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Distribution, sources, and human health risks of heavy metal pollution from mining activities in the Aras River (Iran–Armenia border)

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Abstract

Background: This study evaluates heavy metal contamination resulting from mining activities in the Aras River, with a focus on assessing human health risks, quantifying sediment metal concentrations, and identifying pollution sources through statistical analyses.

Methods: Surface sediment samples were collected from multiple sites along the Aras River on the Iran–Armenia border, particularly near mining operations and tailing dams, during August and December 2020. Twelve samples were obtained from upstream and downstream of the Agharak Mine tailing dams. Concentrations of As, Mo, Cr, Cu, Ni, Pb, Zn, Hg, Fe, and Al were measured using ICP-OES to assess environmental and human health risks.

Results: The estimated Average Daily Dose (ADD) values for all heavy metals studied were below standard guidelines, with higher ingestion rates observed in children. The HQ values for exposure pathways were generally below 1, suggesting low risk, although the HI values for children suggested a potential non-cancer risk in the future. Nickel concentrations exceeded probable effect levels (PELs) in 84% of the samples, while Hg showed unique local sources and minimal correlations with other metals. Principal component analysis (PCA) identified two components, explaining 89.7% of the variance, with PC1 accounting for 50.08% and showing strong loadings for As, Zn, Pb, Cu, and Ni. The main limitation of this study was the small sample size, which may limit the generalizability of the results.

Conclusion: The results underscore the importance of closely monitoring and effectively controlling heavy metal contamination in the Aras River to safeguard the ecosystem and human well-being.

Keywords: Rivers, Health risk assessment, Mining, Sediment analysis, Heavy metals

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Introduction

Environmental pollution and its impact on ecosystems and human health remain pressing concerns in contemporary society (1). Recent findings have highlighted the significant strain on rivers from residential, tourism, industrial, and commercial activities, leading to substantial pollutant discharge (2). As with other aspects of technological progress, rivers have been victims of aggressive human development (3). Heavy metals are considered key pollutants in water systems due to their toxicity, persistence, and bioaccumulation. However, heavy metals such as Cu, Mo, Pb, Al, Hg, and As have been the most common pollutants in recent years. Several acute and chronic toxic effects of heavy metals on different human bodies, such as nervous system disorders, skin lesions, vascular damage, and cancer, have been demonstrated. These metals enter river systems through natural and anthropogenic sources, including mining activities and mineral processing (4). Adsorption, hydrolysis, and co-precipitation processes lead to the accumulation of significant amounts of metal ions in sediments, making them both a source and a reservoir for metal contaminants (5). Over time, sediments serve as a historical record of anthropogenic pollution and play a crucial role in assessing the impact on aquatic ecosystems, affecting benthic organisms, and propagating through the food web (6,7).

Advanced multivariate statistical approaches, such as principal component analysis (PCA) and correlation coefficients, are routinely used to differentiate between different sources of heavy metals in sediments (8,9). By examining the chemical dispersion of high-density metallic elements in river sediments, scientists can gain a thorough understanding of the potential hazards these substances pose to both ecosystems and human communities (10). This knowledge, derived from sediment chemical profiles, offers a holistic perspective on the potential risks of heavy metal exposure, providing valuable insights for risk assessment and management strategies in the environmental and public health domains.

International rivers play a crucial role in global water assessment. There is no doubt that water bodies that cross borders between countries facilitate general cooperation among nations, necessitating greater attention (11). According to a United Nations report, there are nearly 214 international waterways covering 47% of the total continental land surface (12). The Aras transboundary river system is a globally significant international river system in Asia, originating in Bingöl Mountain in Turkey and flowing 275 miles along the international boundary between Armenia and Azerbaijan to the north and Turkey and Iran to the south, passing through the Muğan Steppe of Armenia (13).

In these regions, the population relies heavily on the Aras River as a primary water source for industry, agriculture, energy, and residential users. However, rivers between countries routinely pose broader environmental challenges, such as hydropower, irrigation, flood management, and water pollution (14,15). Concerns about the quality of the Aras River are increasing due to a wide range of human *activities,* mainly agricultural and industrial activities near this area, as well as the lack of an appropriate industrial effluent collection and treatment system that allows for the gradual discharge of industrial effluent into the river (13,16). The Aras River health risk, associated with the lead (Pb), zinc (Zn), nickel (Ni), copper (Cu), and molybdenum (Mo) originating from mine waste and the current crisis in the Kura-Aras River basin, is rooted in active copper and molybdenum mines in the countries around the boundary of the Aras River (17). Mining activities, along with agricultural expansion and urban development, have significantly impacted the Aras River Basin, altering land cover and influencing the river's hydrology and water quality (13,18). Based on existing studies in the study area, severe contamination of agricultural soil and inhabited areas with heavy metals has been reported from Kajaran town (19), Alaverdi (20), and Agarak (21). Possible sources of these elevated levels include dust and waste deriving from mining and processing, as well as from smelting activities. Petrosyan et al show that children's blood lead levels in the mining town Akhtala are higher than in other areas (22). The toxic levels of Mo, Cu, Pb, and Hg are directly linked to the environment, affecting human and ecosystem health and causing complications such as gastrointestinal cancer, neurological effects, diabetes, and infant mortality (23-25). This preliminary study has a limited sampling scope, and the objectives of this study are to (1) assess the human health risk of heavy metals in the Aras River sediment; (2) measure the concentration of heavy metals in sediments of the Aras River (Iranian-Armenian Border); and (3) quantify the contributions of sources using statistical techniques such as PCA and correlation coefficients.

Material and methods

Study area

This study was conducted in East Azerbaijan Province, located on Iran's northwest border. Sampling sites on the Aras River begin at the end of the Iran–Nakhchivan border and extend along the entire shared border between Iran and Armenia (Figure 1). Armenia and Iran host three tailing dams of the Armenian Agarak Mine. The Armenian Agarak Mine is an important center for non-ferrous metallurgy, an industry that produces copper and molybdenum concentrates through bulk-selective flotation recovery of their minerals. It is considered a vital Mo production center. Reports estimate a tailing storage of 80.45 million m^3. However, the total volume of waste tailing storage was reported as 46.7 million m3, with the remaining 33.75 million m3 reported, while satellite images show the filling of tailing dams (26). The lack of an efficient recirculation system and the empty tailing storage volume have led to the discharge of waste containing mineral tailings into the Aras River.

Sample collection and analysis

Surface sediment Samples were gathered at random from different points along the Aras River, including areas upstream of the Agarak mine (Norduz), between tailing dams 2 and 3 (Duzal), and downstream of three tailing dams (Kordasht) over two months (August and December 2020) (Figure 1 and Table S1). A total of six sediment samples (up to 15 cm in depth, about 500 g each) were collected from the riverbed. Sample bottles were washed and rinsed with deionized water in the lab and then rinsed with river water before collecting the sample. A plastic grab sampler device was used to collect sediment samples at each site. Samples were stored at 4° C and were transferred to the laboratory. The sediment samples were air-dried in the shade, homogenized, and sieved through a 2-mm sieve. The chemical analysis comprised digesting 0.5 g of sediment with 10 mL of aqua regia (1:3 HCl: HNO_3). Samples were digested at 80 °C for 3 h, or until the slurry reached 1 mL. The samples were diluted to 25 mL with distilled water.

All liquid sample aliquots were filtered through 0.45 µm membrane filters to *determine* heavy metal concentrations of Arsenic (As), Molybdenum (Mo), Chromium (Cr), Copper (Cu), Nickel (Ni), Lead (Pb), Zinc (Zn), Mercury (Hg), Iron (Fe), and Aluminum (Al) by the Inductively

Figure 1. Location of tailings dams and sampling points on the Aras River

Coupled Plasma Optical Emission Spectroscopy (ICP-OES) *technique* (standard methods US EPA 3050B, Agilent 7500, ICP-Mass, USA) (27). All analyses were carried out in triplicate, and certified reference materials (Agilent Japan standard solution, Lot number 10-160YPY2) were used to ensure accurate and consistent total analysis and analytical quality.

Human health risk assessment

Colloidal-sized metals introduced into the river through tailing dam effluent discharge accumulate in both the water and sediments within the river system. When water containing colloidal particles is used for irrigation, the metals can be transferred to agricultural lands where they may settle. The contaminated sediments can lead to soil pollution, facilitating the uptake of metals by plants. Subsequently, these metals enter the food chain through livestock consumption, accumulate in milk and meat, and ultimately enter the human food cycle. Additionally, fish harvested from this river may be contaminated with heavy metals, posing a risk to human health.

Furthermore, during dry seasons when the water level of the Aras River decreases and sediments are exposed to the atmosphere, colloidal particles may become suspended in the wind, potentially leading to respiratory exposure. Children playing along the riverbank in soil contaminated by these metals may also be exposed to these pollutants through dermal contact. These exposure pathways underscore the need for comprehensive risk assessment across all potential routes to safeguard human health integrity.

The exposure risks posed by sediment metals to the public are calculated using the United States Environmental Protection Agency's recommended health risk assessment formula (28,29). Ingestion of particles (ADDing), inhalation (ADDinhal), and dermal (ADDderm) absorption of trace elements via skin are the three major direct pathways for human exposure (24,30). Presently, heavy metals are used to assess health risks in sediments; As, Cr, Ni, and Pb pose carcinogenic risks, while heavy metals are noncarcinogenic.

We examined children and adults. The noncarcinogenic Hazard quotient (HQ) is obtained by dividing the calculated doses for each element and exposure pathway by the toxicity threshold value, referred to as a specific element's reference dose (RfD). The hazard index (HI) is the sum of the noncarcinogenic risks from all exposure pathways to various pollutants (29). The carcinogenic risk level is determined by multiplying the dose by the carcinogen's corresponding slope factor (SF). This is the likelihood of an individual developing any form of cancer during their lifespan as a result of exposure to carcinogenic hazards (31).

"HQ" refers to noncarcinogenic risks, quantified as the likelihood that a person will experience a negative consequence. The heavy metal toxicity index (HLD) is defined as the ratio of the average daily dose (ADD), expressed in mg/kg-day, to the chronic reference dose (RfD), also expressed in mg/kg-day, for the metal in question. An increase in ingested pollutants occurs when the exposure dose exceeds the oral reference dose and the homogeneity coefficient (HQ) is less than 1. While the exposed population (consumers) is considered safe, a health hazard ratio (HQ) of 1 or higher indicates that human health is at risk. Implementing appropriate preventive measures is necessary. Nevertheless, the HQ parameter does not estimate the risks; it merely defines a degree of risk associated with pollutant exposure. The noncarcinogenic risk was determined by calculating the Hazardous Quantification (HQ) parameter for heavy metals using the following equation:

Therefore, ADD refers to the acceptable daily intake for heavy metal exposure, whereas RfD represents the oral reference dose. The oral reference dose is the amount of a chemical that may be consumed daily without any discernible risk of long-term or daily oral exposure to the human population, and without harmful consequences throughout a person's lifetime. Quantification of heavy metal intake by soil was performed using the following equation:

Equation 1: $$ADD_{ing} = C \times \frac{IR \cdot EF \cdot ED}{BW \cdot AT} \times 10^{-6}$$

ADD_{ing} is the average daily intake of heavy metals, calculated from the heavy metal concentration in soil, expressed in mg/kg. *IR* is the soil ingestion rate in days/year; EF is the exposure frequency; *ED* is the exposure duration in years; *BW* is the body weight; *AT* is the average time.

Equation 2: $$ADD_{inhal} = C \times \frac{InhR \cdot EF \cdot ED}{PEF \cdot BW \cdot AT}$$

ADD_{inhal} is the average daily intake of heavy metals inhaled from the sediment at mg/kg-day, *C* indicates the concentration of heavy metal in the sediment in mg/kg, InhR is the soil inhalation rate in mg/day, and PEF is the particulate emission factor in m^3/kg. *EF*, *ED*, *BW*, and *AT* have been defined earlier in Equation 1.

Equation 3: $$ADD_{derm} = C \times \frac{SA \cdot AF \cdot ABS \cdot EF \cdot ED}{BW \cdot AT} \times 10^{-6}$$

ADD_{derm} is the dosage of exposure over the skin, measured in milligrams per kilogram per day. *C* is the heavy metal content in the sediment in milligrams per kilogram; *SA* indicates the exposed skin area in square centimeters. *SL* is the adherence factor, measured in milligrams per square centimeter per day; *ABS* is the percentage of the administered dose absorbed through the skin, measured as a unit. The variables *EF*, *ED*, *BW*, *CF*, and *AT* were previously expressed in Equation 1.

For the ten heavy metals considered, the noncarcinogenic effect on the population is determined by the sum of all hazard quotients (HQ) for individual heavy metals, collectively known as the hazard index (HI), as described by (28). Equation 4 shows the mathematical representation of this parameter:

Equation 4: $HI = \sum HQi = HQing + HQinhal + HQderm$

For carcinogens, the risk is estimated as the incremental probability that an individual will develop cancer over a lifetime due to exposure. The equation to calculate excess lifetime cancer risk is:

Equation 5: Cancer risk = ADD × SF

Calculation of the total excess lifetime cancer risk for an individual from the average contribution of the individual heavy metals for all pathways is done using the following equation:

Equation 6:
$RiskTotal = Riskingestion + Riskinhalation + Riskdermal$

The risk factor calculation involves multiplying the dosage by the appropriate slope factor to obtain the carcinogenic risk. A risk value ratio of less than 10^{-4} is considered within the permissible limit for carcinogenic risk. The recreational situation was examined for both adult and child receptors, focusing on inadvertent ingestion and skin contact with sediment during swimming activities as the primary routes of exposure (32,33).

No doubt irrigating surrounding farmlands with Aras River water containing suspended mining waste solids could have tragic health effects on agricultural and horticultural products, animal nutrition, and dairy products such as milk and cheese. Fortunately, the drinking water source in the study area is groundwater, and water from the Aras River is not used for drinking. However, the possible infiltration of Aras River water into groundwater could contaminate it.

Sediment quality guideline

Sediment quality guideline (SQG) values are developed to improve protection of the benthic ecosystem, fisheries, and surface water quality and are fundamental components of all sediment quality assessment frameworks (34).

The present study used total concentration-based SQG indices to assess the potential ecological risk posed by metals in the Aras River sediment. When metals are present in concentrations below the threshold effect levels (TEL) of SQGs, they pose low or no eco-risk. When concentrations fall between the TEL and the probable effect levels (PEL), they pose moderate eco-risk. When concentrations exceed the PEL, they pose high eco-risk (35).

Statistical analysis

Pearson's correlation is a statistical technique used to quantify the degree of association between two sets of data. The present work used Pearson's correlation analysis to examine the patterns of correlation among concentrations of multiple heavy metals. The relationships among the amounts of certain heavy metals at all sample locations were analyzed using a Pearson correlation matrix. Statistical correlation analysis serves to validate the findings of factor analysis. To minimize information loss, principal component analysis employs dimensionality reduction to convert multiple indices into a set of independent, comprehensive variables. These fundamental elements are linear combinations of the original variables and capture most of the information about the original measure (36). The Kaiser–Meyer–Olkin (KMO) value and Bartlett's test of sphericity were used to assess the adequacy of PCA. (37). Data were analyzed using SPSS Statistics for Windows, Version 21.0 (IBM SPSS Statistics for Windows).

Results

Human health risk assessment

The health risk assessment of exposure to heavy metals in sediment from the Aras River for residents (children and adults) was estimated based on Equations 1–6. The estimated daily intake (ADD) of heavy metals through ingestion, dermal contact, and inhalation for adults and children is presented in Table 1. We assumed long-term exposure for 24 years in adults and 6 years in children as the worst-case default assumption.

Noncarcinogenic health risk assessment

The noncarcinogenic toxic effects of heavy metals are usually characterized by calculating the hazard quotient (HQ). An HQ value exceeding 1 indicates potential adverse health effects from overexposure. To assess the human health risks associated with exposure to heavy metals, hazard quotients (HQing for ingestion, HQinhal for inhalation, and HQderm for dermal contact) and hazard index (HIing for ingestion, HIinhal for inhalation, and HIderm for dermal contact) were computed as main parameters. The relevant coefficients, such as RfD ingestion, RfD inhalation, and RfD dermal, were available for elements including As, Mo, Cr, Cu, Ni, Pb, Zn, Hg, Fe, and Al. Consequently, a noncarcinogenic risk assessment was conducted for these ten elements. The results presented in Table 2 indicate that the HQ values for sediment exposure pathways (direct ingestion, inhalation, and dermal contact) for adults and children across all investigated metals were lower than 1. Figure 2 illustrates the HQ values for adults and children at each sampling station.

Carcinogenic health risk assessment

After computing the exposure doses to As, Ni, Cr, and Pb, the potential carcinogenic risks associated with sediment exposure were determined using available slope factors. As shown in Table 2, the mean CR_{ing} values for sediments through ingestion exposure were 1.16E−05 (As), 1.85E−07 (Cr), 0.00E + 00 (Ni), and 2.17E−08 (Pb) for adults, and 1.63E−04 (As), 2.15E−06 (Cr), 0.00E + 00 (Ni), and 2.54E−07 (Pb) for children. The value of 1.63E−04 for children exceeds the acceptable threshold and may constitute an unacceptable risk, warranting intervention and management. For the inhalation route, the mean CR_{inhal} (inhalation carcinogenic risk) in the sediments through inhalation exposure was lower than 1.7E−08 (As, Cr, Ni, and Pb) for adults and 3.8E−08 for children. Regarding dermal exposure, CR_{derm} values for adults decreased in the order As > Cr > Pb > Ni, and for children, the order was As > Pb > Cr > Ni. The total human health risks (total CR) were calculated as the sum of the risks from these exposure routes. Carcinogenic risk from ingestion, inhalation, and dermal exposure was observed at the sampling station.

Heavy metal concentration in sediment

The data obtained from the analysis of sediment samples for the assessment of heavy metal concentrations in the Aras River, upstream and downstream of the tailing dams, in August and December 2020, have been summarized in Figure 2 and Table S2 (Supplementary File). In August, the average concentrations of metals in the sediments of the Aras River were ranked as follows: Fe > Al > Ni > Cr > Zn > Cu > As > Pb > Mo > Hg. However, in December, the ranking shifted, with Al being the highest, followed by Fe > Ni > Cr > Zn > Cu > As > Pb > Mo > Hg. In August, the mean concentrations (± SD, mg/kg) of metals in the sediments of the Aras River were: Fe (10483.67 ± 1502.51), Al (9808.00 ± 1886.92), Ni (80.67 ± 26.84), Cr (55.17 ± 15.22), Zn (31.33 ± 6.83), Cu (29.00 ± 10.76), As (5.17 ± 1.61), Pb (4.33 ± 0.76), Mo (0.49 ± 0.00), and Hg (0.05 ± 0.00). In December, the mean concentrations (± SD, mg/kg) were: Al (12187.67 ± 4037.51), Fe (11528.67 ± 844.28), Ni (73.17 ± 53.88), Cr (57.17 ± 41.50), Zn (45.67 ± 7.64), Cu (37.67 ± 7.69), As (9.00 ± 0.87), Pb (7.00 ± 0.50), Mo (0.66 ± 0.29), and Hg (0.05 ± 0.02).

Sediment quality guideline (SQG)

Sediment quality was evaluated by comparing concentrations with the probable effect level (PEL) and the threshold effect level (TEL). The SQG assesses ecological risk based on total concentrations and empirical toxicological experiments without accounting for regional variations in baseline metal levels and chemical compositions. In surface sediments, concentrations of Cr and Cu fell between the TEL and PEL values in 50%

Table 1. Average daily dose(ADD: mg kg-1 day-1) for metals by ingestion and dermal exposure to sediment

Adults	As	Mo	Cr	Cu	Ni	Pb	zn	Hg	Fe	Al
ADD_{ing}	7.76E−06	6.30E−07	6.16E−05	3.65E−05	8.43E−05	6.21E−06	4.22E−05	5.50E−08	1.21E−02	1.21E−02
ADD_{inhal}	1.14E−09	9.27E−11	9.05E−09	5.37E−09	1.24E−08	9.13E−10	6.20E−09	8.08E−12	1.77E−06	1.77E−06
ADD_{derm}	2.35E−08	1.91E−09	1.86E−07	1.10E−07	2.55E−07	1.88E−08	1.28E−07	1.66E−10	3.65E−05	3.64E−05
Children										
ADD_{ing}	9.06E−05	7.35E−06	7.18E−04	4.26E−04	9.83E−04	7.25E−05	4.92E−04	6.41E−07	1.41E−01	1.41E−01
ADD_{inhal}	2.53E−09	2.05E−10	2.01E−08	1.19E−08	2.75E−08	2.02E−09	1.38E−08	1.79E−11	3.93E−06	3.93E−06
ADD_{derm}	1.45E−08	1.18E−09	1.15E−07	6.82E−08	1.57E−07	1.16E−08	7.88E−08	1.03E−10	2.25E−05	2.25E−05

Table 2. Noncarcinogenic and carcinogenic risks of heavy metals in the sediment samples from the Aras River

Adults	As	Mo	Cr	Cu	Ni	Pb	Zn	Hg	Fe	Al	ΣHI
HQ_{ing}	2.59E−02	1.26E−04	2.05E−02	9.13E−04	4.21E−03	1.77E−03	1.41E−04	1.83E−04	1.72E−02	1.21E−02	
HQ_{inhal}	3.81E−06	0.00E+00	3.16E−04	1.19E−07	4.96E−07	2.61E−07	1.77E−08	8.26E−07	2.22E−06	1.27E−03	
HQ_{derm}	1.91E−04	1.00E−06	3.10E−03	9.21E−06	4.72E−05	3.61E−05	2.13E−06	7.92E−06	5.21E−05	3.64E−04	
HI_{total}	2.61E−02	1.27E−04	2.39E−02	9.23E−04	4.26E−03	1.81E−03	1.43E−04	1.92E−04	1.73E−02	1.37E−02	8.84E−02
CR_{ing}	1.16E−05	-	1.85E−07	-	0.00E+00	2.17E−08	-	-	-	-	
CR_{inhal}	1.72E−08	-	2.53E−13	-	1.04E−08	2.92E−12	-	-	-	-	
CR_{derm}	8.59E−08	-	1.12E−11	-	0.00E+00	9.77E−12	-	-	-	-	
TCR	1.17E−05		1.85E−07		1.04E−08	2.17E−08					
Child											
HQ_{ing}	3.02E−01	1.47E−03	2.39E−01	1.07E−02	4.92E−02	2.07E−02	1.64E−03	2.14E−03	2.01E−01	1.41E−01	
HQ_{inhal}	8.43E−06	0.00E+00	7.17E−04	0.00E+00	1.37E−06	6.33E−07	3.93E−08	2.11E−07	4.91E−06	2.81E−03	
HQ_{derm}	1.18E−04	6.19E−07	1.91E−03	5.68E−06	2.91E−05	2.23E−05	1.31E−06	4.89E−06	3.22E−05	2.25E−04	
HI_{total}	3.02E−01	1.47E−03	2.42E−01	1.07E−02	4.92E−02	2.07E−02	1.64E−03	2.14E−03	2.01E−01	1.44E−01	9.75E−01
CR_{ing}	1.36E−04	-	2.15E−06		0.00E+00	2.54E−07	-	-	-	-	
CR_{inhal}	3.82E−08	-	5.62E−13		2.31E−08	6.48E−12	-	-	-	-	
CR_{derm}	5.30E−08	-	6.89E−12		0.00E+00	6.03E−12	-	-	-	-	
TCR	1.36E−04		2.15E−06		2.31E−08	2.54E−07					

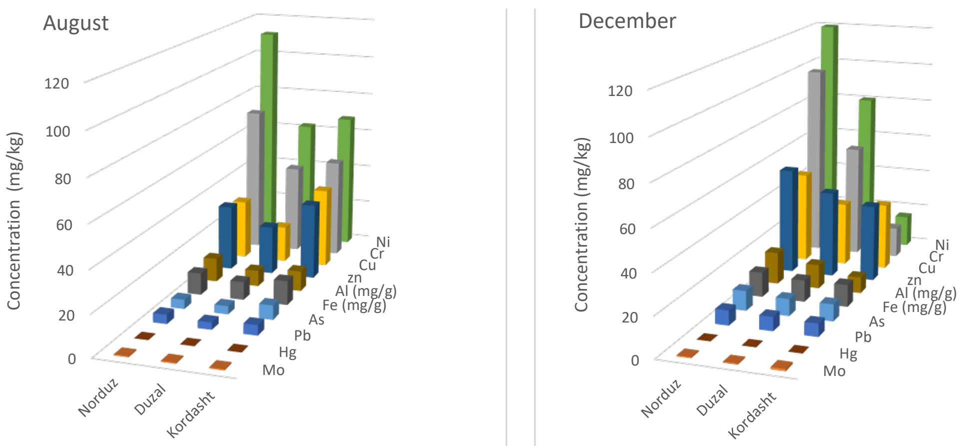

Figure 2. Trend of heavy metal concentration (mg/kg) in the Aras River sediment samples in August and December

and 83% of the samples, respectively, and were below the PEL in 50% and 17% of the samples, respectively. Nickel concentrations exceeded the PEL in 84% of the samples, but were within the TEL and PEL ranges in 16% of the samples. On the other hand, concentrations of As, Pb, and Zn in all samples were below the TEL values Figure 3.

Spatial distribution of heavy metals in the Aras River

The results of the correlation analysis of heavy metals in the sediment of the Aras River are presented in Table 3. PCA is a widely used method for examining the sources of metals in the environment, identifying anthropogenic influences, and determining the parent materials of soil. Table 4 presents the results of PCA before and after rotation, with the latter offering a more accurate representation of empirical realities. The rotation method employed was variance maximization, selecting factors with eigenvalues greater than 1. From the principal component analysis depicted in Table 4, two principal components with eigenvalues exceeding 1.0 were extracted, which explained nearly 89.7% of the data variability. The loading plots for the initial two PCs are shown in Figure 4. PC1 accounts for 50.08% of the cumulative variance. PC1 exhibits strong positive loading on As (0.974), Zn (0.958), Pb (0.949), and Cu (0.910), along with a moderately positive loading on Ni (0.635).

Discussion

Heavy metals enter the human body through various routes, including ingestion, dermal contact, and inhalation. Dermal contact is a significant route of exposure to trace metals in river and coastal sediments. Studies have found that children playing in tide flats can accumulate significant sediment loads on their

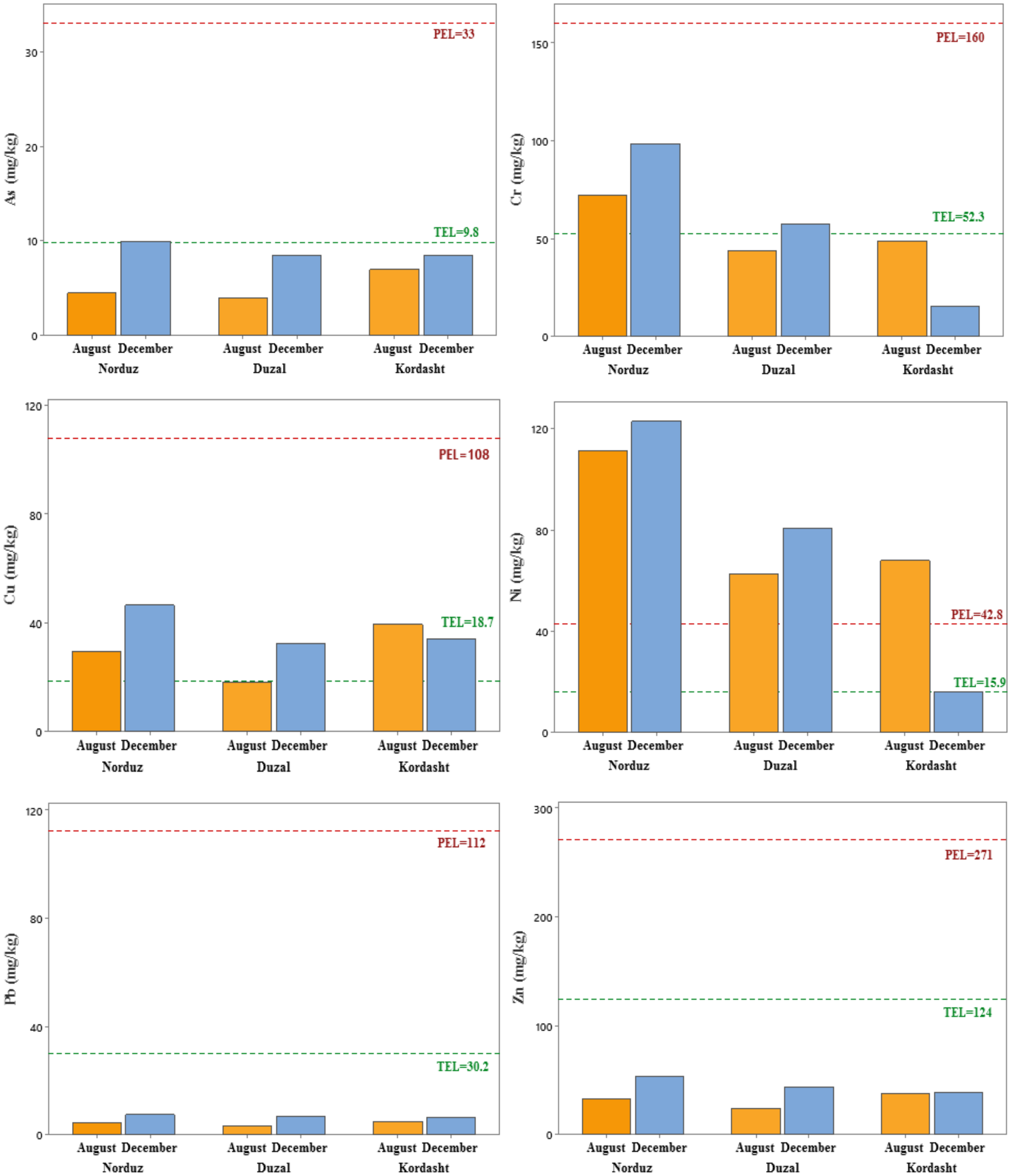

Figure 3. Metal concentrations in sediments of the Aras River in August and December, and the comparisons with the thresholds of sediment quality guidelines (TEL and PEL)

skin, particularly on hands and feet (38-40). Inhalation exposure to river sediments can occur through various pathways, particularly in areas with significant heavy metal contamination. This exposure is concerning due to the potential health risks posed by inhaling dust or aerosols containing toxic metals. Sediments can become airborne during dry conditions or construction activities, leading to the inhalation of contaminated particles.

Additionally, contaminated sediments can be dispersed into the air during rain events, increasing the likelihood of inhalation exposure for nearby populations (29,41,42). In this study, ingestion, inhalation, and dermal contact were considered pathways for metal intake. The estimated ADD from dermal contact for all studied heavy metals

Table 3. Pearson correlation coefficients for heavy metals in sediment Correlation

	As	Mo	Cr	Cu	Ni	Pb	Zn	Hg	Fe
Mo	0.290								
Cr	0.190	−0.712							
Cu	0.823	0.034	0.453						
Ni	0.020	−0.779	0.978	0.324					
Pb	0.965	0.260	0.262	0.757	0.118				
Zn	0.935	0.024	0.512	0.897	0.366	0.941			
Hg	−0.043	0.118	−0.565	−0.394	−0.499	0.009	−0.228		
Fe	0.793	0.051	0.424	0.992	0.314	0.740	0.872	−0.351	
Al	0.568	−0.448	0.908	0.708	0.831	0.640	0.818	−0.452	0.683

*Indicates that correlation is significant at the 0.05 level (2-tailed);

Table 4. Matrix of principal components analysis

Heavy metals	Component		Rotated component	
	PC1	PC2	PC1	PC2
Al	0.953	−0.267	0.635	0.759
Zn	0.934	0.332	0.958	0.254
Cu	0.884	0.321	0.910	0.235
Fe	0.864	0.324	0.896	0.222
Pb	0.778	0.543	0.949	
Cr	0.764	−0.636	0.271	0.957
As	0.750	0.630	0.974	
Hg	−0.626	0.541	−0.210	−0.800
Mo	−0.236	0.830	0.275	−0.818
Ni	0.656	−0.752	0.116	0.991
% of Variance	59.373	30.334	50.081	39.626
Cumulative %	59.373	89.707	50.081	89.707

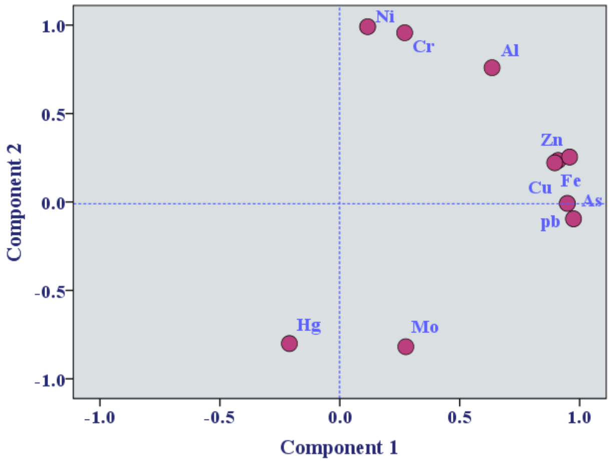

Figure 4. The loading plots of the first two PCs

remained below the standard guideline values (Table 1).

The study results reveal that, for all heavy metals except Al, HQ values were highest for ingestion, followed by dermal contact and inhalation, across age groups (children and adults). This pattern aligns with previous research indicating that ingestion is the primary exposure pathway for heavy metals (19,28). At Norduz, HQ values for children were notably higher for ingestion than for inhalation or dermal contact, with an average exceeding the safe threshold by more than 1. This suggests that children residing near the Aras River may be at risk of experiencing adverse noncarcinogenic health effects.

The HI values for all heavy metals examined through ingestion, inhalation, and dermal contact routes for both adults and children were found to be below the safe level of 1. The total HI value for all metals combined was below 1, suggesting no chronic risk to residents from heavy metals in regional sediment. However, the hazard index values for children were slightly lower than 1, indicating a potential non-cancer risk in the future. Children may be more vulnerable to ingesting small particles because of their lower body weight than adults, which could significantly impact their health (43-45). However, for children, the HI values of As, Cr, Fe, and Al were greater than 0.1, so it is necessary to pay attention to children's health, and especially to guide children to develop good health habits, which can effectively reduce the risk of the ingestion pathway (Table 2) .

Evidently, for both children and adults, the cancer risk (CR) values for As, Cr, Ni, and Pb still indicate that ingestion poses a higher risk than dermal exposure and inhalation. The CR values for As, Cr, Ni, and Pb for adults and children were below 1×10^{-6} via ingestion, inhalation, and dermal exposure, indicating negligible carcinogenic risk. The negligible carcinogenic risk refers to a risk level below established regulatory thresholds, typically considered low enough to pose minimal concern for public health (46). However, the CRing values for As in adults and children were within the acceptable range (1×10^{-6} to 1×10^{-4}). The total CR values for As in adults and children were below the 1×10^{-4} threshold, indicating an acceptable cancer risk from As in the sediment samples. Additionally, the total CR values for Cr, Ni, and Pb in adults and children were below the 1×10^{-6} threshold, suggesting negligible cancer risk from these metals in the sediment samples (Table 3).

The average concentrations of As, Mo, Cr, Cu, Pb, Zn, Fe, and Al in the sediment of the Aras River in December were higher than in August (Fig. 1). This difference could be attributed to factors such as the discharge of tailing dam effluents or the effects of rainfall, dilution, and washing of sediments. It is worth noting that the concentrations of all heavy metals in the Aras River sediment at all stations in August and December were below the average concentrations in Earth's crust, except for Cr, Cu, and Ni. The concentrations of Cr and Cu in sediments at the Norduz station in December were found to be 98.5 mg/kg and 46.5 mg/kg, respectively, which are higher than the average concentrations of these elements in Earth's crust (90 mg/kg for Cr and 45 mg/kg for Cu). Additionally, the concentration of Ni in August at Norduz (111.5 mg/kg) and in December at both Norduz (123 mg/kg) and Duzal (80 mg/kg) were all higher than the average concentration of Ni in Earth's crust (68 mg/kg).

The elevated concentrations of Cr, Ni, and Al at the Norduz station in August suggest that these elements may be entering the river from unidentified upstream sources. Additionally, the higher levels of Cr, Ni, and Al at the Kordasht station compared to Duzal may be attributed to the discharge of tailing-dam effluents into the Aras River. The elevated concentrations of As, Cu, Pb, Hg, and Fe at the Kordasht station in August, following the discharge of tailings dam effluents, exceed those at the other two stations, indicating a direct impact on the Aras River. Additionally, the increasing trend of Mo, Cu, Hg, and Fe concentrations at Kordasht compared to Duzal in December further supports the influence of tailing dams on the metal levels in the Aras River sediment. In this regard, the ecosystem and population around the Aras River are exposed to heavy metals and to water, soil, and agricultural products that are truly polluted (15).

The SQG showed that the sediments had no or low ecological risk in relation to As (100% below TEL), Pb (100% below TEL), and Zn (100% below TEL), moderate ecological risk for Cu (83% above TEL) and Cr (50% above TEL), and high ecological risk from Ni (84% above PEL)(Figure 4). The analysis of sediment samples shows a significant increase in heavy metals at the Duzal and Kordasht stations, likely due to the discharge of mineral waste from the Agarak mine's tailing dams. A 2012 report published by the Georgian Institute of Politics noted that one factor contributing to the pollution of the Aras River with metals is the presence of mining activities near the Kura–Aras River (47). In the study by Ghazaryan et al the soil surrounding Agark's copper–molybdenum mining complex (including the open mine, processing plant, and two recultivated tailing dumps) was compared to a control site. The findings indicate significant soil pollution in these areas (48). In Aazami et al's study, the poor water quality of the Aras River was attributed to wastewater discharge from copper mine excavation, which is a significant source of pollution along the Iranian-Armenian border (49). Given that Aras River water is used to irrigate crops and the region's residents consume these products, it is crucial to assess levels of Cu, Pb, As, and Hg in soil and agricultural products to evaluate the extent of environmental damage. Consequent negative impacts of heavy metals on the ecological environment of the Aras River have also been documented. Environmental pollution with heavy metals has posed significant human health risks in recent years (50). In the Aras River basin, numerous reports of severe health impacts have raised questions about environmental safety and drinking water quality (51).

A correlation value closer to 1 indicates a higher likelihood that the two variables share the same source (52). The *p*-value represents the probability of sampling error; values < 0.05 indicate a significant correlation between the two variables. The correlation analysis revealed a strong positive correlation between the concentrations of As and Cu, Pb, Zn, and Fe; Cr and Ni and Al; Cu and Pb, Zn, Fe, and Al; Ni and Al; Pb and Zn, Fe; Zn and Fe and Al; as well as Mo and Cr and Ni (Table 3). Correlation values exceeding 0.7 suggest a common source or shared distribution of these heavy metals. Conversely, Hg showed no significant correlations with other metals, except for Cr, suggesting a distinct local source. The correlation coefficients between Cr, Zn, and Hg ranged from 0.5 to 0.6, suggesting a moderate correlation and hinting at some familiar sources. Additionally, weak correlations ($P < 0.5$) among other indices indicate a complex origin of heavy metals.

The PCA results are consistent with those of Pearson's correlation analysis (Table 4). PC1 probably reflects anthropogenic sources such as mining activities (mine drainage, tailings, and mining) and vehicle emissions. Upstream of the Duzal and Kordasht stations, there are tailing dams at a copper mine that discharge metallic wastewater containing high levels of heavy metals into the Aras River. This information provides valuable context for understanding the sources and composition of the pollutants identified in PC1 (49,53). The results of the Aras River water sample from the Kordasht station show that rainfall leaching heavy metals in the river exceeded the heavy metal concentration in the downstream region (18). The PC2 accounts for 39.62% of the cumulative variance, with high loadings for Ni (0.991), Cr (0.957), Hg (-0.800), Mo (-0.818), and Al (0.759). PC2 is likely affected by typical anthropogenic sources, including agricultural practices, industrial activities, urban wastewater, and fish farming. This is likely due to the low levels of Hg present in the environment and the fact that Ni, Mo, Cr, and Al are essential components of chemical fertilizers and insecticides (54,55).

The main limitations of this study were as follows: 1) Due to the exploratory nature of this investigation, as a preliminary endeavor within a broader study scope encompassing this river, the sample sizes were intentionally kept minimal. 2) It was not feasible to conduct the Kaiser-Meyer-Olkin (KMO) measure and Bartlett's test of sphericity, both of which serve as indicators of the appropriateness of applying principal component analysis (PCA) for data reliability. However, given the imperative to gain initial insights into the sources of heavy metal pollution in this river, PCA was cautiously executed. 3) The current study did not encompass the assessment of heavy metal concentrations in irrigation water or soil, nor did it explore their bioaccumulation in crops. Furthermore, the potential interconnections among these variables remained unexamined.

Conclusion

The human health risk assessment of heavy metal exposure in sediment from the Aras River indicated that daily intake estimates for heavy metals were below the EPA guideline levels. However, children had a higher intake per kilogram of body weight than adults.

Noncarcinogenic risk assessments indicated low overall risks, while carcinogenic risk assessments showed negligible risks for most metals. The SQG analysis displayed moderate to high ecological risks for some metals. Analysis using correlation and PCA highlighted unique local sources of mercury (Hg) and its minimal correlations with other metals. Anthropogenic sources, such as mining activities, were identified as significant contributors. These findings provide crucial insights into pollutant origins and underscore the impact of human activities on heavy metal distribution in the Aras River, emphasizing the need for ongoing research and monitoring to address contamination and mitigate associated risks to ecosystems and human health.

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Competing interests

The authors declare that there are no competing interests.

Ethical issues

The authors confirm that all data gathered during the study are accurately presented in the manuscript, and no data from this research will be published separately or elsewhere.

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Supplementary file

Supplementary File 1 contains Tables S1 and S2.

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