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Multivariate statistical analysis and risk assessment of dissolved trace metal (loid)s in the cascade-dammed Lancang River

Zhenjie Zhao[a,b,1], Shehong Li[b,2,*], Shilu Wang[b], Weiqi Lu[c], Jingan Chen[b]

[a] Key Laboratory of Environmental Pollution Monitoring and Disease Control, Ministry of Education, School of Public Health, Guizhou Medical University, Guiyang 550025, China
[b] State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, China
[c] Guizhou University of Finance and Economics, Guiyang 550025, China

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ABSTRACT

As a prominent factor impacting water quality, the dissolved trace metal(loid) (DTM) has not been adequately studied in the Lancang River Basin (LRB). Herein, we sampled surface water from 46 sites in the LRB to explore the spatial distribution, primary sources, and associated human health risks of 18 DTMs (As, Ba, Cd, Co, Cr, Cs, Cu, Li, Mo, Ni, Pb, Rb, Sb, Sr, Tl, U, V, and Zn). Affected by geothermal spring input, the concentrations of As, Cs, Li, Mo, Sr, and U were significantly higher in the mainstream than those in tributaries, with a decreasing trend along the flow direction in the mainstream of the LRB. Tl and Rb are mainly related to naturally derived sources, like soil erosion, Cd, Cr, Cu, Pb, and Zn were mainly attributed to anthropogenic processes, Co and Ni were affected by both natural and anthropogenic processes, while Sb was mainly from the Sb deposit in the basin. As a result of the combined influence of multiple sources of pollution, the spatial distribution of these elements does not seem to be regular. Most of the DTM concentrations in the LRB were within the Chinese and WHO drinking water guideline values, while 43.5% of the samples contained arsenic exceeding the guideline value of 10 μg/L. In general, the water quality of the LRB was good. Health risk assessment indicated that As, Sb, and Tl were the primary drivers of the non-carcinogenic risk. These results give the scientific basis needed for metal(loid) cycling in the aquatic environment and the ecological management of the LRB.

1. Introduction

As an essential requirement for life, water is one of the world's most imperiled resources. The quality of water sources has deteriorated due to urbanization, industry, agriculture, and other human activities, resulting in the detriment of their use for drinking, recreation, domestic, and irrigation purposes (Naz et al., 2022; Varol and Tokatlı, 2022; Yeh et al., 2022; Zhang et al., 2022). Thus, assessing the current status of contamination levels in aquatic ecosystems is important, which could strongly support sustainability planning for water resources (Zeng et al., 2020). Particularly, rivers are relatively more vulnerable to contamination due to their open environmental space (Ustaoğlu et al., 2021; Zhang et al., 2021). Therefore, it is significant to gather reliable information about river water quality, distinguish contamination sources, and control river contamination (Iwar et al., 2021; Varol, 2020).

Among the critical issues facing water resources is the contamination of trace metal(loid)s due to their toxic, nonbiodegradable, and bioaccumulative features (Nguyen et al., 2020; Tang et al., 2022, 2020). Large quantities of trace metal(loid)s are released into rivers by natural processes (e.g., soil erosion) and anthropogenic activities (e.g., mining and smelting) (Wang et al., 2021; Wen et al., 2022; Xu et al., 2020). Elevated concentrations of dissolved trace metal(loid)s (DTMs) in surface water could pose serious risks to human health and ecosystem stability (Githaiga et al., 2021; Nuruzzama et al., 2021; Wu et al.,

Abbreviations: DTM, Dissolved trace metal(loid); LRB, Lancang River Basin; T, Water temperature; TDS, Total dissolved solids; DO, Dissolved oxygen; EC, Electrical conductivity; CA, Correlation analysis; PCA, Principal component analysis; WQI, Water quality index; HEI, Heavy metal evaluation index; NI, Nemerow index; HQ, Hazard quotient; HI, Hazard index; K-S, Kolmogorov-Smirnov; SD, Standard deviation; PC, Principal components.

* Corresponding author.
E-mail addresses: zhaozhenjie@gmc.edu.cn (Z. Zhao), lishehong@vip.gyig.ac.cn (S. Li), wangshilu@vip.gyig.ac.cn (S. Wang), 326178956@qq.com (W. Lu), chenjingan@vip.gyig.ac.cn (J. Chen).
[1] 0000-0003-2214-7591
[2] 0000-0003-0151-6407

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2021b). For example, people chronically exposed to DTMs in water can suffer from neurological damage, reproductive abnormalities, liver and kidney damage, cardiovascular problems, and developmental anomalies (Wu et al., 2021a; Xiong et al., 2021).

Originating from the Tibetan Plateau, the Lancang River (Upper Mekong) is a major source of water for Southeast Asian countries (Zeng et al., 2022). Besides nourishing a wide range of ecosystems, it also provides support to the fishing and agricultural sectors along the river and is a prominent transportation hub with great value economically, socially, and ecologically, which is increasingly attracting attention (Cheng et al., 2022). Moreover, the environmental sensibility and fragility of this river also make it of great significance. Considering the diverse geological and human activities in the LRB, the distribution and health risks of DTMs, water quality, and factors that are most responsible for DTM contamination need to be identified for the efficient management of water resources. A few studies have reported the contents of the DTMs in the LRB (Liang et al., 2020; Wen et al., 2022). However, only several heavy metals such as Cr, Cu, Ni, Pb, and Zn were reported, and the sampling sites were insufficient. Moreover, these existing samples were only collected from the mainstream, the status of the tributaries and their influence on the mainstream was not clear.

Therefore, this study was performed to fully understand the spatial distribution of 18 DTMs (As, Ba, Cd, Co, Cr, Cs, Cu, Li, Mo, Ni, Pb, Rb, Sb, Sr, Tl, U, V, and Zn) in the mainstream and tributaries of the LRB, assess the water quality, recognize the potential origins of 18 DTMs, and evaluate their associated human health risks. This research will give scientific support for the rational control of contamination and the improvement of water quality for the LRB and provide a useful guide for other similar areas.

2. Materials and methods

2.1. Study area

The Lancang River is the upper reach of the Mekong River, the largest international river in Asia (Guo et al., 2020b). With a length of 2161 km and a catchment area of 16.7×10^4 km^2, the Lancang River occupies 20.7% of the Lancang-Mekong River Basin (Zeng et al., 2022). There is a 4595-meter elevation drop on the main river and an average annual flow of 2350 m^3/s at its borders (Guo et al., 2020a). Due to its large descending elevation and huge runoff, the Lancang River has an extensive hydraulic resource (Mu et al., 2020; Zhao et al., 2022b). The use and management of LRB water resources are of significant concern to Chinese and other governments. Six dams had been constructed on the river's mainstream by 2016 (Wang et al., 2020). The locations of these cascade dams were presented in Fig. 1, and their main characteristics were displayed in Table S1.

2.2. Sampling and analysis

Surface waters were sampled from 46 sites in the LRB from March to April 2017, including 37 samples from the mainstream (named M1 - M37) and 9 samples from the major tributaries (named T1 – T9). The precise sampling locations were exhibited in Fig. 1 and Table S2. Water temperature (T), pH, total dissolved solids (TDS), dissolved oxygen (DO), and electrical conductivity (EC) were determined in situ with a portable multiparameter water quality analyzer (YSI 6600V2, YSI Ltd., USA). Waters were sampled at ~15 cm beneath the surface and filtered through 0.45 μm cellulose acetate filtration membranes immediately after collection. Waters were saved in 0.05 L acid-washed polypropylene bottles. For each site, two bottles of filtered water were obtained: one was acidified to 2% HNO_3 for cation and metal(loid) analysis, and another was used for anion analysis. Waters were kept at 4 °C before analysis.

Major cations (Ca^{2+}, Mg^{2+}, Na^+, K^+, and SiO_2) were determined by ICP-OES (iCAP6500, Thermo Scientific). Major anions (F^-, Cl^-, NO_3^-, and SO_4^{2-}) were analyzed using ion chromatography (IC, DIONEX ICS-90). DTMs were measured by ICP-MS (Agilent, 7700 ×, USA).

2.3. Statistical analysis

In order to ascertain the normality of the data, Kolmogorov-Smirnov (K-S) was conducted (Zeng et al., 2022). Two sets of data were compared using the independent sample t-test. To determine the potential sources of DTMs, correlation analysis (CA) and principal component analysis (PCA) were adopted (Chen et al., 2022; Islam et al., 2020; Zhao et al., 2022b). The above analyses were completed with SPSS 21.0.

2.4. Indices for water quality status

To determine the comprehensive water quality status of the LRB, the water quality index (WQI), heavy metal evaluation index (HEI), and the Nemerow index (NI) were employed (Jiang et al., 2022; Mokarram et al., 2020; Mukherjee et al., 2020; Şener et al., 2017; Ustaoğlu et al., 2021; Varol and Tokatlı, 2021). Detailed calculations of these indices were provided in the Supplementary Materials (Text S1-S3).

2.5. Health risk assessment

To assess human health risks from river water DTMs, the Hazard quotient (HQ) and Hazard index (HI) were applied (Chai et al., 2021; Maleki and Jari, 2021; Xu et al., 2020). HQ was used to represent the underlying non-carcinogenic risks of metal(loid)s. HI was an aggregate of the HQs. The calculation formula for HQ and HI were as follows:

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

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

where ADD_i refers to the average daily intake of metal(loid) due to direct ingestion and skin contact with calculation details and corresponding parameters given in the Supplementary Materials (Text S4 and Table S3). RfD_i is the reference dose (Table 2). The HQ or HI value > 1 indicates probable detrimental effects on health, and the HQ or HI value < 1 denotes no adverse health effects (Gao et al., 2019; Zhao et al., 2022a).

3. Results and discussion

3.1. Characteristics of DTMs, major ions, and physic-chemical parameters

The statistics of DTMs, major ions, and physic-chemical parameters at 46 sampling sites in the LRB were presented in Table 1 and S4. The K-S test results showed that the data of As, Ba, Mo, Sr, Mg^{2+}, F^-, SO_4^{2-}, pH, EC, TDS, and salinity were found to be normally distributed. For the remaining elements, the K-S test and wide standard deviation (SD) (Table 1 and S4) suggested that outliers might have affected the mean concentration (Wang et al., 2017; Xu et al., 2020). Therefore, median concentrations rather than arithmetic means were applied in our analysis. Nevertheless, arithmetic means were used to compare with the guidelines and the results of other rivers generally reported as arithmetic means.

The pH values of waters from the LRB ranged from between 7.26 and 8.51, with a mean value of 8.11, showing a slightly alkaline characteristic (Table S4). The DO content in surface water was high (8.25–15.50 mg/L). Water temperature values varied between 1.3.3 ℃ and 18.3 ℃. The EC values ranged between 242 and 903 μS/cm, with a mean value of 591 μS/cm. The TDS values of river water varied between 157 and 587 mg/L, with a mean value of 384 mg/L. The values of EC and TDS in the upstream waters were greater than those in the

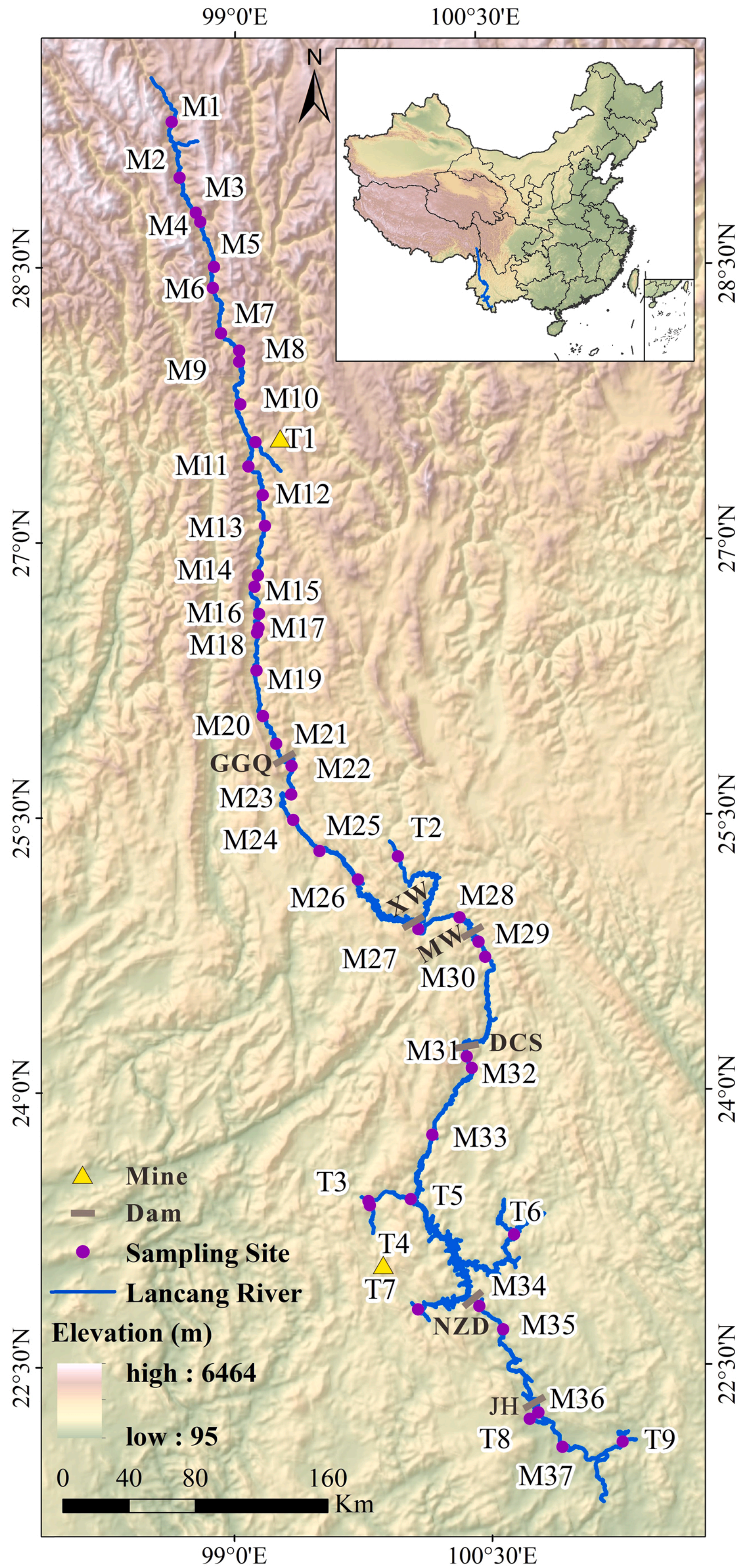

Fig. 1. Location of sampling sites in the Lancang River Basin (LRB), China.

Table 1
Concentrations of dissolved metal(loid)s (μg/L) in the Lancang River Basin (LRB).

	Whole Basin				Mainstream				Tributaries				t-test
	Range	Median	Mean	SD	Range	Median	Mean	SD	Range	Median	Mean	SD	*p*
As	0.93–20.84	9.24	9.43	6.18	2.65–20.84	11.27	11.02	5.75	0.93–9.25	2.06	2.88	2.52	0.000
Ba	23.39–80.40	51.41	51.56	9.40	43.25–64.11	51.80	52.61	5.83	23.39–80.40	51.01	47.27	17.85	0.401
Cd	0.00–0.20	0.01	0.02	0.03	0.00–0.20	0.01	0.02	0.03	0.00–0.08	0.01	0.02	0.02	0.976
Co	0.01–0.42	0.03	0.05	0.07	0.01–0.07	0.03	0.03	0.01	0.03–0.42	0.11	0.14	0.13	0.034
Cr	0.03–0.85	0.20	0.22	0.12	0.09–0.42	0.20	0.21	0.07	0.03–0.82	0.19	0.26	0.25	0.542
Cs	0.00–4.84	0.68	1.49	1.75	0.02–4.84	1.75	1.82	1.80	0.00–0.66	0.07	0.15	0.21	0.000
Cu	0.23–4.60	0.64	0.73	0.66	0.23–1.00	0.57	0.58	0.20	0.46–4.60	0.82	1.37	1.29	0.102
Li	0.62–47.76	17.83	21.08	15.32	7.35–47.76	26.76	25.55	13.64	0.62–11.20	1.67	2.72	3.31	0.000
Mo	0.16–1.85	1.14	1.09	0.49	0.61–1.85	1.27	1.26	0.38	0.16–0.67	0.37	0.40	0.17	0.000
Ni	0.00–4.91	0.09	0.32	0.84	0.00–0.66	0.08	0.12	0.13	0.00–4.91	0.45	1.17	1.70	0.100
Pb	0.02–2.46	0.10	0.21	0.37	0.02–0.39	0.09	0.11	0.09	0.07–2.46	0.28	0.58	0.75	0.097
Rb	0.78–12.92	3.85	4.31	2.53	1.95–8.08	4.88	4.58	2.15	0.78–12.92	1.93	3.24	3.71	0.157
Sb	0.14–17.32	1.14	1.75	2.53	0.56–3.27	1.17	1.57	0.95	0.14–17.32	0.55	2.52	5.59	0.625
Sr	50.5–910.4	485.9	487.5	261.7	241.6–910.4	607.3	571.4	212.4	50.5 – 455.0	108.6	142.4	125.0	0.000
Tl	0.00–0.08	0.02	0.02	0.01	0.01–0.03	0.02	0.02	0.01	0.00–0.08	0.02	0.02	0.02	0.878
U	0.10–1.93	1.15	1.11	0.52	0.61–1.93	1.22	1.31	0.36	0.10–0.62	0.28	0.30	0.16	0.000
V	0.22–2.43	0.43	0.52	0.37	0.22–0.82	0.39	0.42	0.17	0.32–2.43	0.71	0.93	0.62	0.038
Zn	0.69–42.09	3.20	4.95	6.59	0.69–16.44	2.86	3.68	3.09	0.93–42.09	4.80	10.18	12.76	0.167

Table 2
Reference dose (RfD), hazard quotient (HQ), and hazard index (HI) for dissolved metal(loid)s in the Lancang River Basin.

	$RfD^{a}_{ingestion}$	RfD^{a}_{dermal}	$HQ_{ingestion}$		HQ_{dermal}		HI	
	$\mu g\ kg^{-1}\ day^{-1}$	$\mu g\ kg^{-1}\ day^{-1}$	Adult	Child	Adult	Child	Adult	Child
Whole Basin								
As	0.3	0.123	8.44E-01	1.26E+ 00	1.07E-02	3.17E-02	8.55E-01	1.29E+ 00
Ba	70	14	2.01E-02	3.00E-02	5.25E-04	1.55E-03	2.06E-02	3.16E-02
Cd	0.5	0.005	5.48E-04	8.18E-04	2.86E-04	8.44E-04	8.34E-04	1.66E-03
Co	0.3	0.06	2.74E-03	4.09E-03	2.86E-05	8.44E-05	2.77E-03	4.18E-03
Cr	3	0.015	1.83E-03	2.73E-03	1.91E-03	5.63E-03	3.73E-03	8.35E-03
Cu	40	12	4.38E-04	6.55E-04	7.63E-06	2.25E-05	4.46E-04	6.77E-04
Li	20	10	2.44E-02	3.65E-02	2.55E-04	7.52E-04	2.47E-02	3.72E-02
Mo	5	1.9	6.25E-03	9.33E-03	8.58E-05	2.53E-04	6.33E-03	9.58E-03
Ni	20	5.4	1.51E-04	2.25E-04	5.83E-07	1.72E-06	1.51E-04	2.27E-04
Pb	1.4	0.42	1.96E-03	2.92E-03	3.41E-06	1.00E-05	1.96E-03	2.93E-03
Sb	0.4	0.008	7.81E-02	1.17E-01	2.04E-02	6.01E-02	9.85E-02	1.77E-01
Sr	600	120	2.22E-02	3.31E-02	5.79E-04	1.71E-03	2.28E-02	3.48E-02
Tl	0.01	0.01	5.48E-02	8.18E-02	2.86E-04	8.44E-04	5.51E-02	8.27E-02
U	0.6	0.51	5.25E-02	7.84E-02	3.22E-04	9.51E-04	5.28E-02	7.94E-02
V	1	0.01	1.18E-02	1.76E-02	6.15E-03	1.81E-02	1.79E-02	3.57E-02
Zn	300	60	2.92E-04	4.36E-04	4.58E-06	1.35E-05	2.97E-04	4.50E-04
Highest concentration[b]								
As	0.3	0.123	1.90E+ 00	2.84E+ 00	2.42E-02	7.15E-02	1.93E+ 00	2.91E+ 00
Ba	70	14	3.15E-02	4.70E-02	8.21E-04	2.42E-03	3.23E-02	4.94E-02
Cd	0.5	0.005	1.10E-02	1.64E-02	5.72E-03	1.69E-02	1.67E-02	3.32E-02
Co	0.3	0.06	3.84E-02	5.73E-02	4.00E-04	1.18E-03	3.88E-02	5.85E-02
Cr	3	0.015	7.76E-03	1.16E-02	8.10E-03	2.39E-02	1.59E-02	3.55E-02
Cu	40	12	3.15E-03	4.71E-03	5.48E-05	1.62E-04	3.21E-03	4.87E-03
Li	20	10	6.54E-02	9.77E-02	6.83E-04	2.02E-03	6.61E-02	9.97E-02
Mo	5	1.9	1.01E-02	1.51E-02	1.39E-04	4.11E-04	1.03E-02	1.55E-02
Ni	20	5.4	6.73E-03	1.00E-02	2.60E-05	7.67E-05	6.75E-03	1.01E-02
Pb	1.4	0.42	4.81E-02	7.19E-02	8.38E-05	2.47E-04	4.82E-02	7.21E-02
Sb	0.4	0.008	1.19E+ 00	1.77E+ 00	3.10E-01	9.13E-01	1.50E+ 00	2.68E+ 00
Sr	600	120	4.16E-02	6.21E-02	1.08E-03	3.20E-03	4.27E-02	6.53E-02
Tl	0.01	0.01	2.19E-01	3.27E-01	1.14E-03	3.38E-03	2.20E-01	3.31E-01
U	0.6	0.51	8.81E-02	1.32E-01	5.41E-04	1.60E-03	8.87E-02	1.33E-01
V	1	0.01	6.66E-02	9.94E-02	3.48E-02	1.03E-01	1.01E-01	2.02E-01
Zn	300	60	3.84E-03	5.74E-03	6.02E-05	1.78E-04	3.90E-03	5.92E-03

Note: [a] was referenced from Xiao et al. (2019), Xu et al. (2020); [b] the calculation adopted the highest level of each element in the whole basin.

downstream, and their values in the main channel were markedly greater than those in the tributaries ($p < 0.001$).

Table S5 compares DTMs in the LRB with those in other rivers. The mean contents of As, Ba, Cs, Li, Mo, Pb, Rb, Sb, Sr, U, and Zn in the LRB were well above the world average. Whereas the mean levels of Cd, Co, Cr, Cu, and Ni were far lower than the world average, and V was comparable (Gaillardet et al., 2014). Compared to other Chinese rivers, the mean levels of dissolved Co, Cr, Ni, and V were considerably lower than those in the Yangtze River, the Yellow River, the Pearl River, the Tarim River, the Lijiang River, and the Chishui River (Deng et al., 2021; Gaillardet et al., 2014; Gao et al., 2019; Xiao et al., 2014; Xu et al., 2020; Zeng et al., 2019). While the mean levels of dissolved As, Li and Sb in the LRB were significantly greater than those in the above rivers (except for Li in the Tarim River), and Ba, Cd, Cu, Mo, Pb, and Zn were comparable.

3.2. Spatial variation of DTMs

There were three distribution patterns of DTMs in the mainstream and tributaries of the LRB. The first distribution pattern containing the elements As, Cs, Li, Mo, Sr, and U was typically characterized by a significantly higher content in the mainstream than in tributaries ($p < 0.001$, Table 1). Specifically, the average values of As, Cs, Li, Mo, Sr, and U in the mainstream were 11.02, 1.82, 25.55, 1.26, 571.4, and 1.30 μg/L, respectively, whereas those in tributaries were only 2.88, 0.15, 2.72, 0.40, 142.4, and 0.30 μg/L, respectively. In the second distribution pattern, Co and V were significantly more concentrated in tributaries than in the mainstream ($p < 0.05$), with Co and V concentrations of 0.03 and 0.42 μg/L in the mainstream and 0.14 and 0.93 μg/L in tributaries, respectively. The rest elements (Cd, Cr, Cu, Ni, Pb, Rb, Sb, Tl, and Zn) belonged to the third distribution pattern and no significant difference existed between the contents of these elements in the mainstream and tributaries ($p > 0.05$). For major ions, the concentrations of Ca^{2+}, Mg^{2+}, Na^{+}, F^{-}, Cl^{-}, SO_4^{2-}, and HCO_3^{-} in the mainstream were significantly higher than those in tributaries ($p < 0.05$). While the contents of the rest ions (K^{+}, SiO_2, NO_3^{-}) did not differ significantly between the mainstream and the tributaries ($p > 0.05$).

Along the main channel of the LRB, As, Cs, Li, Mo, Rb, Sr, and U presented a decreasing trend from upstream to downstream (Fig. 2). The concentration of As declined from 20.84 μg/L (site M1) to 2.64 μg/L (site M37). Cs declined from 4.84 μg/L (site M4) to 0.02 μg/L (site M36). Li decreased from 47.76 μg/L (site M4) to 7.35 μg/L (site M36). Mo declined from 1.85 μg/L (site M4) to 0.61 μg/L (site M35). Rb declined from 8.08 μg/L (site M3) to 1.96 μg/L (site M35). Sr declined from 910.4 μg/L (site M3) to 262.9 μg/L (site M37). U declined from 1.93 μg/L (site M1) to 0.61 μg/L (site M34).

The concentrations of other DTMs along the mainstream of the LRB displayed different degrees of variations (Fig. 2). The levels of Cu, V, and Cd at the downstream sites (M20-M37) were generally above the levels at the upstream sites (except for Cu at site M8). Pb concentrations fluctuated irregularly, and abnormally high values often appeared at downstream sites (M22, M26, M27, M32, and M34), which might be related to the intense human activities such as industrial activities and transportation in the downstream area. The Sb concentration remained at a stable level, while it increased sharply after the confluence of the tributary where site T1 was located. The abnormally high Sb concentration at site T1 might be influenced by the nearby Gepoluo Antimony Mine (Fig. 1). In contrast, Ba, Cr, Co, Ni, and Tl concentrations were maintained at steady levels with relatively small variations. Notably, the highest levels of Cd, Co, Cr, Cu, Ni, Pb, and Zn in the LRB occurred at site T4, which is adjacent to the Lancang Lead Mine (Fig. 1) where mining has been booming since the 1950 s

3.3. Sources identification of DTMs

CA and PCA have been effective methods for determining the source of metal(loid)s (Ma et al., 2020; Zhao et al., 2022b).

3.3.1. Correlation analysis (CA)

In order to investigate the relationships among the 18 DTMs, CA was conducted (Fig. 3). Strong positive correlations ($p < 0.001$) existed among As, Cs, Li, Mo, Rb, Sr, and U with correlation values ranging from 0.56 to 0.99. Marked positive correlations among Co, Cr, Cu, Pb, and Zn

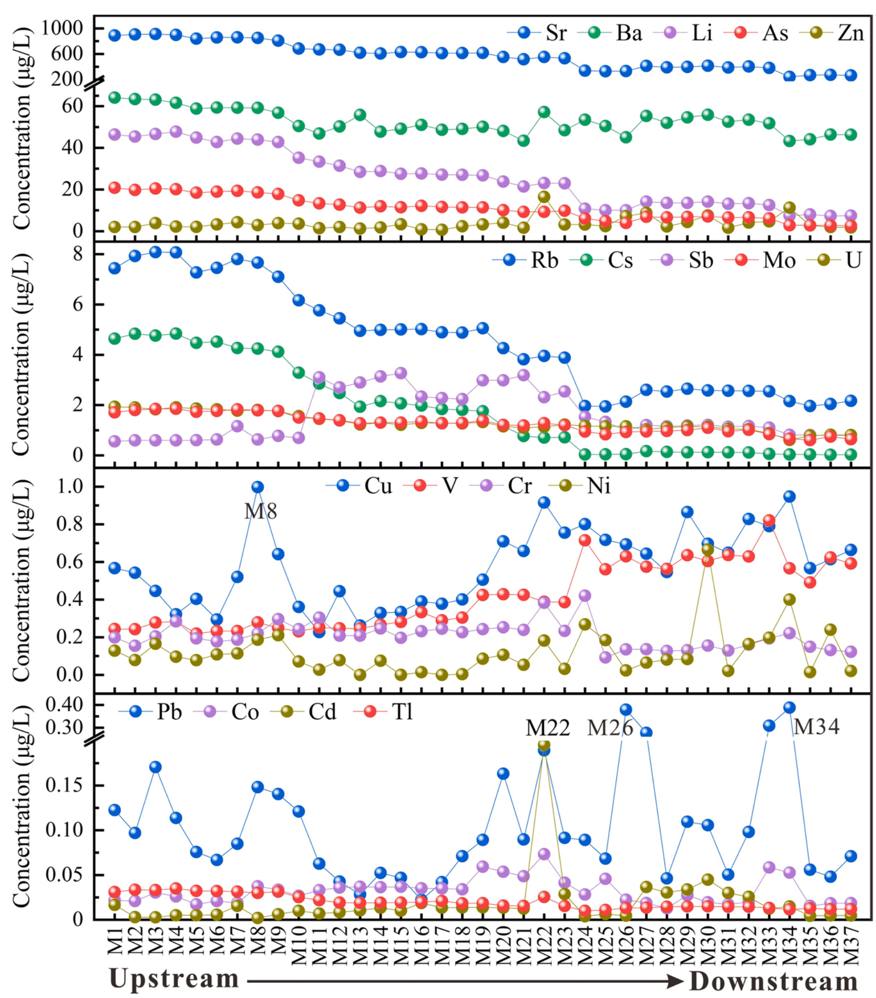

Fig. 2. Spatial variation of 18 dissolved trace metal(loid)s (DTMs) along the mainstream of the LRB.

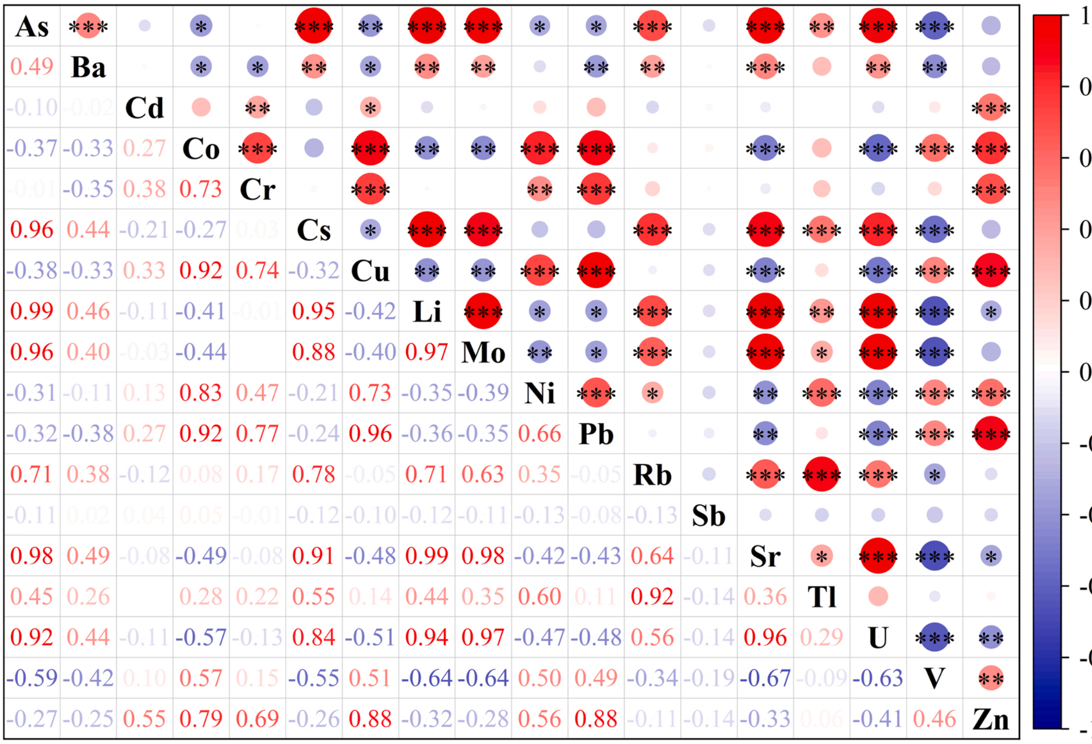

Fig. 3. Correlation coefficients of DTMs in the LRB. * ** Significant at the 0.001 level. * *Significant at the 0.01 level. *Significant at the 0.05 level.

($0.69 \leq R \leq 0.96, p < 0.001$) were also observed. It has been proved that dissolved metal(loid)s with high correlation coefficients could have certain homology or similar chemical behaviors (Wang et al., 2017; Xu et al., 2020; Zhao et al., 2020a). The lack of correlation between Sb and other elements indicated that Sb might have a completely different source.

3.3.2. Principal component analysis (PCA)

PCA was performed to further probe the origins of DTMs in the LRB. The values of KMO and Bartlett's sphericity test were 0.681 and 0.000, respectively, signifying the reliability of the results. As a result, four independent principal components (PC) were extracted, explaining a total of 86.51% variance (Table 3). Based on absolute load values > 0.75, 0.75–0.50, and 0.50–0.30, the factor loads were categorized as "high", "moderate", and "low", respectively (Varol, 2019; Wang et al., 2017).

Table 3
Factor loadings for varimax rotated PCA of metal(loid)s.

	PC1	PC2	PC3	PC4
Mo	**0.982**	-0.084	-0.007	-0.028
Li	**0.979**	-0.132	0.112	-0.021
Sr	**0.977**	-0.176	0.028	-0.005
As	**0.967**	-0.107	0.140	-0.028
U	**0.937**	-0.241	-0.046	-0.069
Cs	**0.908**	-0.098	0.285	-0.053
V	-0.665	0.294	0.130	-0.391
Ba	0.434	-0.308	0.259	0.291
Zn	-0.196	**0.911**	0.018	-0.109
Cr	0.096	**0.891**	0.089	-0.008
Pb	-0.283	**0.883**	0.181	-0.140
Cu	-0.337	**0.866**	0.220	-0.125
Co	-0.375	**0.801**	0.413	-0.009
Cd	0.045	**0.600**	-0.275	0.248
Tl	0.379	0.133	**0.862**	-0.024
Ni	-0.362	0.501	**0.754**	-0.090
Rb	0.645	0.028	**0.745**	-0.013
Sb	-0.127	-0.028	-0.046	**0.909**
% of variance	40.19	26.35	13.36	6.61
% of cumulative	40.19	66.53	79.89	86.51

Accounting for 40.19% of the total variance, PC1 exhibited high positive loadings of Mo (0.98), Li (0.98), Sr (0.98), As (0.97), U (0.94), Cs (0.91); moderate positive loadings of Rb (0.65); and low positive loading of Ba (0.43) and Tl (0.38) (Table 3). Extensive evidence showed that the abundant geothermal springs in Tibet were abnormally enriched in As, Cs, and Li (up to 126, 11.5, and 22.2 mg/L, respectively), and that the discharge of hot springs caused the enrichment of Li, As, and Cs in the upper Brahmaputra and Indus Rivers in Tibet (Zhao et al., 2020b, 2021). Some researchers found that Mo, Ba, Rb, and Sr were also abundant in Tibet's hot springs and that these elements tended to be present in considerably high levels in waters downstream of the spring outlets (Guo, 2012; Guo et al., 2019; Zhang et al., 2008, 2016). Furthermore, arsenic-rich hot springs and sediments have been found near the mainstream of the Lancang River, which also originates from the Tibetan Plateau (Zhang et al., 2017). As previously noted (Section 3.2), As, Cs, Li, Mo, Rb, Sr, and U displayed a decreasing trend from upstream to downstream along the mainstream of the LRB in this study (Fig. 2), and the levels of As, Cs, Li, Mo, Sr and U in the mainstream showed significantly greater than those in tributaries (Table 1). Combined with the significant positive correlations among As, Cs, Li, Mo, Rb, Sr, and U (Fig. 3), we can conclude that PC1 was attributed to geothermal spring input.

Explaining 26.35% of the total variance, PC2 exhibited high positive loadings of Zn (0.91), Cr (0.89), Pb (0.88), Cu (0.87), Co (0.80); and moderate positive loadings of Cd (0.60) and Ni (0.50). Relatively high correlation coefficients existed among Co, Cr, Cu, Pb, and Zn (Fig. 3). Mining, smelting, electroplating, and machine manufacture were the primary origins of Zn, Cu, and Cr (Zhao et al., 2020a). Pb and Cd were mainly from traffic emissions including automobiles and water transportation facilities (Fei et al., 2020; Zeng et al., 2022; Zhao et al., 2022a). After completion of these cascade reservoirs, the quantities of ships accumulated there, and fuel combustion and exhaust emissions could lead to increased Pb and Cd concentrations in water bodies. In addition, as previously noted (Section 3.2), the mining of Lancang Lead

Mine also led to a certain increase in Cd, Co, Cr, Cu, Ni, Pb, and Zn in the waters of the basin. Therefore, PC2 may be related to industrial activities and transportation.

Responsible for 13.36% of the total variance, PC3 was related to Tl (0.86), Ni (0.75), Rb (0.75), and Co (0.41). Considering the low concentrations of these elements, PC3 may be related to natural sources, like soil erosion. Combining the results of PC1, PC2, and PC3, it can be found that Mo, Li, Sr, As, U, Cs, Tl, and Rb were mainly associated with natural inputs, Cd, Cr, Cu, Pb, and Zn were mainly attributed to anthropogenic processes, and Co and Ni were influenced by both natural and anthropogenic processes. This is consistent with our recent findings on the sources of metals in sediment porewaters of the cascade reservoirs from Lancang River, with As primarily from natural inputs, Cu, Zn, Cd, and Pb from anthropogenic processes, and Ni from the combined inputs of natural and anthropogenic sources (Zhao et al., 2022b).

The PC4 explained an additional 6.61% of the total variance, with Sb (0.91) contributing the most. As mentioned previously, the levels of Sb in the basin were potentially affected by the Gepoluo Antimony Mine. The absence of correlations between Sb and other elements suggested that it had a significantly different origin from the other elements and confirmed the above observation. Therefore, PC4 was mainly from the Sb deposit and associated tailing leaching.

Therefore, the first distribution pattern (Section 3.2) containing As, Cs, Li, Mo, Sr, and U can be explained as the influence of hot spring input. Diluted by the continuously converging runoff, the concentrations of these elements in the mainstream decreased along the flow direction and were significantly higher than those in the tributaries. However, the spatial distribution of elements in the second and third distribution pattern did not seem to be regular as a result of the combined influence of multiple sources of pollution. And the concentrations of most elements were not significantly different between the mainstem and the tributaries.

3.4. Water quality assessment

The contents of major and trace elements were compared to the water quality standards (Table S6). All element (including major and trace element) contents in the LRB meet the Chinese surface water standard for Grade I (GB 3838–2002), suggesting the acceptance of the LRB surface waters as clean water sources for the national reserves. Except for As and Sb, the concentrations of other elements (including major and trace elements) in LRB water were below the drinking water guideline values recommended by China (GB 5749–2022) and the World Health Organization (WHO, 2011). There were 20 sites (M1-M20) in the LRB where As concentrations were above the Chinese and WHO drinking water guideline value of 10 μg/L. The Sb concentration in only one site (T1) of the LRB exceeded the Chinese drinking water guideline value of 5 μg/L, but not above the WHO drinking water guideline value of 20 μg/L.

The comprehensive water quality status of the LRB was further ascertained with the WQI, HEI, and NI (Fig. 4). The WQI values of all sampling sites ranged between 33.33 and 8.32 with a mean value of 20.95, which can be categorized as 'excellent' quality water. The HEI values varied between 0.45 and 2.72 with an average of 1.68, reflecting low metal pollution status. The NI values varied between 0.14 and 1.48 with an average of 0.73. Approximately 78.3% and 21.7% of the sampling sites exhibited insignificant and slight metal contamination, respectively. The values of WQI, HEI, and NI in the mainstream were markedly greater than those in tributaries ($p < 0.001$). Along the mainstem of the LRB, these values showed an overall declining trend from upstream to downstream. The relatively higher WQI, HEI, and NI values from site T1 in the tributary were related to the nearby Gepoluo Antimony Mine. Among all the sites, the largest contributor to the WQI and HEI values was As, except for sites T1 and T8, where the largest contributors were Sb and Tl, respectively (Figs. S2 & S3).

3.5. Health risk assessment

Table 2 summarized the HQ and HI values of DTMs via different pathways for adults and children in the LRB. The values of $HQ_{ingestion}$, HQ_{dermal}, and HI for all metal(loid)s (except for As) were less than 1, revealing the limited hazards caused by these elements. However, the $HQ_{ingestion}$ and HI values for As surpassed 1, indicating that potential non-carcinogenic concerns and adverse health effects may be posed by As via daily oral intake. Children had higher risk levels than adults, denoting that children may be more susceptible to toxic metal(loid)s (Table 2), which is consistent with previous research (Canpolat et al., 2020). The values of $HQ_{ingestion}$ for all DTMs except Cr were greater than HQ_{dermal}, indicating that oral ingestion may be the principal exposure route.

The calculated results of HQ and HI values using the maximum content of each metal in the LRB were also presented in Table 2. For both adults and children, the $HQ_{ingestion}$ values of arsenic were highest, followed by Sb and Tl, while the highest HQ_{dermal} values were Sb, followed by As and V, suggesting that the health hazard caused by DTMs via oral ingestion were different from those via the dermal contact. The HI values of As and Sb for both adults and children surpassed 1, followed by Tl which was near 1. Hence, it can be concluded that As, Sb, and Tl were the primary drivers of chronic risks among the study DTMs in the LRB.

Previous studies have demonstrated the harmful effects of toxic metal(loid)s on the human body. Particularly, As can cause a variety of complications such as skin damage, diabetes, hypertension, and neuropathy (Chai et al., 2021; Naujokas et al., 2013). Sb can cause adverse effects on the eyes, skin, respiratory system, and gastrointestinal tract (Jiang et al., 2021). Tl mainly entails polyneuritis (Liu et al., 2020). Therefore, special measures must be taken to control As, Sb, and Tl entering the Lancang River to protect human health and aquatic ecosystems.

Most toxic elements vary in their toxicity based on their form, species, or valence state. As an example, the most toxic form of arsenic is inorganic As, especially As(III). As for chromium, Cr(VI) is more toxic than Cr(III). In the part of the Health Risk Assessment of this study, all

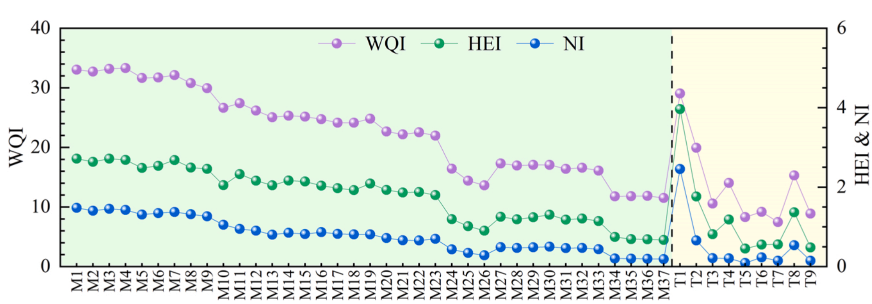

Fig. 4. Spatial variation of the water quality index (WQI), heavy metal evaluation index (HEI), and Nemerow index (NI) along the mainstream and tributaries of the LRB.

arsenic ions were assumed to be inorganic and all Cr ions were assumed to be hexavalent, which may affect the magnitude of the values of HQ and CR of these elements (Varol et al., 2021). These are the limitations of the study.

4. Conclusion

This study provided a comprehensive investigation of DTMs of surface waters in the LRB. Significant spatial heterogeneity was observed in the DTM levels in the LRB. The concentrations of As, Cs, Li, Mo, Sr, and U were significantly higher in the mainstream than those in tributaries, and they presented a decreasing trend along the flow direction in the mainstream of the LRB. From a basin-wide perspective, As, Cs, Li, Mo, Sr, and U were attributed to geothermal spring input, Tl and Rb were mainly associated with naturally derived sources (e.g., soil erosion), Cd, Cr, Cu, Pb, and Zn were mainly attributed to anthropogenic processes, Co and Ni were affected by both natural and anthropogenic processes, while Sb was mainly from the Sb deposit in the basin. Most of the DTM concentrations in the LRB were within the Chinese and WHO drinking water guideline values, while 43.5% of the samples contained arsenic exceeding the guideline value. In general, the water quality of the LRB was good. Health risk assessment suggested that As, Sb, and Tl were the primary drivers of the non-carcinogenic risk for the whole basin. We recommend that effective preventive measures are required to reduce the hot spring input into this important river in China. Mining enterprises should be inspected and the DTM levels along the river would need to be monitored at regular intervals.

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.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.psep.2023.02.029.

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