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Water quality assessment, possible origins and health risks of toxic metal (loid)s in five cascade reservoirs in the upper Mekong

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ABSTRACT

The Mekong River holds significant importance as a transnational water system within the Asian region. This study investigated the pollution characteristics, origins, and health risks associated with eighteen toxic metal (loid)s (TMs) across various depths in five cascade reservoirs located in the upper Mekong. The findings revealed that naturally sourced TMs (As, Cd, Li, Mo, Sb, and Sr) exhibited elevated levels in upstream reservoirs due to the interception effects of cascade dams. Anthropogenically sourced TMs (Co, Cu, Ni, Pb, and Zn) showed heightened concentrations in downstream reservoirs, linked to increased human activities in the downstream region. In deep-water reservoirs, the vertical distribution of redox-sensitive elements was influenced by thermal stratification, with concentrations of As, Cs, Li, Mo, Rb, Sr, and U notably higher in the hypolimnion than the epilimnion. Importantly, the concentrations of all elements in these cascade reservoirs meet Chinese and WHO drinking water standards. Water quality index (WQI: 9.2–19.1), heavy metal evaluation index (HEI: 0.96–1.37), and Nemerow index (NI: 0.35–0.56) confirmed the commendable quality of the cascade reservoir waters. The health risk assessment identified As, Sb, U, and Tl as primary contributors to non-carcinogenic risks in the cascade reservoirs. In the context of a series of cascade dams along this substantial river, this research establishes a solid foundation for preventing TM pollution and ensuring the long-term sustainability of water resources.

1. Introduction

Water pollution from toxic metal (loid)s (TMs) has become a focal point of concern in recent years due to their high toxicity, non-biodegradability, and potential threats to water resources essential for consumption, irrigation, and recreation (Chen et al., 2023; Tang et al., 2022; Yang et al., 2023; Zeng et al., 2021; Zhang et al., 2021b). While both natural and anthropogenic processes contribute to TM pollution in water bodies, elevated TM concentrations are often linked to anthropogenic activities such as agricultural runoff, industrial sewage, and domestic wastewater (Mokarram et al., 2020; Zhang et al., 2021a). The presence of high TM concentrations in water poses severe risks to human and aquatic health (Adimalla, 2020; Muhammad and Ahmad, 2020; Tokatlı and Varol, 2021). For example, chronic exposure to arsenic in water is associated with various cancers, diabetes, skin lesions, and other chronic diseases (Abdul et al., 2015; Naujokas et al., 2013). Similarly, chronic exposure to cadmium in water can cause osteoporosis, lung cancer, and kidney disease (Li et al., 2023a).

Rivers play a pivotal role in the global material cycle, influencing the health of river ecosystems. Understanding the input and discharge status, content characteristics, and cycling processes of TMs is crucial for comprehending river ecosystem dynamics (Wu et al., 2021a; Xiang et al., 2021). However, the growing demand for water resources and energy in socio-economic development has led to widespread cascade damming, disrupting river continuity and altering hydrodynamic conditions, thereby affecting TM transport and geochemical cycling processes (Li et al., 2020; Yin et al., 2022; Zhao et al., 2020, 2022). The stratification of temperature and dissolved oxygen (DO) in overlying

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water, induced by large reservoirs, inhibits mixing and material exchange between surface and deep waters. This prolonged reduced state at the sediment-water interface diminishes sediment adsorption (Krueger et al., 2020; Wang et al., 2016). Consequently, significant quantities of TMs may be released from sediment into overlying water, posing adverse ecological and environmental effects (Gao et al., 2018; Muhammad, 2022; Vink et al., 2017).

Monitoring TMs in freshwater resources is of paramount importance, with various heavy metal risk assessment indices developed globally (Tokatli et al., 2021; Xiang et al., 2021; Zhang et al., 2022). These indices, such as the water quality index (WQI), Nemerow index (NI), heavy metal evaluation index (HEI), and health risk assessment indices, play a crucial role in determining the potential hazards of toxicants in freshwater habitats (Varol and Tokatlı, 2023, Zhang et al., 2021a; Tokatli et al., 2021). They are effective tools for evaluating the synergistic effects of multiple contaminants on ecosystems and human health. Furthermore, applying multivariate statistical techniques serves as a valuable approach to comprehending the mechanisms influencing surface water quality (Wang et al., 2021). Techniques such as correlation analysis (CA) and principal component analysis (PCA) are commonly employed to identify probable pollution sources and provide insights into qualitative data (Haq et al., 2023). These ecotoxicological and statistical assessments contribute to simplifying complex and extensive datasets, facilitating their interpretation for management, decision-making, and communication strategies (Tokatlı et al., 2023).

The Mekong River holds significant importance as a transnational water system within Asia, supporting diverse ecosystems and providing essential freshwater for local communities and economies (Wen et al., 2022). The construction of cascade dams in the upper Mekong (Lancang River), driven by the increasing demand for power and water resources, has led to significant hydrodynamic changes (Cheng et al., 2022). Effective use and management of water resources in the upper Mekong, considering the concerns of governments in China and beyond, are crucial. The rapid industrial, agricultural, and urbanization advancements in recent years raise potential threats to the water environment (Gui et al., 2021; Guo et al., 2020). Reports of metal pollution in natural biofilms and suspended particulate matter in the upper Mekong underscore these concerns (Wen et al., 2022; Zeng et al., 2022). Previous studies have revealed elevated concentrations of As, Cs, Li, Mo, Sr, and U in the upper Lancang River, linked to geothermal spring input. Additionally, mining activities at the Lancang Lead Mine have been associated with increased levels of Cd, Cu, Pb, and Zn in the basin's waters, along with abnormally high Sb concentrations influenced by the nearby Gepoluo Antimony Mine (Zhao et al., 2023). Given the role of reservoirs as key sites for pollutant accumulation, it is imperative to identify the distribution, origins, and health risks associated with TMs (Gao et al., 2022; Varol, 2020). Understanding the influencing factors and assessing the water quality in these cascade reservoirs are essential for effective water resource management.

Therefore, this research aimed to (i) analyze the spatial distribution of 18 TMs in the overlying water of cascaded reservoirs in the upper Mekong, (ii) identify their potential sources, (iii) evaluate overall water quality, and (iv) assess associated health risks. The findings contribute valuable insights for assessing water environments and ensuring the sustainable utilization of water resources in the operational state of cascade reservoirs in the upper Mekong.

2. Materials and methods

2.1. Study area

The upper Mekong, also known as the Lancang River, holds international importance as a vital resource for multiple Southeast Asian countries, contributing to their economic, agricultural, and industrial development. Possible pollution sources of TMs in this river include both natural processes and anthropogenic activities (Wen et al., 2022). Industrial discharges, agricultural runoff, and mining operations are potential contributors to TM contamination (Wang et al., 2012). Additionally, the river's environmental sensitivity makes it susceptible to pollution from various sources (Gui et al., 2021). The population density along the river is notable, particularly in downstream areas, which may lead to increased anthropogenic activities and associated environmental impacts, including potential TM pollution (Zeng et al., 2022). The economic structure of the Lancang River region is diverse, encompassing sectors such as agriculture, industry, and trade. The agricultural potential of the Lancang River basin is significant, with the river water used for irrigation and sustaining agricultural practices. This underscores the importance of maintaining water quality to ensure the productivity and sustainability of agriculture in the region. Industrial potential along the river is also noteworthy, with various sectors contributing to economic growth. However, industrial activities can pose challenges, including the potential release of heavy metals into the river, necessitating careful management to mitigate environmental risks (Song et al., 2013; Zhang et al., 2014).

Abundant hydraulic resources in the upper Mekong, characterized by heavy runoff and a substantial descending elevation, create a favorable environment for the development of cascade hydropower stations (Zhao et al., 2022). The study focuses on five reservoirs located in Yunnan Province—Xiaowan (XW), Manwan (MW), Dachaoshan (DCS), Nuozhadu (NZD), and Jinghong (JH) Reservoirs (Fig. 1). Further information about these reservoirs is provided in Table S1.

2.2. Sample collection and analytical methods

Overlying waters at various depths of the reservoirs were collected using a Niskin Water Sampler (General Oceanics, USA) in April 2017 (Fig. 1). In-situ measurements of electrical conductivity (EC), temperature (T), DO, pH, and total dissolved solids (TDS) were conducted using an automated multiparameter profiler (YSI 6600V2, YSI Ltd., USA). After filtration through 0.45 μm membranes, samples were stored in 50 ml polypropylene bottles. Two filtered waters were collected from each site: one was acidified to 2 % HNO_3 for the detection of TMs and cations, and the other for anion measurement. All samples were kept at 4 °C until analysis. Major cations and anions were detected by ICP-OES (iCAP6500) and ion chromatography (ICS-90, Dionex), respectively. TM analysis was performed using ICP-MS (7700 × , Agilent). The measured values of TMs in the external standard reference solution consistently fell within the range of certified values.

2.3. Indices for water quality

The Nemerow index (NI), heavy metal evaluation index (HEI), and water quality index (WQI) were conducted to assess the water quality status of the five cascade reservoirs, with corresponding details presented in Text S1–S3 (Adimalla and Qian, 2019; Gao et al., 2019; Muhammad and Ullah, 2022; Şener et al., 2017).

2.4. Health risk assessment

Hazard quotient (HQ) and Hazard index (HI) were used to assess the human health risks of TMs in water from these cascade reservoirs (Shil and Singh, 2019; Xiao et al., 2019). These two indices can reflect non-carcinogenic hazards caused by metal (loid)s (Varol et al., 2021). Their calculation formulas were as follows:

$$HQ_i = \frac{ADD_i}{RfD_i}$$

$$HI = \sum \left(HQ_{ingestion} + HQ_{dermal}\right)$$

where ADD_i is the mean daily consumption of metal (loid) via oral ingestion and skin contact. Its detailed calculations can be found in the

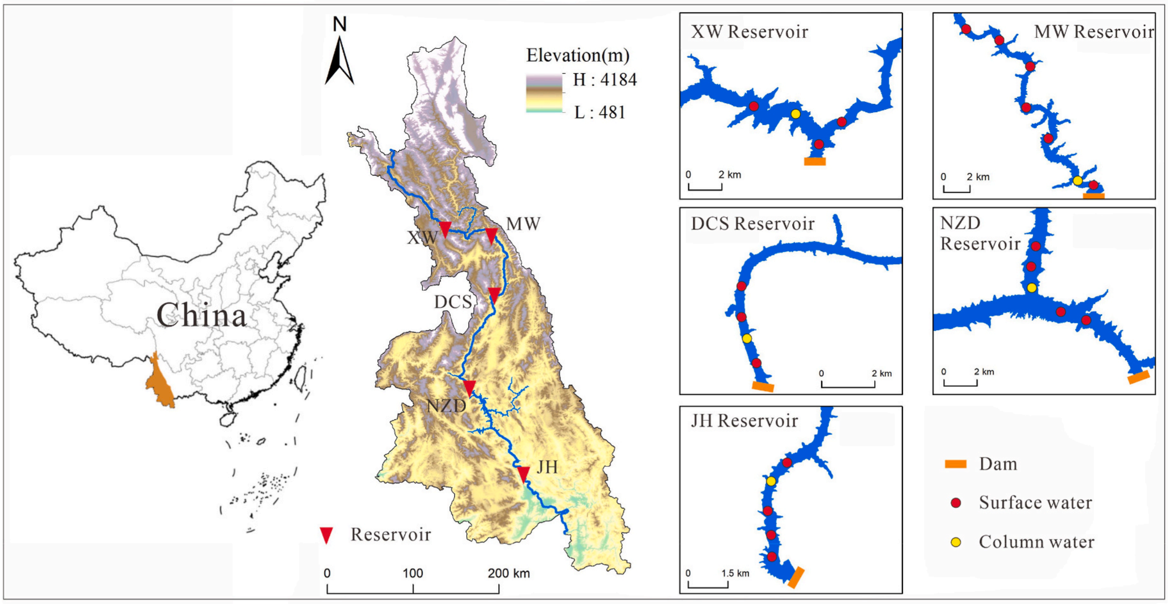

Fig. 1. Locations of the sampling sites.

Supplementary Material (Text S4). RfD_i refers to the reference dose (Zhao et al., 2023). A HQ or HI value greater than 1 suggests potential adverse health effects, while a value below 1 indicates no significant health concerns. (Islam et al., 2020).

3. Results and discussion

3.1. Spatial profiles of TMs in surface water

The surface water of the five cascade reservoirs showed a slightly alkaline nature, with mean pH values ranging from 7.60 to 8.47, and the JH Reservoir exhibiting the lowest pH (Fig. 2). These reservoirs displayed high levels of dissolved oxygen (DO), with the MW Reservoir recording the highest value (9.99 mg/L) and the JH Reservoir the lowest (7.86 mg/L). Average EC and TDS values ranged from 396 to 537 μS/cm and from 258 to 346 mg/L, respectively, with the MW Reservoir registering the highest values and the NZD Reservoir the lowest.

The major ions in these reservoirs exhibited a decreasing order as follows: $SO_4^{2-} > Ca^{2+} > Na^{+} > Cl^{-} > Mg^{2+} > K^{+} > NO_3^{-} > F^{-}$ (Fig. 2). With the exception of K^+, the NZD Reservoir consistently had the lowest concentrations of all ions. The spatial distribution pattern of most ions, including SO_4^{2-}, Cl^-, Ca^{2+}, Mg^{2+}, and Na^+, followed a consistent order across the cascade reservoirs: the MW and DCS Reservoirs had higher concentrations than the XW and JH Reservoirs, which, in turn, had higher concentrations than the NZD Reservoir. This pattern aligned with the distribution of EC and TDS.

The total concentrations of 18 TMs in the surface water of the five cascade reservoirs in the upper Mekong ranged from 241.1 to 483.6 μg/L. Three distinct distribution patterns of TMs were observed in the surface water of these cascade reservoirs (Fig. 3). The first pattern included As, Cd, Li, Mo, Sb, and Sr, with concentrations decreasing in the order of the MW and DCS Reservoirs > the XW Reservoir > the NZD and JH Reservoirs. In the second distribution pattern, Co, Cu, Ni, Pb, and Zn showed higher concentrations in downstream reservoirs compared to upstream reservoirs. The remaining elements (Ba, Cr, Cs, Rb, Tl, U, and V) constituted the third distribution pattern, with no discernible

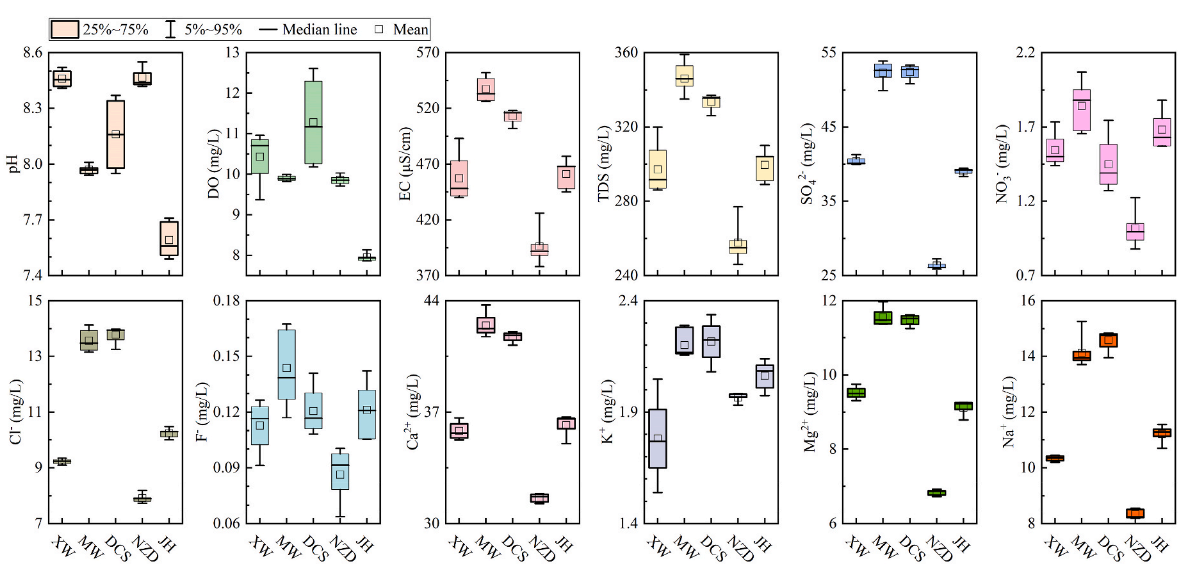

Fig. 2. Spatial profiles of physicochemical parameters of surface water from five cascaded reservoirs in the upper Mekong River.

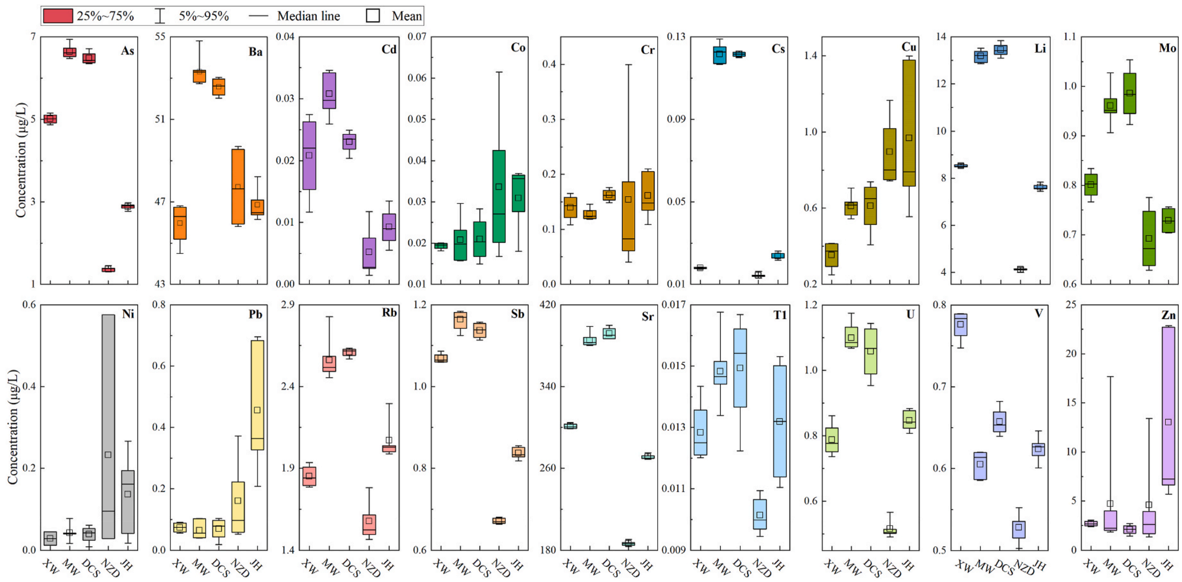

Fig. 3. Spatial profiles of 18 TMs of surface water from five cascaded reservoirs in the upper Mekong River.

regularity observed among these reservoirs.

The average concentrations of TMs in the studied reservoirs were compared with levels reported in other reservoirs (Table S4). Notably, Cd (0.01–0.03 μg/L), Cr (0.14–0.36 μg/L), Ni (0.03–0.23 μg/L), and V (0.53–0.78 μg/L) in the cascade reservoirs were lower than those found in various global counterparts such as the Keban Dam Reservoir in Turkey (Cr 0.63 μg/L, Ni 1.62 μg/L, V 2.74 μg/L) (Canpolat et al., 2020), the Danjiangkou Reservoir in China (Cd 0.30 μg/L, Cr 1.77 μg/L, Ni 2.05 μg/L) (Hao et al., 2021), the Three Gorges Reservoir in China (Cd 0.77 μg/L) (Gao et al., 2016), and reservoirs in the hilly area of southern China (Cr 1.84 μg/L, Ni 0.54 μg/L) (Wang et al., 2018). However, As (5.0–6.6 μg/L) in the XW, MW, and DCS Reservoirs surpassed levels reported in the literature. Similarly, Ba (46.0–53.3 μg/L) concentrations in the cascade reservoirs exceeded those observed in the Keban Dam Reservoir in Turkey (32.4 μg/L). Cu (0.35–0.61 μg/L) in the XW, MW, and DCS Reservoirs was lower than the reported levels in other reservoirs, though comparable in the NZD and JH Reservoirs. Furthermore, Pb (0.40 μg/L) and Zn (10.6 μg/L) in the JH Reservoir were higher than those documented in the Keban Dam Reservoir in Turkey (Pb 0.07 μg/L, Zn 5.2 μg/L), whereas the upper reservoirs did not exhibit elevated

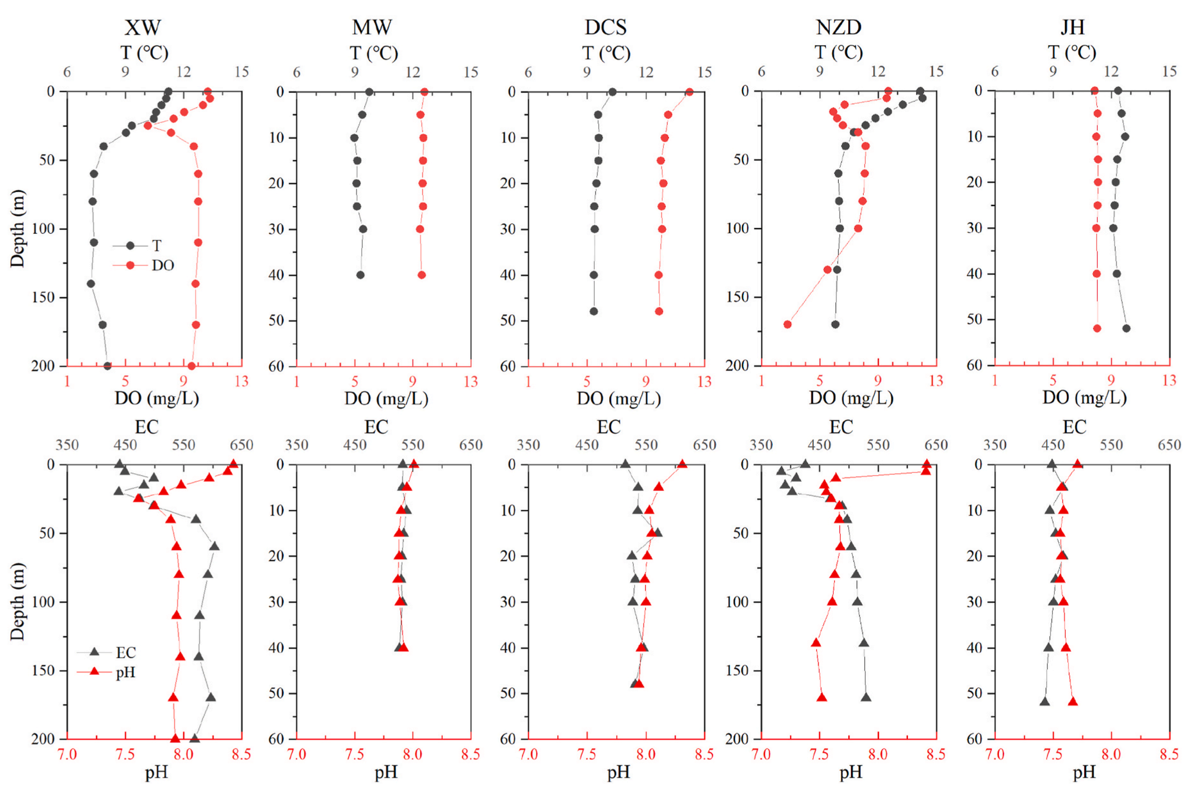

Fig. 4. Depth profiles of water temperature (T), dissolved oxygen (DO), electrical conductivity (EC), and pH in the water column of the five cascaded reservoirs in the upper Mekong River.

concentrations in comparison.

3.2. Vertical profiles of TMs in water column

Distinct chemical stratifications were evident in deep-water reservoirs (Fig. 4). In the XW Reservoir, temperature stratification led to the formation of an epilimnion (0–40 m) and hypolimnion (40–200 m). Within the epilimnion, the DO and pH values progressively decreased with depth, reaching a minimum value at 25 m, and then increased. This pattern suggests intense microbial activity at this depth, impacting DO and pH near this layer. Similarly, in the NZD Reservoir, the presence of an epilimnion (0–40 m) and hypolimnion (40–170 m) due to temperature stratification was observed. In the epilimnion, DO concentrations and pH values decreased with depth, reaching a minimum value at 15 m, and then increased, mirroring the pattern in the XW Reservoir. Conversely, DO concentrations and pH values stabilized in the hypolimnion of the XW Reservoir, while gradually decreasing with depth in the hypolimnion of the NZD Reservoir. The EC value, reflecting the concentration of soluble salts, varied at different depths of the water columns under the influence of these chemical stratifications. No obvious chemical stratification was found in shallow reservoirs (the MW, DCS, and JH Reservoirs).

In deep-water reservoirs, the vertical distribution of redox-sensitive elements was influenced by the thermal stratification of the water column (Fig. 5). Specifically, both the XW and NZD Reservoirs showed significantly higher levels of As, Cs, Li, Mo, Rb, Sr, and U in the hypolimnion compared to the epilimnion. In the XW Reservoir, concentrations in the epilimnion were 6.23, 0.05, 9.12, 0.80, 1.97, 325.0, and 0.80 μg/L, respectively, while in the hypolimnion, they were 7.52, 0.32, 18.13, 1.15, 3.19, 484.4, and 1.33 μg/L, respectively. Similarly, in the NZD Reservoir, concentrations in the epilimnion were 2.14, 0.02, 5.96, 0.63, 1.70, 217.2, and 0.60 μg/L, respectively, while in the hypolimnion, they were 4.12, 0.04, 11.12, 0.87, 2.46, 343.4, and 1.00 μg/L, respectively. Notably, Cu exhibited a depth-dependent decrease in concentration in the NZD Reservoir, potentially influenced by exogenous Cu input from intense anthropogenic activities near the reservoir (Zhao

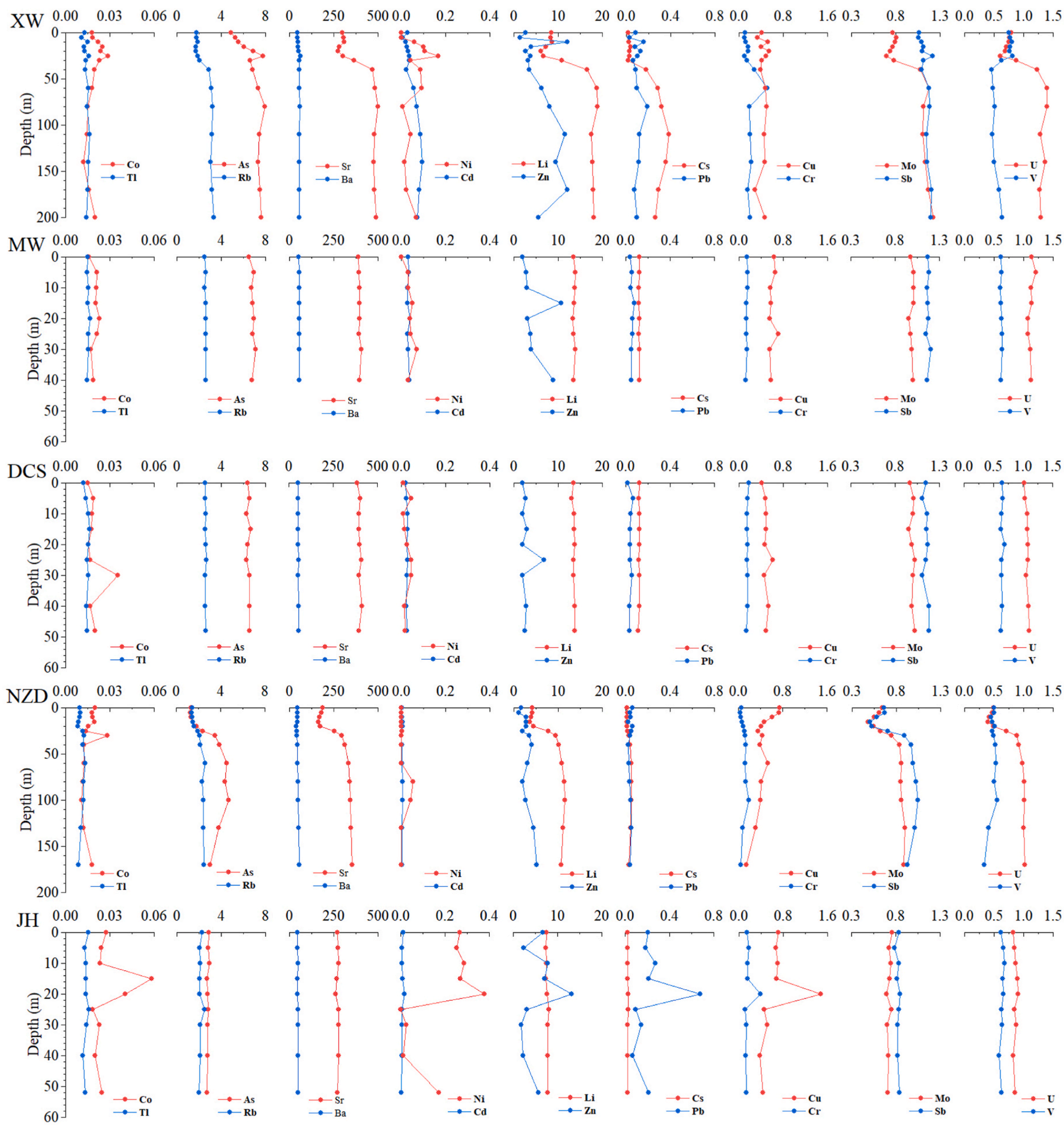

Fig. 5. Depth profiles of 18 TMs in the water column of the five cascaded reservoirs in the upper Mekong River.

et al., 2023). These findings underscore the importance of considering varying water depths for comprehensive water quality assessment and health risk evaluation associated with TMs.

3.3. Sources recognition of TMs

Correlation analysis (CA) and principal component analysis (PCA) have proven effective for discerning TM sources (Varol et al., 2022).

3.3.1. CA

CA aimed to explore interrelationships among the 18 TMs (Fig. 6). It has been demonstrated that TMs that were highly correlated with each other may share common origins or be influenced by similar controlling factors (Canpolat et al., 2022; Luo et al., 2021). Marked positive correlations were observed among As, Ba, Cd, Cs, Li, Mo, Rb, Sb, Sr, Tl, and U ($0.50 \leq R \leq 0.99$, $p < 0.001$), indicating that they might have a common source. Significant positive correlations were found within Co, Cu, Ni, and Pb ($0.53 \leq R \leq 0.85$, $p < 0.001$), but they were negatively correlated with most other TMs, indicating a distinct origin. Additionally, Zn showed positive correlations with Co, Cu, Ni, and Pb ($p < 0.05$) but lacked significant correlations with other TMs.

3.3.2. PCA

The PCA was conducted to further probe TM origins in these cascade reservoirs (Luo et al., 2021; Zeng et al., 2020). Three distinct principal components (PCs) were identified, elucidating a cumulative variance of 80.26 % (Table 1). The factor loads could be categorized as "high", "moderate", and "low" according to absolute load values exceeding 0.75, 0.75–0.50, and below 0.50 (Varol, 2019).

The PC1 component explaining 48.62 % of the total variance, exhibited high loadings for Sr (0.93), Li (0.91), Mo (0.89), As (0.88), Rb (0.88), U (0.87), Cs (0.86), Sb (0.85), Cd (0.84), Ba (0.83) and Tl (0.77). Recent research indicated that dissolved As, Cs, Li, Mo, Rb, Sr, and U in the Lancang River were affected by geothermal spring input (Zhao et al., 2023). Conversely, the relatively low concentrations of Ba, Cd, Sb, and Tl suggested a potential association with natural sources, such as soil erosion. Notably positive correlations among As, Ba, Cd, Cs, Li, Mo, Rb, Sb, Sr, Tl, and U (Fig. 6) further supported the conclusion that PC1 was linked to natural inputs.

The PC2 component, explaining 21.07 % of the overall variance, displayed high loadings for Cu (0.87), Pb (0.87), Zn (0.76), and moderate loadings for Co (0.68), Ni (0.66), Cr (0.55). Notably, Co, Cu, Ni, and Pb displayed relatively strong correlations, and these four elements were also positively correlated with Zn (Fig. 6). The primary sources of Cu and Zn, such as smelting, plating, and machinery manufacturing, have been documented (Chen et al., 2022; Yuan et al., 2023). Pb primarily originates from traffic emissions, including those from cars and barges (Li et al., 2023b; Yuan et al., 2023). The completion of cascade reservoirs could lead to increased Pb concentrations in water due to the accumulation of ships and exhaust emissions. Previous research identified the Lancang Lead Mine, upstream of the NZD Reservoir, as a contributing factor to elevated contents of Co, Cr, Cu, Ni, Pb, and Zn in the Lancang River (Zhao et al., 2023). Hence, PC2 was attributed to industrial activities and transportation.

Therefore, the heightened concentrations of Co, Cu, Ni, Pb, and Zn identified in the surface water of the downstream reservoirs (Section 3.1) were attributable to increased human activities in the downstream region. Conversely, the first pattern encompassing As, Cd, Cs, Li, Mo, Rb, Sb, Sr, and U could be elucidated by natural inputs. The elevated levels of As, Cd, Li, Mo, Sb, and Sr in upstream reservoirs (Section 3.1) might

Table 1
Factor loadings for varimax rotated PCA of TMs.

	PC1	PC2	PC3
Sr	**0.932**	−0.267	0.188
Li	**0.911**	−0.243	0.291
Mo	**0.895**	−0.212	0.237
As	**0.883**	−0.351	−0.203
Rb	**0.880**	−0.155	0.346
U	**0.874**	−0.116	0.341
Cs	**0.861**	−0.063	0.411
Sb	**0.853**	−0.364	−0.278
Cd	**0.837**	−0.015	0.158
Ba	**0.834**	−0.132	−0.127
Tl	**0.773**	0.038	−0.038
Cu	−0.218	**0.873**	−0.032
Pb	−0.290	**0.871**	0.031
Zn	0.093	**0.761**	0.073
Co	−0.403	**0.677**	−0.175
Ni	−0.347	**0.661**	−0.091
Cr	0.181	0.552	0.398
V	−0.115	−0.025	−0.919
% of variance	48.62	21.07	10.57
% of cumulative	48.62	69.69	80.26

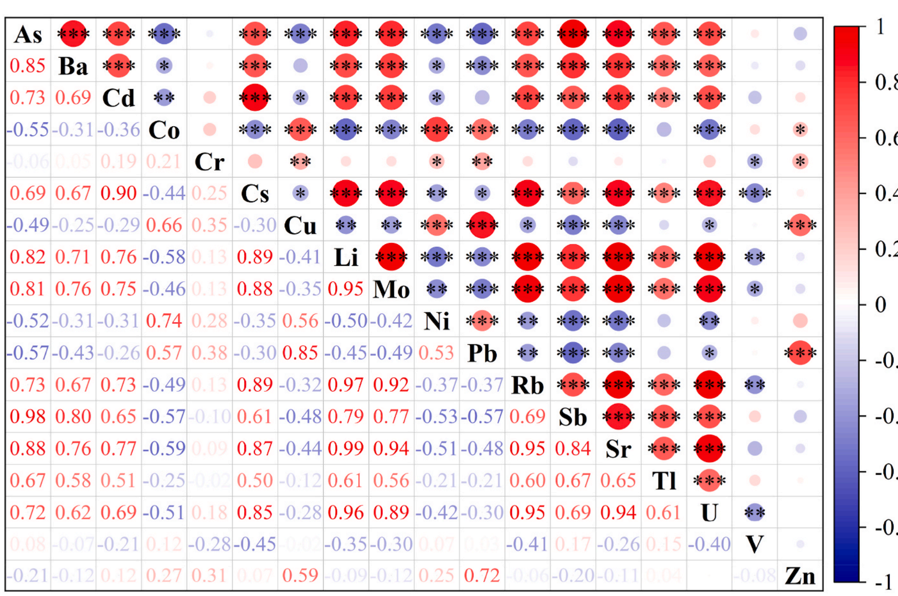

Fig. 6. Correlation coefficients of TMs in the five cascaded reservoirs in the upper Mekong River. *** Significant at the 0.001 level. ** Significant at the 0.01 level. * Significant at the 0.05 level.

be attributed to the interception effects of cascade dams.

3.4. Water quality assessment

A comparison was made between the concentrations of elements in the reservoir water from this study and the established water quality standards (Table S3). The concentrations of all elements (EC, TDS, 8 major ions, and 18 TMs) in the cascade reservoirs complied with the Chinese surface water standard for Grade I (GB 3838–2002), signifying that these reservoirs could be regarded as clean water sources for reserves. Moreover, the concentrations of these elements were below the drinking water guideline set by WHO (2011) and the Chinese standard GB 5749–2022. However, concentrations of As in water at depths below 20 m in the XW Reservoir approached the aforementioned drinking water guideline of 10 μg/L.

The implementation of the WQI, HEI, and NI further assessed the water quality status of these cascade reservoirs (Fig. 7 and S1). The WQI values in these reservoirs ranged from 9.2 to 19.1, significantly below the benchmark value of 100, indicating the 'excellent' water quality. The HEI results varied between 0.96 and 1.37, which were below 10, reflecting low TM pollution in these cascade reservoirs. NI values ranged from 0.35 to 0.56, all less than 1, indicating negligible metal pollution. Notably, WQI values in the hypolimnion were higher than those in the epilimnion in the XW and NZD Reservoirs, suggesting better surface water quality in deep-water reservoirs where thermal stratification existed.

3.5. Health risk assessment

The findings of this study, as presented in Table S5, provided a comprehensive summary of the HQ and HI results for TMs in cascade reservoirs, considering various exposure pathways. The results indicated that values of HQ (including $HQ_{ingestion}$ and HQ_{dermal}) and HI for all TMs in the surface water of these reservoirs consistently remained below 1, reflecting a limited level of risk associated with these elements (Wu et al., 2021b). The heightened health risks for children compared to adults underscored the vulnerability of children to TM exposure in the environment, a trend supported by previous studies (Jawad Ul et al., 2023; Varol and Tokatlı, 2021). The HQ and HI values for naturally sourced TMs (As, Cd, Li, Mo, Sb, and Sr) demonstrated elevated levels in upstream reservoirs (XW, MW, and DCS Reservoirs) due to the interception effects of cascade dams. In contrast, the HQ and HI values for anthropogenically sourced TMs (Co, Cu, Ni, Pb, and Zn) exhibited heightened values in downstream reservoirs (NZD and JH Reservoirs), attributed to increased human activities in the downstream region. The elements As, Sb, U, and Tl exhibited elevated HQ and HI values within the studied reservoirs, indicating their significant contribution to the potential non-carcinogenic risk in the cascade reservoir system. Moreover, the primary mode of exposure to these elements was found to be through ingestion absorption.

Based on the outcomes obtained from $HQ_{ingestion}$, $HQ_{dermal,}$ and HI assessments utilizing the most elevated concentrations of TM in the overlying water of each reservoir, it is evident that the largest $HQ_{ingestion}$ values were observed for As, followed by Sb, U, and Tl for both adults and children. Similarly, the largest HQ_{dermal} values were found for Sb, followed by V, As, and Cr for both adults and children. These findings suggested that the potential health hazard associated with TM ingestion was different from skin absorption. In addition, HI values for As were highest for both adults and children in each reservoir. Hence, arsenic was the primary factor contributing to non-carcinogenic risk, followed by Sb, U, Tl, and V. The $HQ_{ingestion}$ and HI values of As exceeded 1 for children in the XW reservoir and they were close to 1 in other reservoirs, which is noteworthy. Therefore, As can be concluded as the primary driver of potential non-carcinogenic risk, followed by Sb, U, Tl, and V, among the selected TMs in these cascade reservoirs.

While health hazards from some of the existing TMs in the studied cascade reservoirs were acknowledged, heavy metal pollution in these reservoirs did not appear to be extensive, aligning with the results of the WQI. Adverse human health effects associated with TMs have been identified in former research (Xu et al., 2020). For example, exposure to arsenic can give rise to chronic health disorders including diabetes, hypertension, and various cancers (Li et al., 2012). Sb can cause damage to the gastrointestinal tract and respiratory system of humans (Zhao et al., 2023). Consequently, TMs derived from human activities should be restrained from entering the upper Mekong Basin to preserve the superb water quality of the cascade reservoirs.

TMs often exhibit variations in toxicity based on their chemical form, species, or valence state. For instance, inorganic arsenic, particularly As (III), is considered the most toxic form of arsenic. Similarly, hexavalent chromium, Cr(VI), is known to be more toxic than trivalent chromium, Cr(III). In the Health Risk Assessment section of this study, it is important to note that all arsenic ions were assumed to be inorganic, and all chromium ions were assumed to be in the hexavalent state. It is acknowledged that these assumptions may impact the calculated values of HQ and CR for these elements, thus representing limitations in the study.

4. Conclusion

This study conducted a comprehensive investigation into the levels, causes, and sources of major TMs in five cascade reservoirs of the upper Mekong. Naturally sourced TMs (As, Cd, Li, Mo, Sb, and Sr) exhibited elevated levels in upstream reservoirs, attributed to the interception effects of cascade dams. Anthropogenically sourced TMs (Co, Cu, Ni, Pb, and Zn) showed heightened concentrations in downstream reservoirs, attributable to increased human activities in the downstream region. In

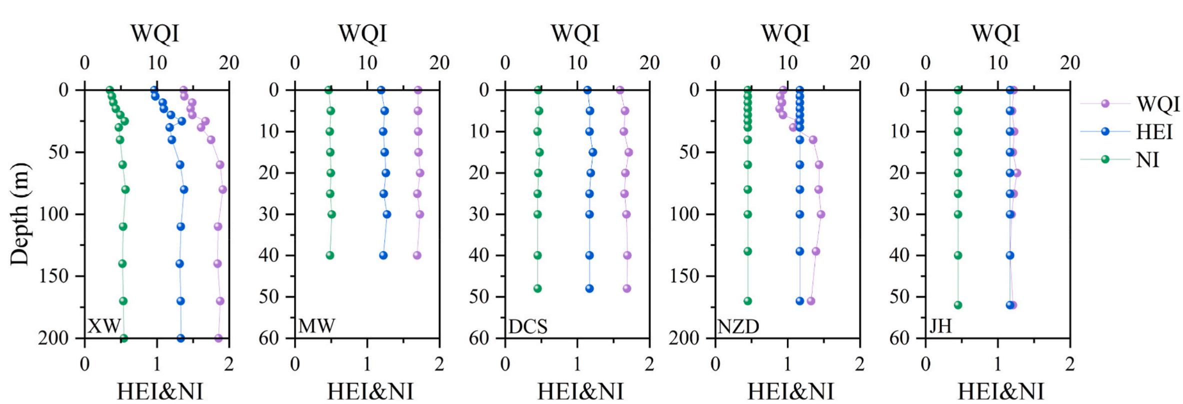

Fig. 7. Depth profiles of the water quality index (WQI), heavy metal evaluation index (HEI), and Nemerow index (NI) in the water column of the five cascaded reservoirs in the upper Mekong River.

deep-water reservoirs, the vertical distribution of redox-sensitive elements was influenced by the thermal stratification of the water column, with concentrations of As, Cs, Li, Mo, Rb, Sr, and U notably higher in the hypolimnion than the epilimnion. These findings underscore the importance of considering TMs at different water depths for assessing water quality and health risks in lakes and reservoirs, facilitating effective water resource management. Importantly, concentrations of all elements in these cascade reservoirs adhered to Chinese and WHO drinking water standards. The water quality index (WQI: 9.2–19.1), heavy metal evaluation index (HEI: 0.96–1.37), and Nemerow index (NI: 0.35–0.56) affirmed the commendable quality of the cascade reservoir waters. The health risk assessment identified As, Sb, U, and Tl as primary contributors to non-carcinogenic risks in the cascade reservoirs. Hence, future monitoring should include more frequent seasonal sampling and sediment analysis as a secondary source of contaminants in aquatic ecosystems.

CRediT authorship contribution statement

Zhenjie Zhao: Writing – original draft, Visualization, Resources, Methodology, Investigation, Formal analysis, Writing – review & editing. **Shilu Wang:** Resources, Conceptualization. **Weiqi Lu:** Resources, Investigation. **Wentao Yang:** Formal analysis. **Shehong Li:** Writing – original draft, Supervision, Resources, Conceptualization, 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.jclepro.2024.141049.

References

Abdul, K.S.M., Jayasinghe, S.S., Chandana, E.P.S., Jayasumana, C., De Silva, P., 2015. Arsenic and human health effects: a review. Environ. Toxicol. Pharmacol. 40, 828–846.
Adimalla, N., 2020. Heavy metals pollution assessment and its associated human health risk evaluation of urban soils from Indian cities: a review. Environ. Geochem. Health 42, 173–190.
Adimalla, N., Qian, H., 2019. Groundwater quality evaluation using water quality index (WQI) for drinking purposes and human health risk (HHR) assessment in an agricultural region of Nanganur, south India. Ecotoxicol. Environ. Saf. 176, 153–161.
Canpolat, Ö., Varol, M., Okan, Ö.Ö., Eriş, K.K., Çağlar, M., 2020. A comparison of trace element concentrations in surface and deep water of the Keban Dam Lake (Turkey) and associated health risk assessment. Environ. Res. 190, 110012.
Canpolat, Ö., Varol, M., Okan, Ö.Ö., Eriş, K.K., 2022. Sediment contamination by trace elements and the associated ecological and health risk assessment: a case study from a large reservoir (Turkey). Environ. Res. 204, 112145.
Chen, L., Zhang, H., Xie, Z., Ding, M., Devlin, A.T., Jiang, Y., et al., 2022. The temporal response of dissolved heavy metals to landscape indices in the Le'an river, China. Environ. Res. 210, 112941.
Chen, Y.L., Mao, W.J., Yang, W.T., Niazi, N.K., Wang, B., Wu, P., 2023. A novel phosphate rock-magnetic biochar for Pb^{2+} and Cd^{2+} removal in wastewater: characterization, performance and mechanisms. Environ. Technol. Innovat. 32, 103268.
Cheng, Y., Zhao, F., Wu, J., Gao, P., Wang, Y., Wang, J., 2022. Migration characteristics of arsenic in sediments under the influence of cascade reservoirs in Lancang River basin. J. Hydrol. 606, 127424.
Gao, B., Gao, L., Gao, J., Xu, D., Wang, Q., Sun, K., 2019. Simultaneous evaluations of occurrence and probabilistic human health risk associated with trace elements in typical drinking water sources from major river basins in China. Sci. Total Environ. 666, 139–146.
Gao, B., Gao, L., Xu, D.Y., Zhou, Y., Lu, J., 2018. Assessment of Cr pollution in tributary sediment cores in the Three Gorges Reservoir combining geochemical baseline and in situ DGT. Sci. Total Environ. 628–629, 241–248.
Gao, L., Lu, J., Xu, D., Wan, X., Gao, B., 2022. Partitioning behavior and ecological risk of arsenic and antimony in the sediment-porewater profile system in the Three Gorges Reservoir, China. Chemosphere, 134409.
Gao, Q., Li, Y., Cheng, Q., Yu, M., Hu, B., Wang, Z., et al., 2016. Analysis and assessment of the nutrients, biochemical indexes and heavy metals in the Three Gorges Reservoir, China, from 2008 to 2013. Water Res. 92, 262–274.
Gui, Z., Chen, X., He, Y., 2021. Spatiotemporal analysis of water resources system vulnerability in the Lancang River Basin, China. J. Hydrol. 601, 126614.
Guo, X., Tang, Y., Xu, Y., Zhang, S., Ma, J., Xiao, S., et al., 2020. Using stable nitrogen and oxygen isotopes to identify nitrate sources in the Lancang River, upper Mekong. J. Environ. Manag. 274, 111197.
Hao, R., Yin, W., Jia, H.Y., Xu, J.F., Li, N.X., Chen, Q.Z., et al., 2021. Dynamics of dissolved heavy metals in reservoir bays under different hydrological regulation. J. Hydrol. 595, 126042.
Haq, A.U., Muhammad, S., Tokatli, C., 2023. Spatial distribution of the contamination and risk assessment of potentially harmful elements in the Ghizer River Basin, northern Pakistan. Journal of Water and Climate Change 14, 2309–2322.
Islam, A.M.T., Islam, H.M.T., Mia, M.U., Khan, R., Habib, M.A., Bodrud-Doza, M., et al., 2020. Co-distribution, possible origins, status and potential health risk of trace elements in surface water sources from six major river basins, Bangladesh. Chemosphere 249, 126180.
Jawad Ul, H., Siddique, M.A.B., Islam, M.S., Ali, M.M., Tokatli, C., Islam, A., et al., 2023. Effects of COVID-19 era on a subtropical river basin in Bangladesh: heavy metal (loid)s distribution, sources and probable human health risks. Sci. Total Environ. 857, 159383.
Krueger, K.M., Vavrus, C.E., Lofton, M.E., McClure, R.P., Gantzer, P., Carey, C.C., et al., 2020. Iron and manganese fluxes across the sediment-water interface in a drinking water reservoir. Water Res. 182, 116003.
Li, D., Yang, C., Xu, X., Li, S., Luo, G., Zhang, C., et al., 2023a. Low dosage fluorine ameliorates the bioaccumulation, hepatorenal dysfunction and oxidative stress, and gut microbiota perturbation of cadmium in rats. Environ. Pollut. 324, 121375.
Li, H., Yao, J., Sunahara, G., Min, N., Li, C., Duran, R., 2023b. Quantifying ecological and human health risks of metal(loid)s pollution from non-ferrous metal mining and smelting activities in Southwest China. Sci. Total Environ. 873, 162364.
Li, S., Xiao, T., Zheng, B., 2012. Medical geology of arsenic, selenium and thallium in China. Sci. Total Environ. 421–422, 31–40.
Li, Y.Y., Gao, B., Xu, D.Y., Peng, W.Q., Liu, X.B., Qu, X.D., et al., 2020. Hydrodynamic impact on trace metals in sediments in the cascade reservoirs, North China. Sci. Total Environ. 716, 136914.
Luo, M., Yu, H., Liu, Q., Lan, W., Ye, Q., Niu, Y., et al., 2021. Effect of river-lake connectivity on heavy metal diffusion and source identification of heavy metals in the middle and lower reaches of the Yangtze River. J. Hazard Mater. 416, 125818.
Mokarram, M., Saber, A., Sheykhi, V., 2020. Effects of heavy metal contamination on river water quality due to release of industrial effluents. J. Clean. Prod. 277, 123380.
Muhammad, S., 2022. Evaluation of heavy metals in water and sediments, pollution, and risk indices of Naltar Lakes, Pakistan. Environ. Sci. Pollut. Control Ser. 30, 28217–28226.
Muhammad, S., Ahmad, K., 2020. Heavy metal contamination in water and fish of the Hunza River and its tributaries in Gilgit-Baltistan: evaluation of potential risks and provenance. Environ. Technol. Innovat. 20, 101159.
Muhammad, S., Ullah, I., 2022. Spatial and seasonal variation of water quality indices in Gomal Zam Dam and its tributaries of south Waziristan District, Pakistan. Environ. Sci. Pollut. Control Ser. 29, 29141–29151.
Naujokas, M.F., Anderson, B., Ahsan, H., Aposhian, H.V., Graziano, J.H., Thompson, C., et al., 2013. The broad scope of health effects from chronic arsenic exposure: update on a worldwide public health problem. Environ. Health Perspect. 121, 295–302.
Şener, Ş., Şener, E., Davraz, A., 2017. Evaluation of water quality using water quality index (WQI) method and GIS in Aksu River (SW-Turkey). Sci. Total Environ. 584–585, 131–144.
Shil, S., Singh, U.K., 2019. Health risk assessment and spatial variations of dissolved heavy metals and metalloids in a tropical river basin system. Ecol. Indicat. 106, 105455.
Song, J., Fu, K., Su, B., Huang, Q., Huang, J., Zhang, J., 2013. Spatial distribution of heavy metal concentrations and pollution assessment in the bed loads of the Lancang River System. Acta Geograph. Sin. 68, 389–397.
Tang, W., Shu, L., Ng, J.C., Bai, Y., Zhao, Y., Lin, H., et al., 2022. Metal(loid) flux change in dongting lake due to the operation of three Gorges dam, China. Environ. Pollut. 306, 119342.

Tokatli, C., Mutlu, E., Arslan, N., 2021. Assessment of the potentially toxic element contamination in water of Sehriban Stream (Black Sea Region, Turkey) by using statistical and ecological indicators. Water Environ. Res. 93, 2060–2071.
Tokatlı, C., Uğurluoğlu, A., Muhammad, S., 2023. Ecotoxicological evaluation of organic contamination in the world's two significant gateways to the Black Sea using GIS techniques: Turkish Straits. Mar. Pollut. Bull. 194, 115405.
Tokatlı, C., Varol, M., 2021. Variations, health risks, pollution status and possible sources of dissolved toxic metal (loid) s in stagnant water bodies located in an intensive agricultural region of Turkey. Environ. Res. 201, 111571.
Varol, M., 2019. Arsenic and trace metals in a large reservoir: seasonal and spatial variations, source identification and risk assessment for both residential and recreational users. Chemosphere 228, 1–8.
Varol, M., 2020. Environmental, ecological and health risks of trace metals in sediments of a large reservoir on the Euphrates River (Turkey). Environ. Res. 187, 109664.
Varol, M., Karakaya, G., Sünbül, M.R., 2021. Spatiotemporal variations, health risks, pollution status and possible sources of dissolved trace metal(loid)s in the Karasu River, Turkey. Environ. Res. 202, 111733.
Varol, M., Tokatlı, C., 2023. Evaluation of the water quality of a highly polluted stream with water quality indices and health risk assessment methods. Chemosphere 311, 137096.
Varol, M., Tokatlı, C., 2021. Impact of paddy fields on water quality of Gala Lake (Turkey): an important migratory bird stopover habitat. Environ. Pollut. 287, 117640.
Varol, M., Ustaoğlu, F., Tokatlı, C., 2022. Ecological risks and controlling factors of trace elements in sediments of dam lakes in the Black Sea Region (Turkey). Environ. Res. 205, 112478.
Vink, J.P.M., van Zomeren, A., Dijkstra, J.J., Comans, R.N.J., 2017. When soils become sediments: large-scale storage of soils in sandpits and lakes and the impact of reduction kinetics on heavy metals and arsenic release to groundwater. Environ. Pollut. 227, 146–156.
Wang, C., Liu, S.L., Zhao, Q.H., Deng, L., Dong, S.K., 2012. Spatial variation and contamination assessment of heavy metals in sediments in the Manwan Reservoir, Lancang River. Ecotoxicol. Environ. Saf. 82, 32–39.
Wang, C., Zhou, H., Kuang, X., Hao, Y., Shan, J., Chen, J., et al., 2021. Water quality and health risk assessment of the water bodies in the Yamdrok-tso basin, southern Tibetan Plateau. J. Environ. Manag. 300, 113740.
Wang, J., Chen, J., Ding, S., Guo, J., Christopher, D., Dai, Z., et al., 2016. Effects of seasonal hypoxia on the release of phosphorus from sediments in deep-water ecosystem: a case study in Hongfeng Reservoir, Southwest China. Environ. Pollut. 219, 858–865.
Wang, X., Zhang, L., Zhao, Z., Cai, Y., 2018. Heavy metal pollution in reservoirs in the hilly area of southern China: distribution, source apportionment and health risk assessment. Sci. Total Environ. 634, 158–169.
Wen, C., Zhu, S., Li, N., Luo, X., 2022. Source apportionment and risk assessment of metal pollution in natural biofilms and surface water along the Lancang River, China. Sci. Total Environ. 843, 156977.
WHO, 2011. Guidelines for Drinking-Water Quality, fourth ed.
Wu, H., Xu, C., Wang, J., Xiang, Y., Ren, M., Qie, H., et al., 2021a. Health risk assessment based on source identification of heavy metals: a case study of Beiyun River, China. Ecotoxicol. Environ. Saf. 213, 112046.
Wu, Z.S., Ma, T.T., Lai, X.J., Li, K.Y., 2021b. Concentration, distribution, and assessment of dissolved heavy metals in rivers of Lake Chaohu Basin, China. J. Environ. Manag. 300, 113744.
Xiang, R., Wang, L.J., Li, H., Tian, Z.B., Zheng, B.H., 2021. Water quality variation in tributaries of the three Gorges reservoir from 2000 to 2015. Water Res. 195, 116993.
Xiao, J., Wang, L., Deng, L., Jin, Z., 2019. Characteristics, sources, water quality and health risk assessment of trace elements in river water and well water in the Chinese Loess Plateau. Sci. Total Environ. 650, 2004–2012.
Xu, S., Lang, Y., Zhong, J., Xiao, M., Ding, H., 2020. Coupled controls of climate, lithology and land use on dissolved trace elements in a karst river system. J. Hydrol. 591, 125328.
Yang, Y., Huang, X., Wu, X.Q., Liu, C.R., Zhao, S.Y., Zhu, X.H., 2023. The spatiotemporal variations characteristic and pollution evaluation of water quality in Qujiang River, China. Appl. Water Sci. 13, 32.
Yin, Y., Zhang, W., Tang, J., Chen, X., Zhang, Y., Cao, X., et al., 2022. Impact of river dams on phosphorus migration: a case of the Pubugou Reservoir on the Dadu River in China. Sci. Total Environ. 809, 151092.
Yuan, B., Cao, H.L., Du, P., Ren, J., Chen, J., Zhang, H., et al., 2023. Source-oriented probabilistic health risk assessment of soil potentially toxic elements in a typical mining city. J. Hazard Mater. 443, 130222.
Zeng, J., Han, G., Yang, K., 2020. Assessment and sources of heavy metals in suspended particulate matter in a tropical catchment, northeast Thailand. J. Clean. Prod. 265, 121898.
Zeng, J., Han, G.L., Hu, M.M., Wang, Y.C., Liu, J.K., Zhang, S.T., et al., 2021. Geochemistry of dissolved heavy metals in upper reaches of the three Gorges reservoir of yangtze river watershed during the flood season. Water 13, 2078.
Zeng, J., Han, G., Zhang, S., Liang, B., Qu, R., Liu, M., et al., 2022. Potentially toxic elements in cascade dams-influenced river originated from Tibetan Plateau. Environ. Res. 208, 112716.
Zhang, F., Wang, Y., Liao, X., 2022. Recognition method for the health risks of potentially toxic elements in a headwater catchment. Sci. Total Environ. 839, 156287.
Zhang, J., Fu, K., Wang, B., Chen, L., Song, J., Su, B., 2014. Assessment of heavy metal pollution of bed sediment in the Lancang River. Prog. Geogr. 33, 1136–1144.
Zhang, J., Wu, Q., Wang, Z., Gao, S., Jia, H., Shen, Y., 2021a. Distribution, water quality, and health risk assessment of trace elements in three streams during the wet season, Guiyang, Southwest China. Elementa: Science of the Anthropocene 9, 00133.
Zhang, J.H., Li, X.C., Guo, L.Q., Deng, Z.M., Wang, D.W., Liu, L.S., 2021b. Assessment of heavy metal pollution and water quality characteristics of the reservoir control reaches in the middle Han River, China. Sci. Total Environ. 799, 149472.
Zhao, Z., Li, S., Wang, S., Liao, J., Lu, W., Tan, D., et al., 2022. Heavy metal characteristics in porewater profiles, their benthic fluxes, and toxicity in cascade reservoirs of the Lancang River, China. Environ. Sci. Pollut. Res. 29, 36013–36022.
Zhao, Z., Li, S., Xue, L., Liao, J., Zhao, J., Wu, M., et al., 2020. Effects of dam construction on arsenic mobility and transport in two large rivers in Tibet, China. Sci. Total Environ. 741, 140406.
Zhao, Z.J., Li, S.H., Wang, S.L., Lu, W.Q., Chen, J.A., 2023. Multivariate statistical analysis and risk assessment of dissolved trace metal(loid)s in the cascade-dammed Lancang River. Process Saf. Environ. Protect. 172, 241–249.