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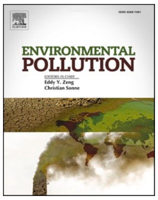

Heavy metal pollution in surface water bodies in provincial Khanh Hoa, Vietnam: Pollution and human health risk assessment, source quantification, and implications for sustainable management and development[☆]

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ABSTRACT

The global issue of heavy metal pollution in surface water poses a significant concern, with the potential to harm public health through various pathways. Given that pollution levels are dependent on water bodies and seasons and their potential impacts on human health vary with children and adults, it is crucial to identify and quantify pollution sources for the development of sustainable management strategies. The current study aimed to evaluate pollution levels and associated health risks of heavy metals and to quantify their pollution sources in various surface water bodies in Khanh Hoa, Vietnam. Water samples were taken from three water bodies (reservoirs, rivers, and narrow waterways) during two seasons (dry and rainy) from 2016 to 2020 and analyzed for seven heavy metals. The results showed that iron had the highest concentration of 392.4 ($\mu g\ L^{-1}$), followed by zinc (25.7 $\mu g\ L^{-1}$), arsenic (3.93 $\mu g\ L^{-1}$), copper (3.77 $\mu g\ L^{-1}$), lead (2.77 $\mu g\ L^{-1}$), chromium (2.71 $\mu g\ L^{-1}$), and cadmium (0.57 $\mu g\ L^{-1}$). Narrow waterways were more polluted with heavy metals (heavy metal pollution index, HPI = 29.5) than other water bodies, such as rivers (23.3) and reservoirs (21.7), and the dry season had a higher HPI (26.5) than the rainy season (24.0). The hazard index for children varied from 1.2 to 1.48, while that for adults was less than 1, suggesting that surface water may have adverse impacts on children's health. The factor analysis identified three primary sources of contamination, namely combustion emissions/street dust, agricultural run-off, and other sources. Cadmium is the most critical metal in determining HPI, while arsenic and chromium are the two key elements potentially influencing children's health. Managing pollution sources, reducing the metal concentration, and controlling the pathways through which metals enter the human body should be implemented for a healthier environment and long-term development.

1. Introduction

Heavy metals are dense metals and metalloids with atomic densities greater than 4000 kg m^{-3} (Vardhan et al., 2019). They pose a significant global concern as their presence in surface water can be toxic to organisms and humans even at low concentrations. These metals have characteristics such as long-term persistence in the environment, bio-accumulation in the food chain, and bio-toxicity, necessitating the control of their levels below acceptable thresholds. Nevertheless, the concentration of heavy metals in surface water resources worldwide has been increasing in recent decades (Zhou et al., 2020). Heavy metals can enter surface water bodies through various sources, including mining, the discharge of metal-containing wastes, effluent from industrial areas, leaded gasoline and paints, fossil fuel combustion, agricultural fertilizers and pesticides, animal manures, sewage sludge, wastewater from residential areas (Anubhav et al., 2022; Briffa et al., 2020; Sonone et al., 2020). Both natural processes and human activities contribute to heavy metal contamination, emphasizing the need for further research and effective management strategies.

Many studies have been conducted to examine heavy metal pollution in surface water, focusing on metal concentrations in comparison to maximum permeable thresholds, such as the WHO's guideline (WHO,

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2011), and identify the pollution sources (Astatkie et al., 2021; Zhou et al., 2020). A study conducted by Kumar et al. (2020) showed that heavy metal concentrations varied greatly between water bodies. The finding indicates that heavy metal pollution may differ depending on the type of water bodies, including canals, streams, rivers, and reservoirs. Each water body possesses distinct features influencing metal presence. The reservoirs may store large volumes of freshwater for daily consumption and irrigation, while rivers can contain freshwater, which can flow from one place to the others and may connect the reservoirs to the sea. Canals and streams, referred to as narrow waterways, act as channels for pollutant transport, including metals, from their sources to rivers or reservoirs. The great variation in the water speed, storage capacity, and spatial position may contribute to the significant differences in heavy metal pollution among these water bodies. Nevertheless, limited research has explored the effects of different water bodies on heavy metal levels in surface water. Thus our first hypothesis is that narrow waterways are more polluted with heavy metals than the other types of water bodies.

Studies have examined the influence of natural factors, such as seasonal variations, on heavy metal pollution in surface water, yielding mixed results. In the Ganges River of northwestern Bangladesh, the heavy metal pollution index (HPI) was higher during the summer than during the monsoon season (Haque et al., 2019). The presence of certain metals was greater during summer, while others exhibited higher levels during the rainy season, likely due to varying pollution sources (Banerjee et al., 2016). The pollution levels of metals in water samples were higher in the pre-monsoon season compared to the monsoon and post-monsoon seasons (Kumar et al., 2013). The seasonal variation in heavy metal concentration in surface water can be involved in water flow, transporting metals from their origins to temporary storage places of surface water bodies. During the rainy season, increased water may carry more metals, leading to a higher metal concentration in water bodies. Conservely, increased water flow during the rainy season can dilute metal concentration. During the dry season, water scarcity due to evaporation may elevate metal concentrations as water loss of water condenses metals in surface water bodies. These findings lead to the development of a second hypothesis that water samples collected during the dry season would show a higher HPI than those collected during the rainy season.

Surface water heavy metal pollution poses a significant risk to human health, as these toxic metals can accumulate in living organisms over time (Mitra et al., 2022). Heavy metals in surface water can enter the human body through various pathways, including drinking, skin absorption, and ingestion of contaminated products (Mawari et al., 2022; Witkowska et al., 2021). These pathways of individual metals are taken into account to calculate the hazard quotient and hazard index, being used to evaluate the potential risks to human health (EPA, 2004; Selvam et al., 2022). The potential impacts on public health vary with specific metals and the development stage of individuals. Compared to adults, children may exhibit heightened sensitivity to heavy metal pollution levels in surface water, due to factors such as lower body weight, educational level, and weaker immune function. Consequently, our third research hypothesis is that children are more vulnerable to heavy metal pollution than adults. Therefore, this study aimed to assess heavy metal pollution levels, associated health risks, and pollution sources in different surface water bodies in Khanh Hoa province, Vietnam. Furthermore, the current study was designed to capture the effects of seasons on heavy metal concentration in surface water.

2. Materials and methods

2.1. Study area

The current study was conducted in Khanh Hoa province, located in the South Central Coast region of Vietnam at 11°48′22″ to 12°52′39″ N and 108°40′8″ to 109°27′50″ E (Supplementary Fig. 1). It covers 5197 km^2 and has a population of around 1232 823 people (Khanh Hoa statistics offices, 2020). Khanh Hoa is situated near the Truong Son mountain range, with mountains covering the majority of its land area. The terrain of the province is relatively high, with an average elevation of around 60 m above sea level. Khanh Hoa has a dense network of water systems with short and steep rivers that finally flow into the eastern sea of Vietnam, potentially contaminating the international seas if they carry out elevated levels of heavy metals. Khanh Hoa has a tropical savanna climate, with two distinct dry and rainy seasons. The rainy season is short, lasting from September to December, contributing 70–80% of the total annual rainfall, while the remaining months constitute the sunny season. The province has an average annual temperature of around 26.7 °C, a relative humidity of approximately 80.5%, and an annual total rainfall of about 2000 mm.

Khanh Hoa is undergoing rapid economic growth with tourism services contributing 45%, industry and construction making up 42%, and agriculture, forestry, and aquaculture accounting for 13%. The province has strong traditional industries like shipbuilding, seafood processing, construction materials, and garments. With 72 ore mines registered by 2003, Khanh Hoa is also rich in minerals. Despite having a small and narrow plain system, farming, particularly rice cultivation, remains a significant economic activity, alongside short-term industrial crops like sugarcane and peanuts, and food crops such as cassava and corn. These productive activities generate waste, posing a threat to the local surface water system.

2.2. Experimental factors

Khanh Hoa province has a diverse surface water system, which contains four types of water bodies, including rivers, reservoirs, streams, and canals. While rivers and reservoirs have distinct characteristics that set them apart, the latter two share common features with narrow widths (less than 5–7 m). The current study distinguishes three types of water bodies, including reservoirs, rivers, and narrow waterways (streams and canals), forming an experimental factor potentially influencing heavy metal status. In addition, the study area has two distinct seasons. The rainy season lasts from September to December, contributing roughly 70–80% of the total annual rainfall and the remaining months are the dry season. These seasons are considered additional experimental factors expected to influence heavy metal concentrations in the three water body types.

2.3. Water sampling and analysis

Surface water samples were taken for the current study, based on the two experimental factors, which are water body type and season. The sampling was implemented for five consecutive years from 2016 to 2020, and 12 months a year. Nineteen sampling sites (seven reservoirs, eight rivers, and four narrow waterways) were pre-selected to take surface water samples repeatedly over five years (Supplementary Fig. 1 and Supplementary Table 1). The distribution of sampling sites across water bodies was determined by local conditions and the proportional representation of each water body in the study area. For one sampling campaign, a GPS (Global Positioning System, **GPS Garmin MONTANA 680**) device was used to locate the pre-selected sites based on their *latitude* and *longitude* GPS coordinates. A total of 1140 surface water samples (19 sampling sites per month x 12 months per year x 5 years) were taken for the study.

The Van Dorn water sampler was employed to collect surface water samples from the 0–50 cm layer, maintaining a relatively consistent sampling depth across all 1140 samples. For rivers and narrow waterways, each surface water sample was composed of eight samplers distributed across the cross-section of these water bodies. For the reservoirs, eight to ten samplers distributed over the surface of the water bodies and a few hundred meters away from their shores were implemented. Collected water samples were securely capped in plastic bottles, placed in an ice box at 4 °C, and transported to a laboratory. Chemical analyses of seven metals, including iron (Fe), zinc (Zn), arsenic (As), copper (Cu), lead (Pb), chromium (Cr), and cadmium (Cd) were implemented based on the procedure by Giri and Singh (2013). The samples were first prepared by filtering them through Whatman No. 42 filter paper into centrifuge tubes. The filtrate was then acidified to a pH of 2 using concentrated nitric acid before being stored in a refrigerator at 4 °C until analysis. The concentrations of the seven heavy metals were measured using Inductively coupled plasma-optical emission spectrometry (ICP-OES, Spectroblue ICP-OES Analyzer, Spectro Analytical Instruments GmbH & Co. KG) following Nguyen et al. (2020). The analysis commenced with six standard solutions featuring known metal concentrations to construct a standard curve with a coefficient of determination exceeding 0.98. To validate the accuracy of the analysis, a calibration blank sample and a standard solution of known metal concentration were reanalyzed after every 15 samples.

2.4. *Calculation and statistical analysis*

One important objective of this study was to identify sources of the harmful elements and quantify their contributions, utilizing multivariate regression analyses such as principal component analysis/factor analysis and multiple linear analysis (Le et al., 2023; Shihab, 2022; Tokatlı et al., 2023b) were employed. Additionally, the potential impacts of pollutants on human health were further assessed for early warning purposes, utilizing various risk indexes, such as the heavy metal pollution index (HPI), hazard quotient (HQ), and hazard index (HI) (Haq et al., 2023). The HPI gauges overall water quality concerning metal pollution, the HQ evaluates the risk associated with individual pollutants, and the HI provides a cumulative assessment of the potential risk from exposure to multiple pollutants (Nguyen et al., 2023; Tokatlı et al., 2023a; Tokatli and Ustaoğlu, 2020). These ecotoxicological indexes and statistical methods have been found widespread application in studies worldwide to assess heavy metal pollution and its potential impacts on living organisms (Haq et al., 2023; Le et al., 2023; Tokatlı et al., 2023a; Tokatlı et al., 2023b).

2.4.1. *Heavy metal pollution index (HPI)*

The HPI was computed using the method by (Uugwanga and Kgabi, 2021), as shown in the following equation (Eq. 1) below.

$$\mathrm{HPI} = \frac{\sum_{i=1}^{n} Q_i W_i}{\sum_{i=1}^{n} W_i} \tag{1}$$

Where n is the number of heavy metals examined; W_i is equal to $\frac{1}{S_i}$; Si is the water quality standard of the ith metal (Supplementary Table 2); $Q_i = \frac{V_i}{S_i}x100$; V_i is the measured concentration of the ith metal. A value of HPI less than 100 indicates that the water is at low risk of heavy metal pollution; otherwise, the water is at high risk.

2.4.2. *Hazard index (HI)*

The health risk assessment was implemented using the method developed by EPA (2001) and (Xiao et al., 2019) to compute the HI for children and adults. Details for the calculation of these indexes were shown in Supplementary Text 1.

2.4.3. *Other statistical methods*

Principal component analysis/factor analysis (PCA/FA) was applied to the whole dataset of the concentrations of the seven heavy metals in 1140 water samples, following the procedure described by (Le et al., 2023; Shihab, 2022). Multiple linear analysis was employed to calculate the percentage contribution of individual heavy metals to the explanation of the HPI and HIs for children and adults (Nguyen et al., 2020; Putri et al., 2018). The inter-relationship among the seven metals and three indexes (HPI and HIs for children and adults) was examined using a Pearson correlation matrix to aid in identifying pollution sources. Analysis of variance was performed on all data, including metal concentrations, HPI, HIs, and HQs, using a two-factor (water body and season) completely randomized design with varying replicates (Ott and Longnecker, 2016). When the ANOVA result indicated a significant effect at $P \leq 0.05$, Tukey's Honest Significant Difference test was employed to classify treatment means.

3. Results

3.1. *Heavy metal status*

The heavy metal pollution index (HPI), computed for the seven metals analyzed in the current study, was shown in Fig. 1. Over the five-year period from 2016 to 2020, HPI varied from 9.2 to 59.3 with an average of 24.3. The highest HPI occurred in 2016, while 2020 exhibited the lowest (Fig. 1a), indicating that HPI decreased from 2016 to 2020. HPI was significantly affected by both seasons and water bodies separately (Fig. 1b). Among the three water bodies, reservoirs had the lowest HPI (21.7), while narrow waterways exhibited the highest (29.5). The dry season had a higher HPI (25.7) than the rainy season (24.02).

The concentration of Pb, Cu, Zn, and As was not significantly influenced by the interaction between the water body and reason (Fig. 2). The Pb concentration was significantly affected by the water body but not by the season (Fig. 2a). Reservoirs had the lowest Pb concentration (2.3 μg L^{-1}), while narrow waterways had the highest concentration (4.21 μg L^{-1}). In narrow waterways, surface water samples collected during the rainy season had a higher Cu concentration than those collected during the dry season. Conversely, in reservoirs and rivers, water samples collected during the two seasons had a similar Cu concentration. Dry season samples from narrow waterways had the highest Zn concentration (36.3 μg L^{-1}) and reservoir samples during the same season exhibited the lowest (20.3 μg L^{-1}). Reservoirs had the lowest Zn concentration (21.4 μg L^{-1}), while narrow waterways showed the highest (37.6 μg L^{-1}). The As concentration was highest in narrow waterways during the rainy season (4.7 μg L^{-1}) and lowest in reservoirs during the dry season (3.7 μg L^{-1}). Narrow waterways had the highest As concentration, while the other water bodies showed similar As concentrations. In addition, surface water had pH values varying from 6.0 to 8.8 and dissolved oxygen (DO) levels ranging from 0.8 to 7.2 (mg L^{-1}) and reservoirs exhibited higher pH and DO levels than narrow waterways.

The interactive effect between the water body and the season on the concentration of Cd, Fe, and Cr was also not significant (Fig. 3). In narrow waterways, surface water samples taken during the dry season had a higher Cd concentration (0.70 μg L^{-1}) than those taken during the rainy season (0.58 μg L^{-1}). Meanwhile, in rivers and reservoirs, the Cd concentration was similar in both seasons. The highest Fe concentration occurred in the rivers during the rainy season was the highest (574.5 μg L^{-1}), while the lowest was in reservoirs during the dry season was the lowest (270.9 μg L^{-1}) (Fig. 3b). The Cr concentrations in surface water samples in three water bodies and during the two seasons were not significantly different from each other (Fig. 3c).

3.2. *Human health risk*

The human health risk was assessed through the hazard index (HI) and hazard quotient (HQ), presented in Fig. 4 and Table 1, respectively. The HI for both children and adults was significantly influenced by the water body but not by the season or their interaction (Fig. 4). Narrow waterways had the highest HI for children (1.4) and adults (0.67), while reservoirs and rivers showed similar HIs, varying from 1.20 to 1.27 for children and from 0.55 to 0.59 for adults. For both children and adults, the HQ of all seven metals was not significantly affected by the interaction between the water body and the season (Table 1). For Fe, the HQ

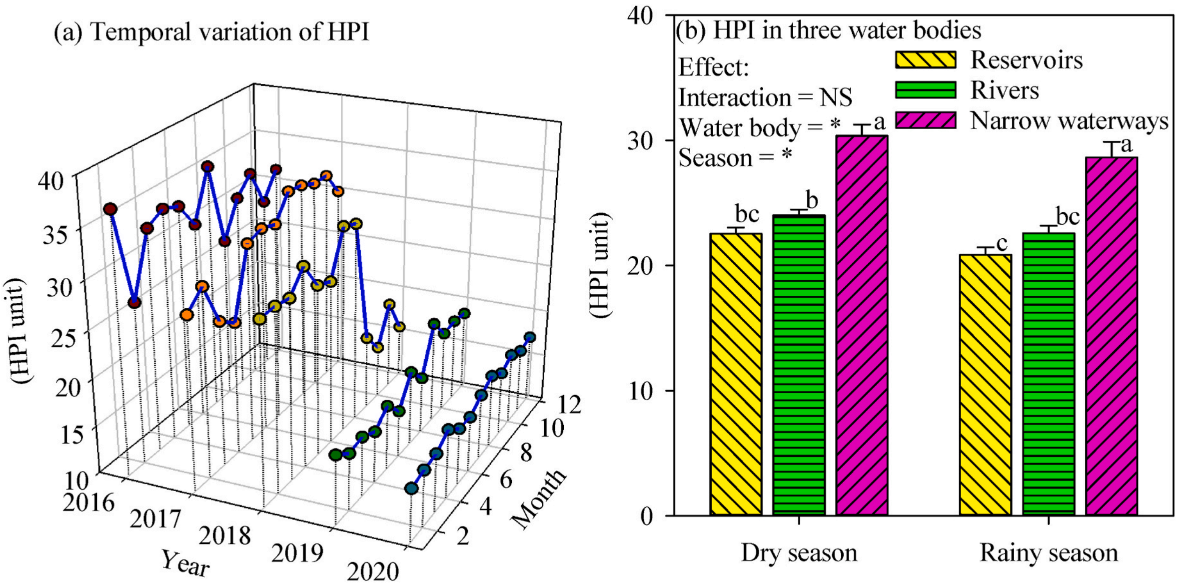

Fig. 1. Temporal variation of heavy metal pollution index (HPI) (a) and HPI of surface water collected in three water bodies and two seasons (b). NS and * indicate the associated effect was insignificant and significant, respectively. Error bars indicate the standard deviation of the mean. Within panel b, bars attached with the same letter were not significantly different from each other.

was significantly highest in rivers during the rainy seasons for both children (0.014) and adults (0.004), and lowest in reservoirs during the dry season (0.006 for children and 0.002 for adults). The HQ of Cr was not significantly affected by the water body and the season, varying from 0.33 to 0.37 for children and from 0.096 to 0.107 for adults. The HQ of Zn varied from 0.0015 to 0.0028 for children and 0.00066 to 0.00126 for adults. Narrow waterways had the highest HQ of Zn (0.0027 for children and 0.0012 for adults), while reservoirs showed the lowest (0.0016 for children and 0.00069 for adults). The HQ of Cu, Cd, and As for both children and adults was highest in narrow waterways and relatively similar in the other two water bodies. The HQ of Pb was significantly affected by the water body, with narrow waterways showing the greatest HQ for both children and adults, while reservoirs exhibited the lowest.

3.3. Multivariate analysis and relationship

The principal component analysis/factor analysis was conducted using the entire dataset of seven metals measured in the current study, revealing three latent factors. Factor 1 explained 20.08% of the total variance of the entire dataset, factor 2 explained 16.49%, and factor 3 explained 15.68% (Table 2). These three factors together accounted for 58.25% of the total variance. Factor 1 had a strong relationship with Cu, Zn, and As; factor 2 exhibited high loading values with Cd and Fe and factor 3 showed a significant correlation coefficient with Cr. Supplementary Table 3 indicated that the HI for both children and adults had a significant correlation with all examined metals (excluding Fe) and HPI with Pearson correlation coefficients greater than 0.35. The HPI demonstrated a strong relationship with five metals, including Zn, Cu, Pb, Cd, and As, with correlation coefficients greater than 0.73.

The percentage contribution of individual metals to the variance of HPI and HI for children and adults was shown in Table 3. The HPI showed a significant correlation with Fe, Cr, Pb, Cd, and As, but an insignificant correlation with Zn and Cu. Among the seven metals measured in the current study, Cd had the highest percentage (84.4%) contributing to the total variance of the HPI, followed by Pb, As, Fe, Cr, Zn, and Cu. All seven metals accounted for nearly 100% of the total variance of the HI for both children and adults. Among the seven metals analyzed, As showed the greatest contribution to the total HI variance, representing 53.91% and 80.97% for children and adults, respectively, while Zn and Cu had negligible contributions, with percentages close to zero.

4. Discussion

4.1. Heavy metal pollution

The present study observed varying levels of elements, with Fe ranging from 12.1 to 2830 μg L^{-1}, Cr from 1.0 to 10.0 μg L^{-1}, Zn from 1.0 to 187 μg L^{-1}, Cu from 0.2 to 14.80 μg L^{-1}, Pb from 0.3 to 11.5 μg L^{-1}, Cd from 0.05 to 2.40 μg L^{-1}, and As from 1.48 to 8.60 μg L^{-1}. These findings align with the ranges reported by Haq et al. (2023), indicating Cr levels spanning 2.63–440 μg L^{-1}, Zn from 12.1 to 1140 μg L^{-1}, Cu from 2.02 to 450 μg L^{-1}, Pb from 0.96 to 55 μg L^{-1}, Cd from 0.04 to 53 μg L^{-1}, and As from 1.25 to 9.36 μg L^{-1} across various water bodies in different countries. The findings from Fig. 1b confirmed the first research hypothesis that water derived from narrow waterways was more polluted with heavy metals than that derived from rivers and reservoirs. The variation in metal pollution levels across three water bodies could be attributed to their respective sources and physical characteristics. Three main characteristics that differ among these three water bodies may include water-storage capacity, flow velocity, and spatial position. Narrow waterways, characterized by narrow channels (less than 5–7 m wide), generally have the smallest water-storage capacity among the three water bodies examined. In terms of spatial position, narrow waterways are situated in locations directly connected to the sources of pollutants, such as agricultural and residential areas. Hence, they are more susceptible to pollution as they may directly receive wastewater discharged from these pollutant-generating regions. Similarly, Kumar et al. (2020) found varying metal levels in different water bodies, attributing the differences to pollutant sources and the spatial distribution of these water bodies. Likewise, ponds demonstrated the highest total concentration of toxic metals among the three water bodies—reservoirs, ponds, and lakes—due to their comparatively low water volume (Tokatlı and Varol, 2021). On the other hand, reservoirs, with their larger capacity, can effectively dilute pollutants due to their ability to hold a greater volume of water.

Moreover, the pollution of heavy metals in the three water bodies may be affected by their water residence time (WRT), which refers to the average time that a volume of water remains within a given water body.

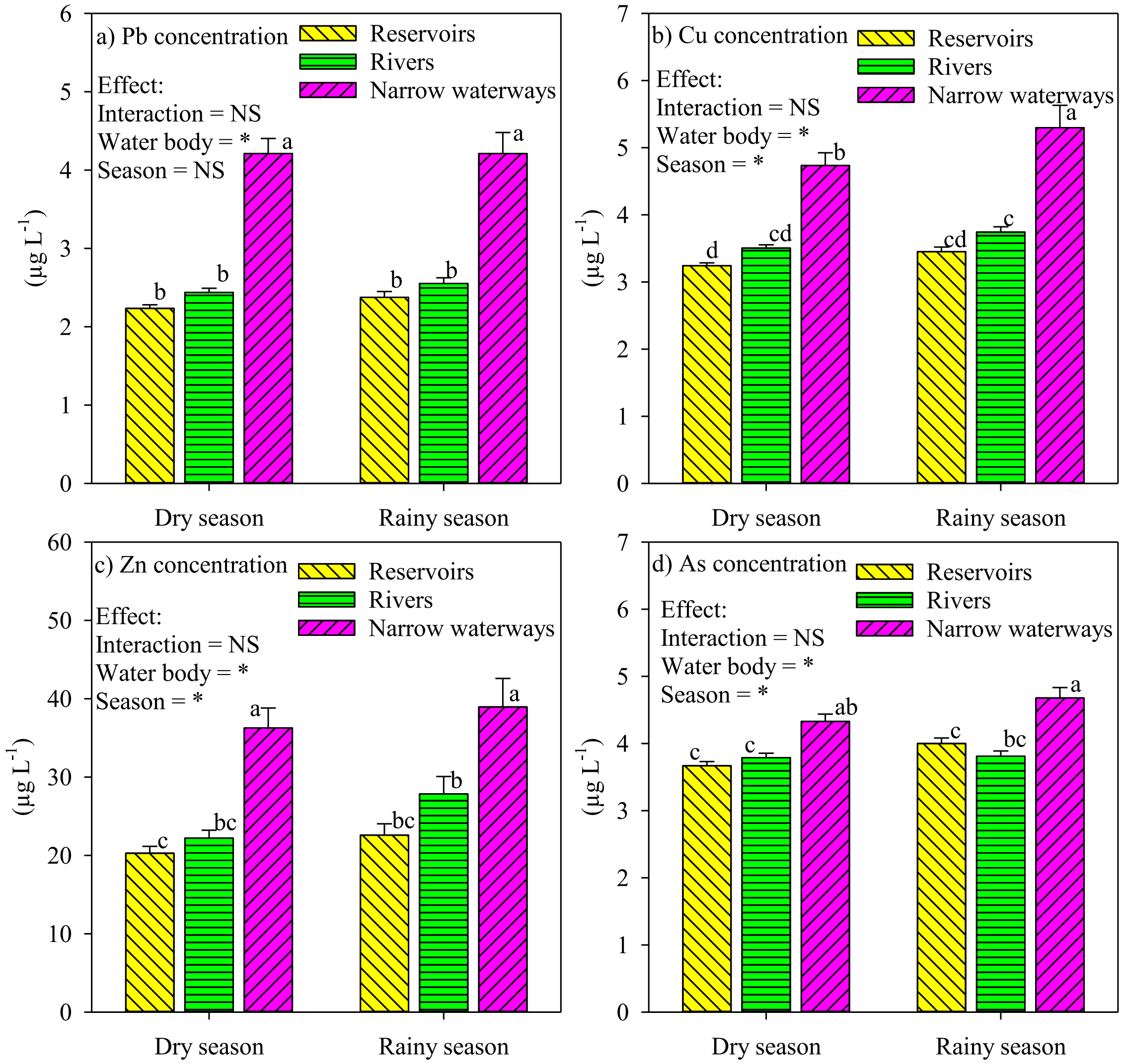

Fig. 2. The concentration of Pb (a), Cu (b), Zn (c), and As (d) in surface water collected from three water bodies in two seasons. NS and * indicate the associated effect was insignificant and significant, respectively. Error bars indicate the standard deviation of the mean. Within a panel, bars attached with the same letter were not significantly different from each other.

WRT is determined by the water-storage capacity and the flow velocity, which is additionally dependent on the total discharge, slope, and drainage area for the water bodies (Feng et al., 2018). A water body with a longer WRT may be cleaner due to self-purification and pollutant sedimentation (Zhang et al., 2019). As MRT is commonly estimated by dividing the water-storage capacity by flow velocity (Rueda et al., 2006), narrow waterways likely have the shortest WRT due to their smallest water-storage capacity and the greatest flow velocity, while reservoirs may have the longest WRT. Consequently, the longer WRT of reservoirs may contribute to greater water quality, leading to reduced heavy metal pollution, relative to rivers and narrow waterways, which was in line with Md Anawar and Chowdhury (2020).

The HPI was significantly higher during the dry season than during the rainy season (Fig. 1), validating our second research hypothesis. The finding suggests that water in the three water bodies was more polluted with heavy metals during the dry season than during the rainy season. High precipitation during the rainy season may have dual effects on metal concentrations in surface water. The first effect may be linked to the rapid and strong transport of metals from their origins to water bodies during the rainy season, leading to more pollution of heavy metals in the tested water bodies. This effect was observed in Cu, Zn, As, and Fe, which showed higher concentrations during the rainy season than during the dry season (Fig. 2b, c, 2d, and 2b). Similarly, the concentration of some metals such as Cr, Zn, and Cd was found to be higher during the monsoon season than during the pre-monsoon and post-monsoon seasons (Kshetriya et al., 2021). Conversely, the rainy season may introduce another important mechanism of the dilution effect. More rainwater during the rainy season may dilute pollutants including heavy metals, lowering their concentration in various water bodies (Meng et al., 2022). This effect was observed for Cd (Fig. 3a), contributing the majority (84.4%) to the total variance of HPI (Table 3), lowering HPI (Fig. 1b) during the rainy season compared to the dry season. This dilution effect was responsible for the higher concentration of trace elements in water samples collected during the dry season than during the rainy season in the Saigon River (Nguyen et al., 2020).

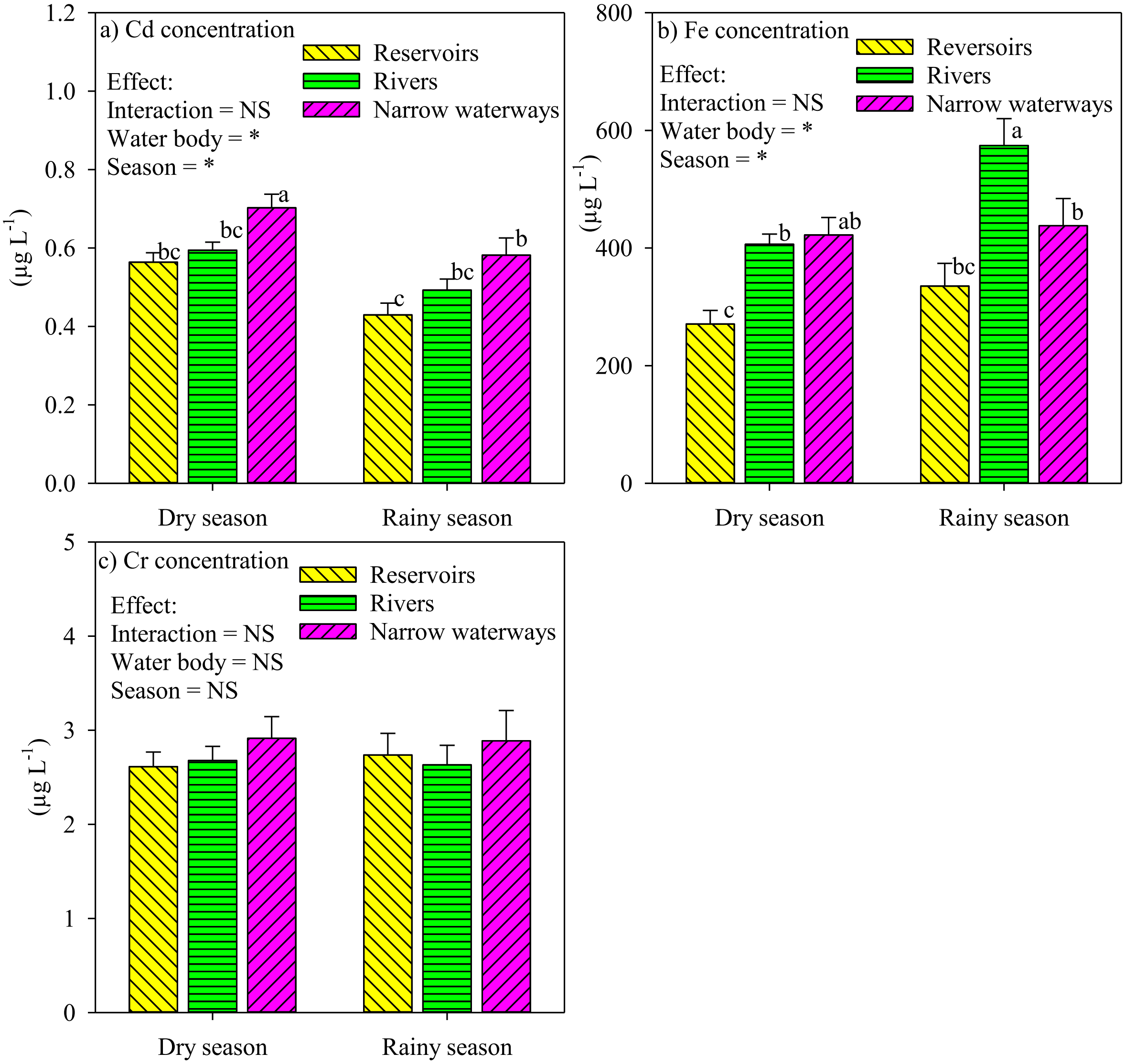

Fig. 3. The concentration of Cd (a), Fe (b), and Cr (c)) in surface water collected from three water bodies in two seasons. NS and * indicate the associated effect was insignificant and significant, respectively. Error bars indicate the standard deviation of the mean. Within a panel, bars attached with the same letter were not significantly different from each other.

4.2. Human health risk assessment

The varying levels of heavy metal pollution in three water bodies could potentially influence human health differently, depending on age. As shown in Fig. 4, the HI for children was significantly higher than that for adults, confirming our third hypothesis. In all three water bodies, the HI for children, but not for adults, exceeded one, indicating that the potential exposure to surface water could pose a significant non-carcinogenic risk to the health of children, in agreement with previous studies (Karimi et al., 2020; Shafiuddin Ahmed et al., 2021). These findings suggest that children, with weaker immune function and lower education levels than adults, are more sensitive to heavy metal pollution, which aligns with previous studies (Karimi et al., 2020; Shu et al., 2020). Consequently, more attention must be paid to children in avoiding the harmful impacts of heavy metals from surface water systems.

In general, a heavy metal may adversely affect human health when its HQ exceeds one (Ustaoğlu and Aydin, 2020). Although individual heavy metal concentrations in the current study were below the adverse health threshold (HQ < 1), the HI values for children (Fig. 4a) exceeding one indicate that a combination of the seven metals may have adverse impacts on children's health. The metals can enter and accumulate in the human body through various pathways, such as direct ingestion, dermal contact, inhalation, and the food chain (Naveedullah et al., 2014). Water from reservoirs and rivers may be used for daily drinking after processing, while that from all three water bodies may be used for irrigation in agricultural production. Daily contact with surface water from these water bodies may also happen, which may influence human health. Therefore, besides reducing the metal concentration in surface water bodies, it is essential to regulate pathways of metal entry to minimize the risk of heavy metal pollution in surface water systems and safeguard human health.

4.3. Pollution sources and implications

In the present study, traffic and industrial emissions, the discharge of industrial and domestic sewages, and paddy-field runoff could be the main source of the examined heavy metals (Meng et al., 2022; Nazarpour et al., 2019; Varol and Tokatlı, 2021). The PCA/FA suggested three primary pollution sources responsible for emitting the seven heavy metals examined in the current study (Table 2). The first significant source, responsible for Pb, Cu, Zn, and As, having inter-relationships

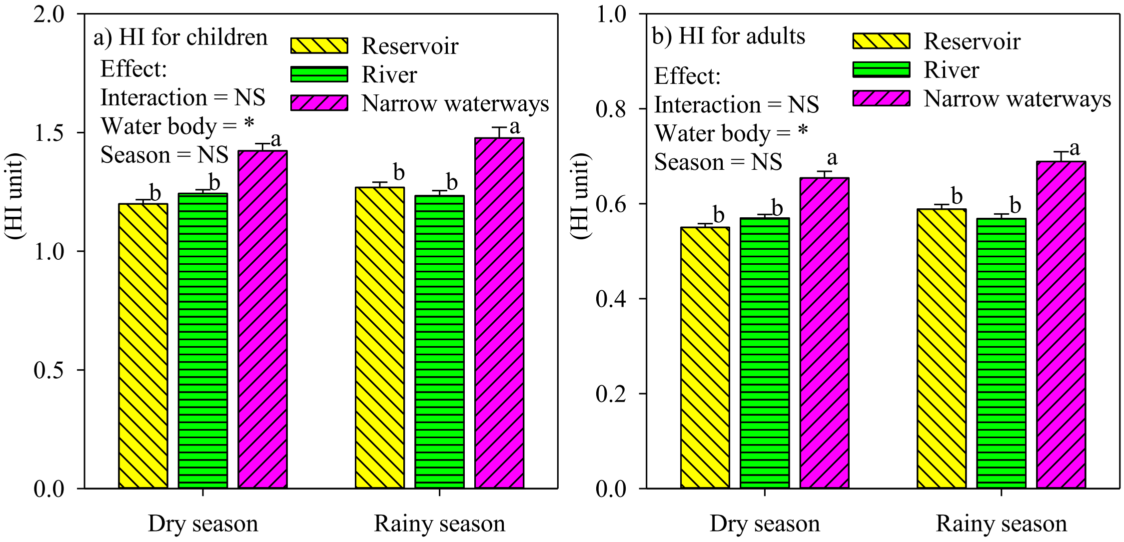

Fig. 4. Children and adult hazard index (HI) of surface water collected in three water bodies and two seasons. NS and * indicate the associated effect was insignificant and significant, respectively. Error bars indicate the standard deviation of the mean. Within panel b, bars attached with the same letter were not significantly different from each other.

Table 1
The hazard quotient (HQ) of individual heavy metals in water samples collected in three water bodies and two seasons. NS and * indicate the associated effect is insignificant and significant, respectively. The numbers in the parenthesis are the standard deviation of the mean. Within a column, data attached with the same letter were not significantly different from each other.

Water bodies	Season	Fe	Cr	Zn	Cu	Pb	Cd	As
For children								
Reservoir	Dry season	0.006^{c}	0.331^{a}	0.001^{c}	0.004^{d}	0.028^{b}	0.075^{b}	0.753^{c}
		(0.001)	(0.012)	(0.0001)	(0.00005)	(0.0006)	(0.003)	(0.01)
	Rainy season	0.008^{b}	0.347^{a}	0.002^{bc}	0.004^{d}	0.030^{b}	0.057^{c}	0.821^{bc}
		(0.001)	(0.018)	(0.0001)	(0.00008)	(0.0009)	(0.004)	(0.02)
River	Dry season	0.010^{b}	0.340^{a}	0.002^{bc}	0.004^{d}	0.031^{b}	0.079^{b}	0.778^{c}
		(0.0004)	(0.011)	(0.0001)	(0.00006)	(0.0007)	(0.003)	(0.01)
	Rainy season	0.014^{a}	0.334^{a}	0.002^{b}	0.004^{c}	0.032^{b}	0.065^{bc}	0.783^{c}
		(0.0011)	(0.016)	(0.0002)	(0.00009)	(0.0009)	(0.004)	(0.02)
Narrow waterway	Dry season	0.010^{b}	0.370^{a}	0.003^{a}	0.005^{b}	0.053^{a}	0.093^{a}	0.889^{ab}
		(0.0007)	(0.018)	(0.0002)	(0.00021)	(0.0024)	(0.005)	(0.02)
	Rainy season	0.010^{ab}	0.366^{a}	0.003^{a}	0.006^{a}	0.053^{a}	0.077^{bc}	0.961^{a}
		(0.0011)	(0.024)	(0.0003)	(0.00038)	(0.0034)	(0.006)	(0.03)
Effect test	Water body	*	NS	*	*	*	*	*
	Season	*	NS	*	*	NS	*	*
	Water body x season	NS	NS	NS	NS	NS	NS	NS
For adult								
Reservoir	Dry season	0.002^{c}	0.096^{a}	0.00066^{c}	0.0019^{d}	0.011^{b}	0.022^{b}	0.417^{c}
		(0.0002)	(0.003)	(0.00003)	(0.00002)	(0.0002)	(0.001)	(0.01)
	Rainy season	0.002^{bc}	0.100^{a}	0.00073^{bc}	0.0020^{cd}	0.012^{b}	0.017^{c}	0.454^{bc}
		(0.0003)	(0.005)	(0.00005)	(0.00004)	(0.0004)	(0.001)	(0.01)
River	Dry season	0.003^{b}	0.098^{a}	0.00072^{bc}	0.0020^{cd}	0.012^{b}	0.024^{b}	0.430^{c}
		(0.0001)	(0.003)	(0.00003)	(0.00003)	(0.0003)	(0.001)	(0.01)
	Rainy season	0.004^{a}	0.096^{a}	0.00090^{b}	0.0021^{c}	0.013^{b}	0.020^{bc}	0.433^{c}
		(0.0003)	(0.005)	(0.00007)	(0.00005)	(0.0004)	(0.001)	(0.01)
Narrow waterways	Dry season	0.003^{b}	0.107^{a}	0.00117^{a}	0.0027^{b}	0.021^{a}	0.028^{a}	0.492^{ab}
		(0.0002)	(0.005)	(0.00008)	(0.00011)	(0.001)	(0.001)	(0.01)
	Rainy season	0.003^{ab}	0.106^{a}	0.00126^{a}	0.0030^{a}	0.021^{a}	0.023^{bc}	0.531^{a}
		(0.0003)	(0.007)	(0.00012)	(0.00019)	(0.0014)	(0.002)	(0.02)
Effect test	Water body	*	NS	*	*	*	*	*
	Season	*	NS	*	*	NS	*	*
	Water body x season	NS	NS	NS	NS	NS	NS	NS

with each other, likely originated from anthropogenic combustion emissions/street dust, including traffic, vehicle emissions, and industrial activities (Nazarpour et al., 2019). Similarly, local traffic and industrial emissions have been identified as the primary sources of Pb, Cu, and Zn found in surface/street dust (Cheng et al., 2011; Hou et al., 2019; Kumar et al., 2020). Additionally, various metals were found in street dust in

Table 2

Loading values of seven heavy metals from principal component analysis/factor analysis. Bold numbers are those greater than 0.5.

Parameter	Factor 1	Factor 2	Factor 3
Pb	**0.74**	0.01	0.20
Cu	**0.69**	−0.36	0.07
Zn	**0.63**	0.23	−0.01
As	**0.57**	0.32	−0.47
Cd	−0.01	**0.83**	−0.04
Fe	0.14	**0.55**	0.31
Cr	0.10	0.02	**0.82**
Eigenvalue	1.83	1.15	1.10
Percent	26.08	16.49	15.68
Cumulative Percent (%)	6.08	42.57	58.25

various parts of Vietnam (Nguyen et al., 2022; Nguyen et al., 2021; Thuy et al., 2022). Arsenic, naturally present in the earth's crust and thus in rocks and soils, may be released into surface water bodies through various processes such as mining, natural weathering, and the oxidation of As-contained minerals (Komorowicz and Barałkiewicz, 2016; Uugwanga and Kgabi, 2021). Arsenic could be present in street dust, which could be derived from decomposed animal and plant residues, damaged and aged pavement substrates, worn and aged automobile tires, automobile exhaust emissions, and waste gas from industrial emissions (Cai and Zhang, 2021). Deposited dust could be more likely eroded and transported to the water bodies, leading to higher metal concentrations in the rainy season than in the dry season, as shown in Fig. 2.

The second pollution source, accounting for the presence of Fe and Cd, may be linked to soil erosion or run-off from agricultural land. Iron, naturally abundant in soil and essential for living organisms (Abbaspour et al., 2014), can be carried out to various water bodies through the transportation of surface soil. Enrichment of Fe in water bodies can also result from the decomposition and transport of organic waste. Furthermore, Fe can bind organic matter in the form of mineral-organic aggregates (ThomasArrigo et al., 2018), leading to higher Fe content in organic matter-rich soils. On the other hand, cadmium, a widely distributed heavy metal in the environment, can originate from various sources such as mining, combustion emissions, and especially the use of Cd-containing fertilizers (Kubier et al., 2019). Cadmium can form water-soluble complexes with dissolved organic matter, enhancing its abundance in water bodies (Kubier et al., 2019). Therefore, agricultural soils can contain elevated levels of Fe and Cd due to various reasons such as the application of Cd-containing fertilizers, the presence of Fe-containing soils, or the use of compost/organic matter. Erosion and transport of these soils with rainwater contribute to the presence and abundance of these metals in water bodies. Nevertheless, Fig. 4 revealed significant seasonal variations, with higher Cd levels during the dry season and greater Fe levels during the rainy season. While the exact reasons are unknown and require more studies for confirmation, these disparities may be involved in rice production in the study area. Khanh Hoa boasts the largest rice cultivation area among various types of agricultural land use, covering nearly half of the total crop areas, approximately 100,000 ha of the province (Kim and Minh, 2022). Rice cultivation in the province occurs primarily in two seasons: the winter-spring crop (December to April and the summer-autumn crop (April to August) each year. Consequently, during the dry season, the application of fertilizers for rice cultivation may potentially elevate Cd content in surface water due to surface runoff of Cd-contained fertilizers (Kubier et al., 2019). Conversely, the runoff of Fe-contained soil from agricultural areas could be more pronounced during the rainy season than the dry season, contributing to higher Fe levels during the rainy season, as observed in Fig. 4.

chromium in the current study may originate from a variety of anthropogenic activities and natural processes (Brasili et al., 2020; Kazakis et al., 2017). Human activities such as mining and metal works, steel and metal alloy production, paint manufacturing, and dyeing can contribute to the release of Cr into the environment (Tumolo et al., 2020). Furthermore, fertilizers used in agriculture may contain a high amount of Cr (Krüger et al., 2017), enriching Cr in surface water through agricultural soil erosion. Natural weathering of ultramafic rocks also contributes to elevated Cr levels in its associated soils and sediment as well as groundwater (Chrysochoou et al., 2016). Due to the diverse Cr sources, the Cr concentration as well as its HQ did not show significant differences among the three examined water bodies or between the two seasons in the current study (Fig. 3a and Table 1). These suggest that the management of Cr pollution requires efficient approaches due to the involvement of various sources.

The current study identifies Cd, As, and Cr as the most important heavy metals in the study area, potentially causing environmental problems and toxicity to human health, particularly for children. In terms of environmental pollution, Cd is highlighted as the most crucial metal among the seven examined in the current study, accounting for the highest percentage (84.44%) of the total variance of HPI (Table 3). Regarding human health risks, As emerges as the most important element with the greatest HQs for children and adults (Table 1), contributing the greatest proportion to explaining the total variance of the HIs (Table 3). Following As, chromium could be considered the second most important metal, accounting for a considerable proportion of the total variance of the HI for children and adults. Three elements, Cd, As, and Cr can be present in various environmental compartments (Komorowicz and Barałkiewicz, 2016; Kubier et al., 2019; Weerasundara et al., 2021; Xing et al., 2022) and poses serious environmental and human health concerns (Xing et al., 2022). While managing pollution sources to lower their concentration in water bodies may be challenging, considering options to remove them from polluted water could be a potential solution. Nevertheless, further studies focusing on pollution source management and the development of technologies for heavy metal removal from surface water bodies are necessary to promote a better environment, protect human health, and contribute to sustainable development.

Table 3

Contributive percentage of individual metals in explaining the total variance of the HPI and HIs in the study area. *$P < 0.05$ indicates that the contribution of the associated metal is statistically significant.

Metal	HPI			HI for children			HI for adult		
	Sum of Squares	Prob > F*	Contribution (%)	Sum of Squares	Prob > F*	Contribution (%)	Sum of Squares	Prob > F*	Contribution (%)
Fe	778.7	<.0001	1.00	0.1	<.0001	0.10	0.0	<.0001	0.05
Cr	14.8	<.0001	0.02	45.8	<.0001	42.91	3.8	<.0001	17.50
Zn	0.00	0.5	0.00	0.0	<.0001	0.00	0.0	<.0001	0.00
Cu	0.00	0.6	0.00	0.0	<.0001	0.00	0.0	<.0001	0.00
Pb	6965.2	<.0001	8.92	0.3	<.0001	0.33	0.1	<.0001	0.26
Cd	65,937.9	<.0001	84.44	2.9	<.0001	2.74	0.3	<.0001	1.22
As	4388.1	<.0001	5.62	57.5	<.0001	53.91	17.6	<.0001	80.97
Error	1.1		0.00	0.0		0.00	0.00		0.00
Total variance	78,085.8		100.0	106.7		100.0	21.7		100.00

5. Conclusion

The present study observed varying levels of heavy metal pollution in the surface water, influenced by different water bodies and seasonal changes. Narrow waterways exhibited higher levels of heavy metal pollution (29.5 units) compared to rivers (23.3 units) and reservoirs (21.7 units). Additionally, the dry season showed a higher heavy metal pollution index than the rainy season. The effect of the seasonal changes may be involved in the transport of metals from their sources to the water bodies. While heavy metal pollution in the three water bodies exhibits some potentially adverse influences on children's health, it does not produce similar effects on adults' health. The seven examined metals could originate from three primary sources, including combustion emissions/street dust (26.08%), run-off from agricultural land (16.49%), and various sources (15.68%). Cadmium was identified as the most influential in determining the heavy metal pollution index, while arsenic and chromium were highlighted as the two key elements potentially affecting children's health. Because children are more sensitive to the potential impacts of heavy metal pollution than adults, protecting children from its potential impacts should be prioritized over protecting adults.

CRediT authorship contribution statement

Thang Viet Le: Formal analysis, Investigation, Resources, Writing - original draft, Methodology, Software. **Binh Thanh Nguyen:** Conceptualization, Visualization, Writing - original draft, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.123216.

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