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Heavy Metals in Halda River Sediments: Index-Based Evaluation and Multivariate Interpretation

Md. Abdul Aziz[1]*, Md. Rashid Awal Chowdhury[2], A. Ghosh[3], Sudip Kumar Pal[4], Akramul Haque[5], Md. Shahriar Hossain[6] and Jushan Abdullah[7]

[1]Department of Civil Engineering, CUET, Bangladesh (yasinmdabdulaziz@gmail.com)
[2]Department of Civil Engineering, PCIU, Bangladesh (rashidacm07@gmail.com)
[3]Department of Civil and Environmental Engineering, South Dakota State University, USA, (avishekg90@gmail.com)
[4]Department of Civil Engineering, CUET, Bangladesh (sudip@cuet.ac.bd)
[5]Department of Civil Engineering, DUET, Bangladesh (akramul@duet.ac.bd)
[6]Department of Civil Engineering, PCIU, Bangladesh (shahriarhossaince927@gmail.com)
[7]Department of Civil Engineering, CUET, Bangladesh (jushan.abdullah@yahoo.com)

Abstract

Heavy metal contamination in river sediments poses significant ecological and health risks, especially in rapidly urbanizing regions. This study assessed the distribution, contamination levels, and ecological risks of Cu, Fe, Mn, Cr, and Zn in sediments from 10 sites along the Halda River, Bangladesh spanning upstream (S1–S3), midstream (S4–S6), and downstream (S7–S10) sections. Sediment samples were collected using a Van Veen grab, oven-dried, sieved (<0.075 mm), digested with aqua regia, and analyzed via spectrophotometer. Contamination was evaluated using Contamination Factor (CF), Pollution Load Index (PLI), Geo-accumulation Index (I_{geo}), Enrichment Factor (EF) and Ecological Risk Index (RI). Results showed Cu, Fe, and Mn as the primary pollutants, with peak concentrations at S5 (Cu: 25.701 mg/kg; Fe: 9443.55 mg/kg) and S7 (Mn: 1963.9 mg/kg). PLI values exceeded 1 (highest contamination level) at all sites, reaching 22.90 at S5, indicating severe pollution. I_{geo} values classified Cu and Fe at S3, S5, and S8 as “heavily to extremely polluted,” while EF values (<1.0) suggested mostly natural origins. RI indicated high ecological risk at S3, S5, and S8, predominantly from Cu. Correlation and PCA revealed Cu, Fe, Cr, and Zn shared common sources, while Mn originated separately. The findings highlight urgent management needs to preserve the Halda River’s ecological integrity.
Keywords: Halda River, heavy metals, sediment pollution, risk assessment, PCA

1. Introduction

The increment of heavy metal contamination in aquatic environments has become a significant concern in recent decades. (Fernandes et al., 2008; Bhuyan et al., 2019) due to the indestructibility and the toxicity of their harmful effects (Islam et al., 2015; Wu et al., 2016). Heavy metals can reach aquatic ecosystems through point sources such industrial, municipal, and home wastewater effluents, as well as through surface runoff, erosion, and atmospheric deposition.(Akcay et al., 2003; Bhuyan et al., 2019). The impacts of unplanned urbanization and industrialization are contributing to an increase in metal contamination in aquatic habitats (Nilin et al., 2013; Ali et al., 2016). This becomes the main quality concern in fast developing countries since population and urbanization growth is more rapid than water and sediment quality maintenance systems (Bhuyan et al., 2019).

Sediments have been a vital, important, and dynamic element of the river basin with its diverse range of habitats and environments, often providing critical information for environmental and geochemical pollution levels. Suspended particle matter and sediments in aquatic settings account for more than 90% of the total heavy metal load.(Varol & Şen, 2012). Sediments have long been regarded as environmental sinks for the assessment of metal pollution in aquatic ecosystems. (Islam et al., 2015).

The Halda River is the sole natural breeding place for major Indian carps, located in the southeastern part of Bangladesh, establishing it as a distinctive heritage of this country. This river water is not only a natural breeding place for big carps, but it is also used a lot in farming, irrigation, raising cattle, fish farming, bathing, recreation, and drinking. But in the last few years, the Halda has been under more and more anthropogenic pressures like industrial waste, city sewage, and agricultural runoff. As a result, large volumes of toxic and hazardous heavy metals are discharged in the river (Bhuyan & Bakar, 2017; Dey et al., 2021).

Numerous indices have been developed to evaluate the risk that heavy metals pose to the environment. Among the developed indices contamination factor (CF), pollution load index (PLI), enrichment factor (EF), geo-accumulation index (I_{geo}), and risk index (RI) were calculated for individual heavy metal in sediment. The objective of this study is to assess the magnitude of heavy metal pollution by cataloging the concentrations and their distribution within a riverine environment. Also, to assess spatial variation of heavy metals with respect to water flows in different streams by using ArcGIS and explore the relationship between the metals by statistical analysis.

2. Materials and methods

2.1 Study location

The study was conducted on 16.44 km of Halda River, geographically located between 22° 25′ 13″–22° 48′ 51.37″ N and 91° 45′ 00″–91° 52′ 33″ E. Originating from the hill ranges of Chattogram hill tracts, it flows through the Chattogram district before merging with the Karnaphuli River. During Oct- Dec, 2024, the data were collected. Between the selected areas, there are many heavy industries and industrial and sewage wastes are carried by a large number of canals in to the river. Human activities (e.g. farming, harvesting, and bathing) are conducted near the riverbank. These are contributing in increased amount pollution in sediment and aquatic environment, so for the selection of sampling, these factors are considered.

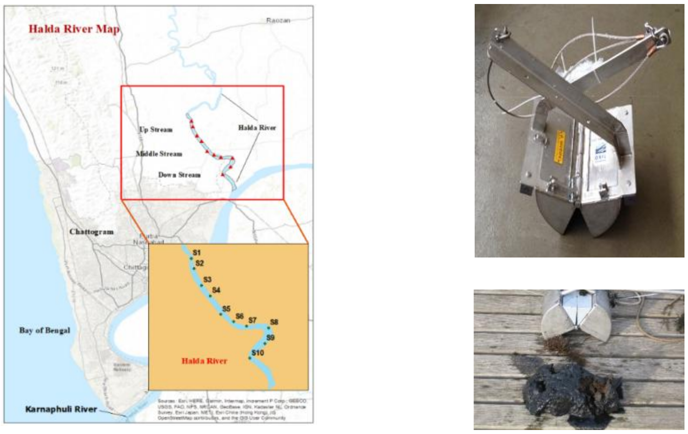


Figure 1. Sampling points in Halda River (left); Instrument: Van Veen grab used to collect sediment from river bed (right).

2.2. Sample collection

Three sample locations were located in the Halda River's upstream (S1–S3), three in the middle stream (S4–S6), and four in the downstream (S7–S10) portions. (Figure 1: left). The 'Van Veen grab' machine (Company: osil, country: UK) was used to collect sediment samples from river bed by submerging the sampler to the bottom of the water and to prevent contamination, the obtained samples were kept apart in airtight bags. (Figure 1: right).

2.3. Sample preparation

The sample materials were oven dried at 110 °C for 24 hours and air-dried, homogenized, crushed for sieving. After that, all of the standard sieve sizes were used to filter the samples and finally sample passing through #200 no sieve (0.075 mm) was collected. The collected samples were then weighed and all information were recorded. Finally, the retained mass, weight of % retained, cumulative % retained, % finer and FM were calculated. Aqua regia was then used to digest the sediment samples. ($HCl: HNO_3 = 3:1$) on a hotplate at 90–95 °C for 1 h, filtered, and diluted with deionized water to volume. The digests were analyzed for Cu, Fe, Mn, Cr, and Zn using a spectrophotometer (Model HI802-02, HANNA Instruments) following calibration with appropriate standards.

2.4. Index Analysis

2.4.1 Contamination factor (CF)

In terms of contamination factor (CF), the degree of metal contamination in sediment is represented as follows:

$$CF = \frac{C^i}{C_n^i} \tag{1}$$

Where CF is the single element pollution factor, C^i is the content of the element in samples and C_n^i is the reference value of the element. Contamination factor (CF) is classified as Class 1: CF < 1 (low), Class 2: 1 ≤ CF < 3 (moderate), Class 3: 3 ≤ CF < 6 (considerable), and Class 4: CF ≥ 6 (very high) contamination (Ali et al., 2016).

2.4.2 Pollution load index (PLI)

The PLI is defined as the nth root of the multi plications of the contamination factor (CF) of metals (Ali et al., 2016).

$$PLI = (CF_1 \times CF_2 \times CF_3 \times ......\times CF_n)^{1/n} \tag{2}$$

Here, A PLI value of zero signifies perfection, a value of one denotes the existence of merely baseline pollution levels, while values over one indicates a continual deterioration of the site and estuary quality.

2.4.3 Enrichment factor (EF)

The enrichment factor (EF) is considered to be a useful tool for assessing the level of pollutants in the environment. EF is represented as follows

$$EF = (C_M / C_{Fe})_{\text{sample}} / (C_M / C_{Fe})_{\text{Earth's crust}} \tag{3}$$

Where, Where, $(C_M / C_{Fe})_{\text{sample}}$ is the ratio of concentration of heavy metal (C_M) to that of iron (C_{Fe}) in the sediment sample, and $(C_M/C_{Fe})_{\text{Earth's crust}}$ is the same reference ratio in the Earth's crust (Mohiuddin et al., 2016).

2.4.4 Geo-accumulation index (I_{geo})

The geoaccumulation index (I_{geo}) values were computed using the following formula to describe the degree of pollution in the sediment:

$$I_{geo} = \text{Log}2[C_n/1.5B_n] \tag{3}$$

where C_n is the measured concentration of metal n in the sediment and B_n is the geochemical background value of element n in the background sample. The factor 1.5 is introduced to minimize the possible variations in the background values which may be qualified to lithogenic effect. Geoaccumulation index (I_{geo}) values were interpreted as: $I_{geo} \leq 0$—practically uncontaminated; $0 \leq I_{geo} \leq 1$—uncontaminated to moderately contaminated; $1 \leq I_{geo} \leq 2$—moderately contaminated; $2 \leq I_{geo} \leq 3$—moderately to heavily contaminated; $3 \leq I_{geo} \leq 4$—heavily contaminated; $4 \leq I_{geo} \leq 5$—heavily to extremely contaminated; and $5 < I_{geo}$ —extremely contaminated (Nilin et al., 2013).

2.4.5 Ecological risk index (RI)

The Risk Index (RI), as suggested by Hakanson (1980), is used to evaluate the level of heavy metal pollution. The toxic response factors are Cu = 5, Pb = 5, Zn = 1, Cd = 30, Ni = 6, Cr = 2, As = 10, and Hg = 40. In this study, a modified RI classification was applied in order to evaluate the RI by the analyzed six heavy metals Table. Potential ecological risk (RI) is graded as: RI < 50 (low risk), 50 ≤ RI < 100 (moderate risk), 100 ≤ RI < 200 (considerable risk), and RI ≥ 200 (high risk) (Ali et al., 2016).

3. Result and Discussion:

3.1 Size distribution of sediment samples

The gradation curve (Figure 2: left) shows the particle size distribution of 10 sediment samples (S1–S10) from the Halda River. Most samples exhibit well-graded behavior with smooth and continuous curves, indicating a wide range of particle sizes, while sample S2 stands out with a coarser and poorly graded profile, having a higher percentage of coarse particles above 0.3 mm. The rest of the samples display finer sediments, with over 80% finer than 1 mm, suggesting dominance of silts and fine sands typical of riverine depositional environments.

3.2 Heavy metal Concentration

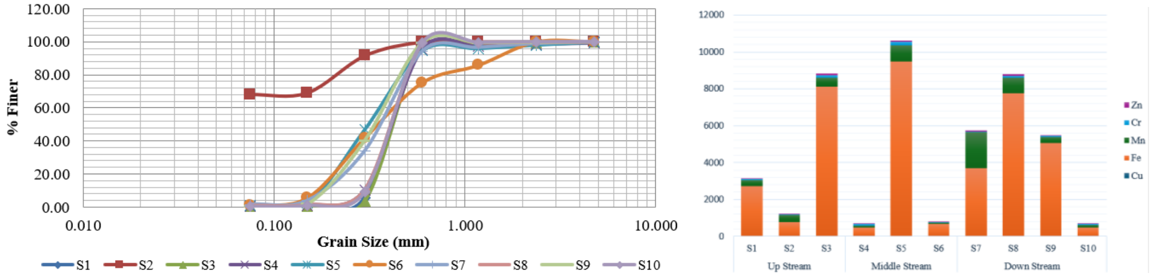
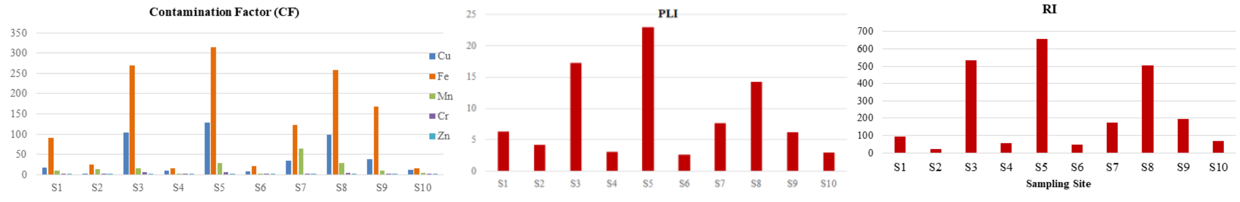


Figure 2. Gradation curve for sediments for all sampling sites (S1-S10), Heavy metal Concentration (mg/kg units)

The concentration of heavy metals across the Halda River shows spatial variation from upstream to downstream (Figure 2: right). In the upstream, Fe peaked at 8096.2 mg/kg (S3) and Cu at 20.836 mg/kg (S3), while Mn and Cr were 476.25 mg/kg and 148.83 mg/kg, respectively. In the middle stream, Fe and Cu reached maximums of 9443.55 mg/kg and 25.701 mg/kg at S5, with Mn at 896.54 mg/kg, Cr at 179.31 mg/kg, and Zn at 83.68 mg/kg. Downstream sites showed elevated Mn at 1963.9 mg/kg (S7) and Fe at 7730.5 mg/kg (S8), with Zn peaking at 85.11 mg/kg (S8), indicating diverse metal inputs likely from anthropogenic sources. Heavy metals As, Cr, Cd, and Pb in Karnaphuli River, Bangladesh, showed concentrations in water (Cr > As > Pb > Cd) and sediment (Cr > Pb > As > Cd) with water levels exceeding safe drinking limits (Ali et al., 2016). The samples of sediments from Buriganga River showed high contamination with mean concentrations of Cr (173.4 µg/g), Pb (31.4 µg/g), Cd (1.5 µg/g), Ni (153.3 µg/g), Fe (481.8 µg/g), Cu (344.2 µg/g), Zn (12,989 µg/g), and Mn (4,036 µg/g), with Cr, Cu, and Ni exceeding severe effect levels (Mohiuddin et al., 2016). Heavy metals Cr, Ni, Cu, As, Cd, and Pb in sediments of Paira River, Bangladesh, averaged 45, 34, 30, 12, 0.72, and 25 mg/kg respectively, with higher concentrations in winter (Islam et al., 2015).

3.3 Index Analysis

3.1.1. Contamination factor (CF) & Pollution load index (PLI|)



(a) (b) (c)

Figure 3. Contamination factor (CF) (a); pollution load index (b) and Risk Index (c) for sediment in Halda River, Chattogram

The Contamination Factor (CF) graph (Figure 3a) shows extremely high Cu and Fe levels across all sites, especially at S3, S5, and S8, indicating severe contamination. Mn contamination peaks at S7 and S5, while Cr shows moderate to considerable levels at S3 and S5, but Zn CF remains low at all sites. Overall, midstream (S3–S6) and downstream (S7–S10) have higher metal pollution than upstream (S1–S2), likely from cumulative anthropogenic sources. Sediments contamination factors and pollution load index (>1) indicating moderate to high sediment pollution, posing ecological and health risks in Karnaphuli River (Ali et al., 2016).

All ten sites show PLI values greater than 1, indicating varying degrees of heavy metal contamination. S5 shows the highest pollution load (PLI = 22.90), driven by extremely high Cu, Fe, and Mn levels. S3, S8, and S7 also exhibit severe contamination, with PLI values well above 10. S4, S6, and S10 show the lowest PLI values (around 2–3) but still exceed baseline levels. Zn levels remain low across all sites, but Cu, Fe, and Mn are the main contributors to pollution (Figure 3c).

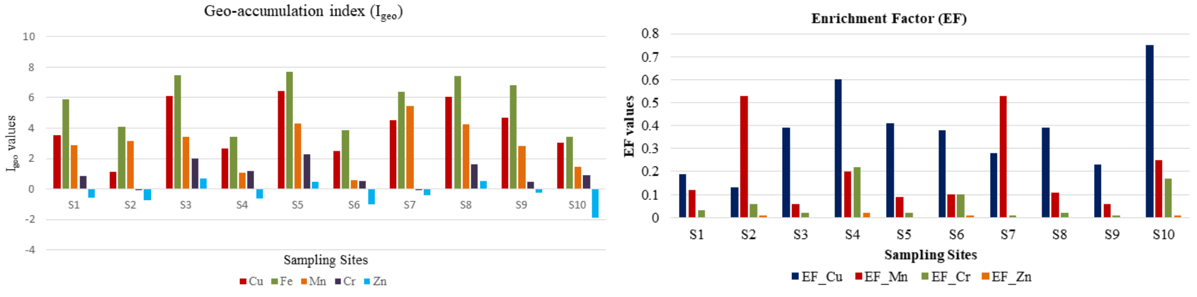


Figure 4. Geo-accumulation index (I_{geo}) (left) and Enrichment factor for sediment samples (right)

2.2 Geo-accumulation index I_{geo}

Cu, Fe, and Mn show high contamination, with several samples falling into the "heavily" to "extremely polluted" categories especially Cu (S3, S5, S8) and Fe (S3, S5, S8). Cr levels are mostly in the "moderately polluted" range (S3, S4, S5, S8), with some locations being unpolluted to slightly polluted. Zn is the least polluted, with most I_{geo} values below 0, indicating it is generally unpolluted across the sampling sites. S5 and S8 are the most contaminated sites overall, with high I_{geo} values for multiple metals (Figure 4 left). According to I_{geo} classification, the majority of sediments are affected by anthropogenic inputs, particularly with Fe, Cu, and Mn, requiring urgent attention. For Al, Cu, Cr, and Zn, I_{geo} results showed moderate to severe contamination, particularly at sites in the inner Ceará River estuary in Brazil. (Nilin et al., 2013).

2.3 Enrichment Factor (EF)

Enrichment Factor (EF) values for Cu, Mn, Cr, and Zn at all sites (S1–S10) along the Halda River are below 1.0, indicating no significant contamination and mainly natural sources. The highest EF was for Cu at S10 (0.75), well under the anthropogenic threshold, with no metals exceeding EF > 1.5, confirming minimal human impact (Figure 4 right). The Ceará River estuary in Brazil showed sediment metal contamination following Fe > Al > Zn > Cr > Pb > Cu, with moderate to strong enrichment of Al, Cu, Cr, and Zn in inner sites (Nilin et al., 2013). Sediment samples from Buriganga River exceeding toxicity reference values; enrichment factors and pollution indices indicated extreme pollution, posing serious risks to the ecosystem and local population (Mohiuddin et al., 2016).

2.4 Risk Index (RI)

Ecological risk assessment shows high risk at S3, S5, and S8 (RI > 200) due to elevated Cu levels (up to 25.70 mg/kg), considerable risk at S7 and S9 (100 ≤ RI < 200), and moderate risk at S1, S4, and S10 (50 ≤ RI < 100). S2 and S6 are low risk (RI < 50), with Cu as the dominant contributor, and midstream/downstream areas having higher risk than upstream sites. Pasur River water, pollution load and contamination factors indicating progressive sediment deterioration and highest ecological risk from Cd, urging urgent treatment measures (Ali et al., 2018).

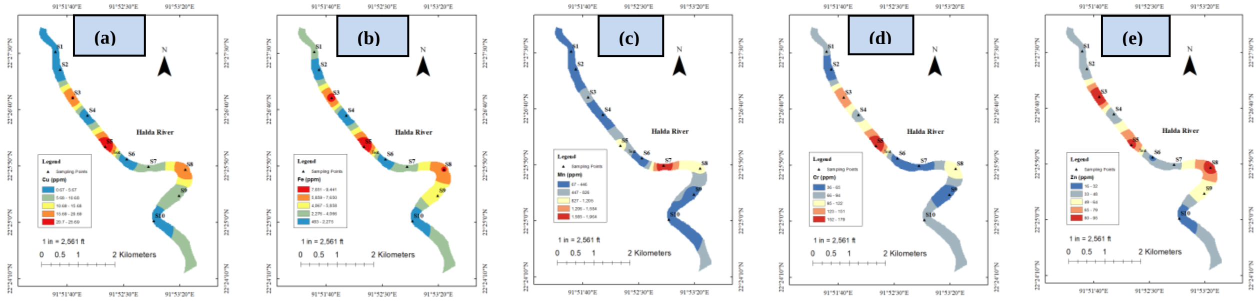


Figure 5. Spatial maps of concentration across Halda River, (a) Cu, (b) Fe, (c) Mn, (d) Cr, and (e) Zn

3.6 Spatial Analysis

The IDW maps (Figure 5. (a-e)) show Cu ranging from 0.67 mg/kg (S2) to 25.70 mg/kg (S5), with high spots at S3, S5, and S8. Fe peaked at 9443.55 mg/kg (S5) and 8096.2 mg/kg (S3), while Mn reached 1963.9 mg/kg at S7, indicating downstream enrichment. Cr peaked at 179.31 mg/kg (S5) and Zn at 95.25 mg/kg (S3), with the lowest Zn at 16.33 mg/kg (S10).

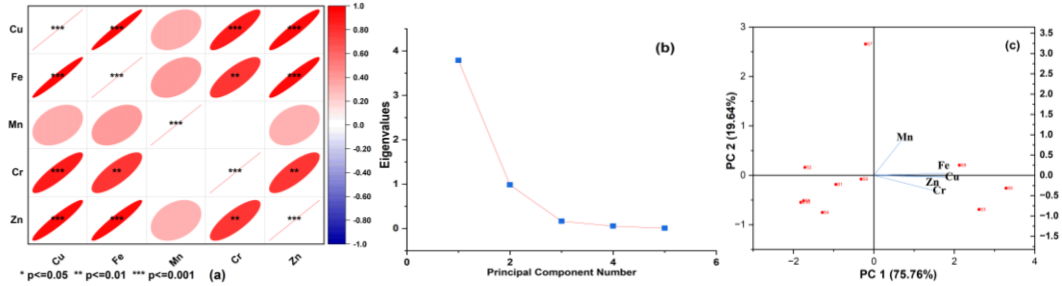


Figure 6. Pearson Correlation (a), PCA (biplot (b), Scree plot (c) between the metals

3.6 Pearson Correlation Analysis

Pearson correlation analysis shows strong positive links among Cu, Fe, Cr, and Zn, especially Cu–Fe (r = 0.97), Zn–Cu (r = 0.94), and Zn–Fe (r = 0.95) indicating common sources, with Cr also strongly tied to Cu (r = 0.88) and Zn (r = 0.80), while Mn showed weak correlations, suggesting a separate origin. (Figure 6a).

3.7 Principal Component Analysis (PCA)

Principal Component Analysis (PCA) of the five heavy metals (Cu, Fe, Mn, Cr, Zn) in sediment samples from 10 sites (S1–S10) reveals that the first two principal components (PC1 and PC2) explain 95.4% of the total variance, with PC1 alone accounting for 75.76%. PC1 is strongly influenced by Cu (0.51), Fe (0.50), Cr (0.45), and Zn (0.50), indicating these metals have a common source or similar behavior. PC2 is primarily dominated by Mn (0.91), suggesting a distinct input or geochemical behavior. The biplot shows clear grouping of metal influences, while the scree plot confirms the dominance of the first component in explaining variability (Figure 6b, 6c).

Conclusion

The Halda River's sediment analysis shows that the majority of the material is fine-grained, with well-graded textures except for S2, which is coarse and poorly graded. Heavy metal concentrations show clear spatial variation, with Cu, Fe, and Mn being the primary contaminants, particularly in midstream and downstream sites (S3, S5, S7, S8). Pollution indices (CF, PLI, I_{geo}) reveal moderate to extreme contamination, especially for Cu and Fe, while EF values suggest most metals are of natural origin. Ecological risk assessment identifies S3, S5, and S8 as high-risk sites, with Cu as the dominant contributor to ecological threats. Multivariate analysis (correlation and PCA) further supports that Cu, Fe, Cr, and Zn share common sources, while Mn originates from distinct inputs, emphasizing how human activities and natural processes both affect the quality of sediments.

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