was recorded in monsoon (Table 1). Zn concentration was found to be 0.212 mg/L in Halda River water by Alam et al. (2022). Khatun et al. (2021) could not detect Zn in the Karnaphuli River water, whereas Islam et al. (2013) recorded 0.28 mg/L Zn concentration in the Karnaphuli River. In the water of the Brahmaputra River, Zn was recorded at 0.02 mg/L, as determined by Islam, Rahman, and Khan (2016). Bhuyan and Bakar (2017)

TABLE 3 | Heavy metal pollution index [HPI] of the collected water samples.

Seasons	HPI values	Status
Monsoon	129.866	High heavy metal pollution
Winter	161.519	High heavy metal pollution
Spring	92.139	Low heavy metal pollution
Pre-monsoon	74.691	Low heavy metal pollution

found a 0.48 mg/L concentration of Zn in the surface water of the Halda River. Zn of 0.036 mg/L was recorded in the Meghna River water by Hasan et al. (2015), and 0.20 mg/L Zn was recorded by Shanbehzadeh et al. (2014) in the upstream of the Tembi River. Didar-Ul Islam and Azam (2015) recorded Zn concentrations of 0.0431, 1.021 and 0.452 mg/L in pre-monsoon; 0.017, 0.338 and 0.331 mg/L in monsoon; and 0.081, 0.846 and 0.561 mg/L in post-monsoon in Sitalakkhya, Buriganga and Turag River, respectively. The current study showed that during winter, Zn concentration becomes highest in the Halda–Karnaphuli confluence (Table 1). The introduction of Zn into the waterbodies can occur through artificial pathways, such as coal-fired power stations, steel production and burning of waste materials (Wuana and Okieimen 2011).

Pb concentration in the present study is 0.0391 mg/L, and the highest concentration found for monsoon that crosses the WHO and USEPA (2006) permissible limit (Table 1). Bhuyan and Bakar (2017) found 0.07 mg/L Pb in Halda River. Ali et al. (2016) found 0.013 mg/L in the Karnaphuli River, 0.112 mg/L concentration was found in the Buriganga River recorded by Bhuiyan et al. (2015) and 0.014 mg/L concentration recorded by Zakir, Sharmin, and Shikazono (2006) in the Turag River. Hasan et al. (2015) and Khatun et al. (2021) found Pb concentration below the detection limit from the Meghna and the Karnaphuli River water, respectively. The minimum concentration for Pb was found during pre-monsoon in this study (Table 1). The major sources of Pb are vehicles, exhaust, metal plating, wastewater discharge and fertilizers (Karrari, Mehrpour, and Abdollahi 2012). The high concentration of Pb found in this study may be related to the release of effluents from industries located on the nearby riverbank.

3.2 | HPI in Water Samples

For the determination of the water quality for the studied four seasons, HPI was assessed. The HPI presented in Table 3 was calculated by using the mean concentrations for the winter, spring, pre-monsoon and monsoon.

The mean HPI recorded for the four seasons (monsoon—129.866, winter—161.519, spring—92.139 and pre-monsoon—74.691) indicates that water quality was degraded because of high metal pollution during monsoon and winter, according to Prasad and Bose (2001). The HPI value was found to be highest in winter (161.519), which leads to the conclusion that for elevated sunlight, water volume decreases, which results in higher metal concentrations. Besides this, during the start of the dry season (winter), the riverbank is used for agricultural activities, which leads to the high HPI value. The water volume plays a very important role in the heavy metal contamination rate. The high HPI values for monsoon and winter cross the critical index value (100), which means Halda River water is highly contaminated with heavy metals during these seasons. Increased runoff from urban areas, agricultural lands and industrial sites due to heavy rainfall might be the reason for high HPI value during the monsoon season.

TABLE 4 | Metal index [MI] of water samples for the four seasons.

Seasons	$\mathrm{MI} = \sum_{i=1}^{n} C_i/(\mathrm{MAC})_i$	Status
Monsoon	1.531	Slightly affected
Winter	2.212	Moderately affected
Spring	1.632	Slightly affected
Pre-monsoon	1.498	Slightly affected

3.3 | Metal Index

MI is used to evaluate the level of metal contamination in river water. The mean MI value presented in Table 4 for the four seasons shows that, in recent years, the Halda River water is affected by metal pollution. According to Caeiro et al. (2005), (1 < MI < 2) indicates river water samples are slightly affected during monsoon, spring and pre-monsoon. In the winter season, the Halda River water is moderately affected by the studied metals (MI > 2). This result could be correlated with various large-scale as well as many small-scale industry discharges released in different seasons located near the Halda River.

3.4 | Favourability Assessment for Carp Breeding Environment

To assess the favourability of breeding environment for carp fishes (Rui, Catla, Mrigal and Kalibaush) and the level of metal contamination in the Halda River water, different contamination indices such as contamination factors (C_f), degree of contamination (C_{deg}) and the pollution load index (PLI) were analysed.

The contamination factors (C_f) and the degree of contamination of the sampling sites are presented in Table 5. The contamination factor analysis indicates that very high contamination ($C_f > 6$) and considerable contamination ($3 \le C_f < 6$) for Fe were found in the Halda–Karnaphuli confluence and in Napiter Ghona, respectively. Moderate Fe contamination ($1 \le C_f < 3$) was found in Chayar Char, Khandakia Khal and West Binajuri. All the sampling sites were moderately contaminated with Pb ($1 \le C_f < 3$). The contamination factors for Cr, Cu, Cd and Zn indicate low metal contamination with these metals in all the sampling sites (Suryawanshi et al. 2016).

Moderate ($6 \le C_{\mathrm{deg}} < 12$) degree of contamination was found in the Halda–Karnaphuli confluence (Table 5). Loadings of the metals are highest in Halda–Karnaphuli confluence, which is presented in Figure 4. The presence of industries, power plants

TABLE 5 | The contamination factors and the degree of contamination of six sampling sites.

	The contamination factor (C_f)						
Sampling sites	**Cr**	**Cu**	**Cd**	**Fe**	**Zn**	**Pb**	**Degree of contamination (C_{deg})**
Confluence	0.166	0.815	0.026	6.218	0.241	1.875	9.340
Chayar Char	0.076	0.346	0.053	2.774	0.112	1.404	4.766
Khandakia Khal	0.056	0.184	0.026	1.641	0.080	1.313	3.30
Horekrishno-Mohajoner Tek	0.034	0.293	0.052	0.975	0.049	1.125	2.53
Napiter Ghona	0.194	0.405	0.039	3.539	0.132	1.312	5.623
West Binajuri	0.056	0.323	0.026	1.349	0.082	1.312	3.149

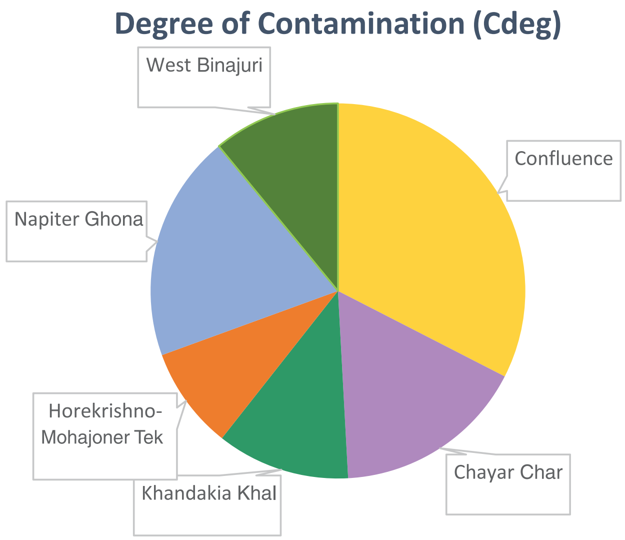

FIGURE 4 | Representation of degree of contamination in the sampling sites.

and mills at Kalurghat near the Halda–Karnaphuli confluence may be the reason for a higher percentage of the degree of contamination than the other sampling sites. A low degree of contamination was found for other sampling sites. The degree of contamination of heavy metals found in Napiter Ghona, which is one of the breeding zones of the river, may be due to the flowing upstream water, which carries pesticides and herbicides thrown from riverbank tea gardens and agricultural lands. As the degree of contamination illustrates the metal enrichment of each metal, in this study the degree of contamination of the sampling sites increases because of the slightly high concentration of Fe and Pb. The discharges from textiles, leather tanning chemical industries and so on established near the riverbank may be responsible for the high Fe contamination. The reason for a high degree of Pb contamination may be the discharge of untreated industrial, agricultural and urban effluent into the river.

To evaluate the overall contamination level of the heavy metals and the sampling sites, the PLI was assessed in this study. The PLI for all sampling sites and metals is presented in Table 6. In the present study, the PLI ranges from 0.189 to 0.463 in the studied sampling sites, which indicates no metal pollution in the studied sampling sites (Harikumar et al. 2009). On the other hand, the PLI values for Cr, Cu, Cd, Fe, Zn and Pb range from 0.035 to 6.865. As metals, such as Fe and Pb have PLI > 1, it indicates that the sampling sites are contaminated with these heavy metals, which might be due to the discard of untreated industrial, agricultural and urban effluents into the river water.

3.5 | Relationship Among the Concentrations of Heavy Metals Analysed in the Water Samples

In river water systems the correlations among metals give information about their sources and their pathways. Correlations like very strong, strong and moderate express the significant relations of metals and their similar source of origin. So, for determining the relationships among studied metals and their sources, a Pearson correlation matrix ($p < 0.05$ and $p < 0.01$) is shown in Table 7, which indicates that the studied metals are significantly correlated. It suggests that Fe versus Zn (0.967) shares a very strong linear positive relationship at the significance level of 0.01. A strong relationship was found in Zn versus Pb (0.866) and Fe versus Pb (0.793).

The strong positive relationship of Fe with other metals like Zn and Pb reveals that these metals belong to the same origin, which may be lithogenic (Islam et al. 2020). Cu versus Fe (0.472) shows moderate and Cu versus Zn (0.380), Cu versus Pb (0.314), Cr versus Zn (0.291) and Cr versus Fe (0.257) show weak relationships presented in Table 7. All the positive relationships among the studied metals in Halda River water express that a potential common source, which can be an anthropogenic source, is mainly responsible for the metal contamination. On the other hand, the negative relationship was also observed in Cr versus Cd (−0.388), Cu versus Cd (−0.150) and Cr versus Cu (−0,031), which means a decrease in Cr and Cu results in an increase of Cd, and more Cd concentrations indicate more contamination.

3.6 | Principal Component Analysis

The PCA was performed to find out the potential sources of studied heavy metals, contributing to the river water metal contamination. Two principal components (PC1 and PC2) with eigenvalues >0.5 were extracted by applying Varimax rotation, which

TABLE 6 | The pollution load index from the collected water samples.

Pollution load index (individual metals)					
Cr	**Cu**	**Cd**	**Fe**	**Zn**	**Pb**
0.080	0.355	0.035	2.258	0.102	6.865

Pollution load index (sampling sites)					
Halda–Karnaphuli confluence	**Chayar Char**	**Khandakia Khal**	**Horekrishno-Mohajoner Tek**	**Napiter Ghona**	**West Binajuri**
0.463	0.291	0.189	0.203	0.351	0.202

TABLE 7 | Pearson's correlation matrix of heavy metals of Halda River water.

	Cr	**Cu**	**Cd**	**Fe**	**Zn**	**Pb**
Cr	1					
Cu	−0.031	1				
Cd	−0.388	−0.150	1			
Fe	0.257	0.472*	−0.055	1		
Zn	0.291	0.380	−0.028	0.967**	1	
Pb	0.082	0.314	0.221	0.793**	0.866**	1

*Correlation is significant at the 0.05 level (2-tailed).
**Correlation is significant at the 0.01 level (2-tailed).

TABLE 8 | Principal component analysis.

	Component	
Element	**PC1**	**PC2**
Cr	0.922	−0.204
Cu	0.936	−0.156
Cd	−0.108	0.957
Fe	0.988	0.059
Zn	0.973	0.099
Pb	0.815	0.346
Eigenvalue	4.323	1.115
% Total variance	72.057	18.576
Cumulative % variance	72.057	90.633

Note: Extraction method: principal component analysis.

explains 90.633% of the total sample variance (Table 8). The first principal component (PC1) accounted for about 72.057% of total variance, which indicates the highest loadings for all the metals except Cd. Because PC1 explains high loadings of Cr, Cu, Fe, Zn and Pb, the sources of these metals might be geological or anthropogenic. Generally, effluents from different industries or mining activities contain Cr compounds. Cr compounds are being used in leather tanning, dyes, chrome plating and wood preservation in different industries for commercial purposes that can be the probable sources of Cr in Halda River water (Barnhart 1997). The presence of a power plant adjacent to the river bank might be the source of Cu in river water, as Cu can be produced from untreated furnace oil released from oil-based power plants (Dey et al. 2021). The sources of Fe can be both geological and anthropogenic. Weathering of rocks and sewage in steel industry might be the reason responsible for Fe in Halda River water (Reimann and Caritat 1998). In addition, Zn and Pb can be introduced into the Halda River water by agricultural runoff. Besides this, the burning of waste materials, coal production and water vehicles and metal plating might be the sources of Zn and Pb respectively (Karrari, Mehrpour, and Abdollahi 2012; Wuana and Okieimen 2011).

The second principal component (PC2) accounted for 18.576% of the total variance with high loading of Cd and Pb. The industries near the Halda River which use Cd in electroplating, battery manufacturing and pigments might be the major sources of Cd into the river water (Rzętała 2016). Moreover, agricultural activities like the use of fertilizers and pesticides can contribute to Cd contamination of river water (Islam et al. 2020). On the contrary, the high loadings of Pb indicate the presence of anthropogenic sources, such as different industrial zones near the riverbank.

4 | Conclusion

The recent status of heavy metal contamination in Halda River water has been assessed in this study, where the concentration of heavy metals follows the order: Fe > Pb > Zn > Cr > Cu > Cd. Metals like Fe cross the permissible limit set by WHO and ECR (1997), and Pb crosses the WHO, USEPA and ECR critical limits. A moderate seasonal variation is seen to have existed among the sampling sites. The seasonal water movements and discharges from various sources impact greatly the concentration of metals. In this study, high heavy metal concentrations were found for the winter and spring, respectively, which means the agricultural activity, the release of untreated effluents from industries situated near the riverbank and the low volume of river water during the dry season (winter) impact greatly the metal concentration of the Halda River water. Due to the accumulation of higher metals in the river water, it can be said that the river water quality is potentially risky for aquatic biota during these seasons. Moreover, different risk assessment indices revealed that the sampling sites are slightly contaminated with Fe and Pb. If we don't take proper initiatives to control the sources of heavy metal contamination, the river water may lose its favourability for spawning of fish soon. Therefore, the establishment of mills, factories, power plants and brickfields in the vicinity of the Halda River should be prohibited immediately, along with further studies essential in order to protect this unique carp breeding area and increase its ecological sustainability.

Author Contributions

Afsana Kamal: Data curation, formal analysis, writing–original draft, investigation, and editing. **Md. Manzoorul Kibria**: supervision, conceptualization. **Shahadat Hossain**: visualization, methodology, resources, project administration. **Chowdhury Kaiser Mahmud**, **Rashmi Roy**, and **Nighat Sultana Resma**: software, validation, writing–review.

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Ethics Statement

We, the authors of this research article, hereby affirm our commitment to upholding the highest standards of ethical conduct throughout the entire research process. We declare that the research conducted and presented in this manuscript complies with established ethical principles, guidelines, and regulations.

Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data for this study is available on request from the corresponding author.

Peer Review

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