

Spatial heterogeneity and source-to-sink processes of heavy metals in a karst Pb-Zn mining area

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Abstract

Purpose: To identify heavy-metal source-to-sink relationships in a karst mining area without treating qualitative associations as quantitative source contributions.

Methods: We developed a transparent cross-media evidence framework combining spatial-unit contrasts, distance gradients, multivariate associations, and sediment-water responses. The framework was applied to 66 composite surface-soil samples collected at 0–20 cm, 13 channel-sediment samples, 24 surface-water samples, and 7 groundwater samples from a Pb-Zn mining area in southwestern China.

Results: Cd, Cu, Pb, and Zn met three solid-phase criteria for a mining-river association: a mine/plain mean ratio of at least 2, both distance correlations at or below -0.30 , and a PC1 loading of at least 0.70 . Their mine/plain ratios were 5.70, 4.53, 13.90, and 7.08, respectively. Cr and Ni were more strongly associated with karst background and pedogenic controls, whereas As showed mixed behavior. High sediment/soil ratios for Zn (6.29) and Cd (5.78), together with high surface-water/groundwater ratios for Zn (29.55) and Cd (32.07), supported particulate storage and a subsurface attenuation hypothesis.

Conclusion: The framework identifies a mine source, a river-sediment pathway, and localized plain accumulation while distinguishing evidence-supported inference from unmeasured fluxes. It provides a reproducible intermediate approach for subsequent isotope tracing, event monitoring, and calibrated transport modelling.

1 Introduction

Mining activities introduce metal-bearing waste rock, tailings, mine drainage, and readily erodible particles into connected soil-water systems. Mining contamination is therefore not only a concentration problem but also a mass-transfer problem: source areas release metals, pathways transport them, and receiving units temporarily store, attenuate, or redistribute them. Reviews of environmental contamination emphasize that heavy metals persist in soils and waters and that sources, transport, and remediation contexts should be distinguished (Briffa et al. 2020; Xu et al. 2024). Reviews of mining areas in China further show that Cd, Pb, Zn, Cu, and As are often controlled jointly by geological background and anthropogenic inputs, and that whole-area averages alone provide limited insight into transport processes (Li et al. 2014; Chen et al. 2015; Hu et al. 2020; Wu et al. 2022).

Karst terrain further complicates source discrimination. Weathering of carbonate rocks can produce naturally elevated metal backgrounds, whereas thin soils, fractures, and conduits can promote rapid surface-water/groundwater exchange. Studies in karst areas indicate that combining spatial patterns with auxiliary variables, source models, or isotopic evidence is more reliable than assigning sources from a single principal component alone (Huang et al. 2021; Zhang et al. 2021; Zhang et al. 2022; Tai et al. 2026; Wang et al. 2026).

Existing methods are moving from qualitative identification toward multi-evidence and quantitative apportionment. GIS, self-organizing maps, and PMF can estimate source contributions when receptor

samples are sufficient and uncertainty is evaluated (Li et al. 2022b; Wang et al. 2021). Combining PMF with a geographic detector can link factor distributions to parent material, roads, and industrial activity (Fei et al. 2020). Sediment fingerprinting can further discriminate source areas and test mixing models (Sellier et al. 2021), while hydrogeochemical monitoring can compare source contributions across discharge conditions (Onnis et al. 2023). These methods generally require source samples, rainfall-event observations, auxiliary spatial data, or tracers, which are not available for every historical survey.

Transport models provide another route from concentration patterns to process understanding. SWAT-HM can represent partitioning, erosion, runoff, deposition, resuspension, and burial at the catchment scale, but calibration requires continuous hydrological, sediment, and metal observations (Meng et al. 2018). Rainfall-runoff studies likewise stress the need to distinguish dissolved from particle-bound transport and to prioritize event monitoring (Qiao et al. 2023). In karst water systems, filtered and total-dissolved concentrations together with stable isotopes can help distinguish dissolved inputs, particulate remobilization, and surface-water/groundwater exchange (Qin et al. 2021). Joint studies of river water and sediments also show that both media should be considered when evaluating metal mobility (Islam et al. 2015).

This study addresses a narrower, testable question: can multiple mutually supportive observations from a single spatial survey divide a connected mining area into source, transition, transport-pathway, and receiving units? We therefore developed a cross-media evidence framework comprising spatial-unit contrasts, distance gradients, multivariate associations, and sediment-surface-water-groundwater responses. The framework is a transparent screening and synthesis tool; it does not replace PMF, isotopic source apportionment, or calibrated transport models. Specifically, we aimed to (1) quantify metal heterogeneity in the mining-area, midslope, footslope, and plain units; (2) formalize evidence criteria for the Cd-Pb-Zn-Cu mining-river association; (3) distinguish karst-pedogenic control on Cr-Ni from mixed behavior of As; and (4) use cross-media contrasts to evaluate pathway storage and subsurface attenuation while identifying observations needed for quantitative validation.

2 Materials and methods

2.1 Study area

The study area is a peak-cluster karst mining area in southwestern China affected by historical Pb-Zn mining. The area contains carbonate-rock terrain, discontinuous soil cover, interconnected river corridors, and fracture-conduit groundwater pathways. Ore-bearing strata and mine-related deposits occur in the upper part of the catchment, while the midslope, footslope, and plain units are distributed sequentially along the connected terrain. We treated the four units as components of the same mining area rather than independent land-use classes. Because the area remains under environmental management, specific mine and village names are not disclosed; the regional setting, hydrological connectivity, and sampling geometry needed for analysis are retained. For spatial comparison, the study domain was divided into plain (I), footslope (II), midslope (III), and mining-area (IV) units. This order follows topographic position and does not indicate a pollution ranking.

2.2 Sample collection and chemical analysis

The dataset comprised 66 composite surface-soil samples, 13 channel-sediment samples, 24 surface-water samples, and 7 groundwater samples. Soils and sediments were collected in 2022. Surface soils (0–20 cm) were collected following the technical principles of HJ/T 166–2004; at each point, 3–5 subsamples were collected within a 20–50 m radius, composited in equal mass after removing visible roots and stones, and retained at approximately 1 kg. The 66 soil samples comprised 9 from the mining area, 5 from the midslope, 6 from the footslope, and 46 from the plain. Channel sediments were collected along the river corridor by combining three grab samples at each site, with the upper 0–10 cm retained. Water sampling followed HJ 494–2009. Samples were acidified to $\text{pH} < 2$, refrigerated at 4 degrees C, filtered through 0.45- μm membranes, and analyzed within one week. Soil and sediment samples were air-dried, coarse particles removed, and ground with nonmetallic tools before sequentially passing 0.830-mm and 0.149-mm nylon sieves. Soil pH was measured at a soil-to-water ratio of 1:2.5. Clay content (0–2 μm) was measured by laser particle-size analysis after removal of carbonates and organic matter. For total-metal determination, 0.2 g of < 0.149-mm material was microwave-digested with HNO_3 , HCl , and HF . Cr, Cu, Ni, Pb, and Zn were measured by ICP-OES, while As and Cd were measured by ICP-MS; all seven elements in water were measured by ICP-MS. Each batch included 20% duplicate samples, procedural blanks, and the certified reference soil GBW 07404. Recoveries ranged from 73.67 to 126.71%, and duplicate results were averaged before statistical analysis.

2.3 Cross-media evidence framework and statistical analysis

The framework was designed to identify converging source-to-sink evidence rather than estimate source mass fractions. It comprised four evidence domains: (i) spatial-unit evidence, comparing means among the mining area, midslope, footslope, and plain and using P90 to identify high values within the dataset; (ii) distance evidence, calculating Spearman rank correlations between concentrations and distances from the mine and river corridor; (iii) association evidence, using Varimax-rotated principal component analysis (PCA) to identify co-varying element groups; and (iv) cross-media evidence, comparing overall sediment/soil and surface-water/groundwater means to identify possible particulate storage and subsurface attenuation. An element was classified as part of the mining-river association when it met all three solid-phase criteria: a mine/plain mean ratio of at least 2, both distance correlations at or below -0.30 , and a PC1 loading of at least 0.70. These ratio and correlation thresholds were dataset-specific screening rules, not regulatory criteria. The criteria were specified before interpretation and were intended to distinguish clear source-area contrasts from small mean differences. Cross-media ratios were used only as pathway evidence and were not interpreted as fluxes because discharge, sediment load, and paired event observations were unavailable. Descriptive statistics included the minimum, median, arithmetic mean, P75, P90, maximum, and coefficient of variation. Because some elements were right-skewed, the overall mean was used for description only; spatial interpretation relied mainly on unit-specific means, medians, percentiles, and individual high values. Metal concentrations were $\log_{10}$ -transformed and standardized before PCA; the first three rotated components explained 90.49% of the variance. Each

interpretation was required to be supported by at least two evidence domains, and process arrows not directly measured were labeled as hypotheses or potential pathways.

As shown in Fig. 1, the study area was divided by topographic position into four spatial units: plain, footslope, midslope, and mining area. The figure retains the study boundary, river corridor, and sampling units needed to interpret the analysis.

3 Results

3.1 Overall metal concentrations in surface soils

All surface-soil data showed clear elemental differences and spatial heterogeneity (Table 1). Pb had the highest mean concentration ($960.38 \text{ mg kg}^{-1}$) and the largest CV (3.19), followed by Zn ($662.12 \text{ mg kg}^{-1}$, CV 2.24), Cu (65.67 mg kg^{-1} , CV 1.78), and Cd (3.14 mg kg^{-1} , CV 1.59). As, Cr, and Ni had lower variability, with CVs of 0.99, 0.36, and 0.50, respectively. The means of Pb, Zn, and Cu were markedly higher than their medians, and their P90 values were high, indicating that a small number of high-value samples strongly influenced the overall means. Accordingly, overall statistics were used to describe the dataset, whereas spatial interpretation was based mainly on statistics for the four units. P90 values for Pb and Zn were 1352.90 and $1306.16 \text{ mg kg}^{-1}$, respectively, and their maxima reached 18078.16 and $10977.15 \text{ mg kg}^{-1}$; the maximum Cu concentration was $931.23 \text{ mg kg}^{-1}$. These extreme values were not treated as representative concentrations for the whole mining area; instead, they were interpreted as evidence of local enrichment together with sampling-unit, distance-gradient, and multivariate evidence.

3.2 Contrasts among the four spatial units

The four-unit comparison separated the source signal from the lower-concentration downstream transition units (Table 1). The mining area had the highest mean concentrations of Pb, Zn, Cu, and Cd, at 4867.62 , 2443.79 , 192.20 , and 10.32 mg kg^{-1} , respectively. Relative to the plain, the mine/plain mean ratios were 13.90, 7.08, 4.53, and 5.70, respectively. The midslope and footslope units generally showed intermediate concentrations, although the elements did not all follow a strictly monotonic pattern. Mean Cd, Cu, Pb, and Zn concentrations were lower in the plain, but some plain samples still had high P90 and maximum values. These results indicate a clear source-area contrast, but incomplete spatial attenuation; the pattern cannot be attributed to a single distance effect.

3.3 Distance gradients and multivariate associations

Cd, Cu, Pb, and Zn were negatively correlated with both distance from the mine and distance from the river corridor (Table 2a). The two correlation coefficients were -0.40 and -0.46 for Cd, -0.39 and -0.45 for Cu, -0.48 and -0.54 for Pb, and -0.50 and -0.56 for Zn. This pattern is consistent with combined effects of mine proximity and river-corridor transport. In contrast, Cr and Ni had weak or positive distance correlations and were positively correlated with the 0–2 micrometer clay fraction, suggesting stronger control by geological material, particle size, and pedogenesis. As was positively correlated with clay content but showed no consistent distance gradient. The first three Varimax-rotated components

explained 90.49% of the variance (Table 2b). PC1 explained 53.17% of the variance and had high loadings for Cd, Cu, Pb, and Zn (0.929, 0.912, 0.941, and 0.974, respectively). PC2 explained 29.88% and was dominated by Cr (0.751) and Ni (0.885), whereas PC3 explained 7.44% and was dominated by As (0.955). Together with the spatial-unit contrasts and distance gradients, these results indicate a relatively consistent Cd-Cu-Pb-Zn mining-river association, a karst-background-pedogenic Cr-Ni association, and mixed behavior of As. The evidence categories in Table 2c are qualitative and indicate convergence among observations; they do not provide source contribution percentages.

3.4 Cross-media responses

The cross-media data provided two supplementary lines of evidence (Table 3). Mean Zn and Pb concentrations in channel sediments were 4164.40 and 1106.03 mg kg⁻¹, respectively, and mean Cd was 18.15 mg kg⁻¹. Sediment/overall surface-soil mean ratios were 6.29 for Zn and 5.78 for Cd, whereas ratios for As, Cr, and Ni were below or close to 1. This contrast is consistent with preferential enrichment of particle-bound Zn and Cd in the river corridor. Because sediment loads, discharge, and repeated event observations were unavailable, we interpreted the ratios as storage signals rather than transport fluxes. Mean surface-water concentrations of Zn and Cd were 737.05 and 8.66 µg L⁻¹, respectively, compared with 24.94 and 0.27 µg L⁻¹ in groundwater; the corresponding surface-water/groundwater ratios were 29.55 and 32.07. These differences may reflect dilution, adsorption, precipitation, or other attenuation processes during subsurface transfer, but the available data cannot distinguish among these mechanisms. The water ratio for Pb was lower (3.90), while the surface-water/groundwater ratio for Cr was below 1 (0.27), showing that cross-media responses were element-specific rather than uniform across the mining area.

Table 1
Descriptive statistics and mean concentrations in the four spatial units

(a) Overall statistics for all surface-soil samples (n = 66)								
Element	Minimum	Median	Mean	P75	P90	Maximum	CV	Unit
As	0.19	26.10	42.40	41.06	109.98	162.34	0.99	mg kg ⁻¹
Cd	0.01	1.37	3.14	4.17	5.69	28.24	1.59	mg kg ⁻¹
Cr	18.49	61.64	58.05	74.06	83.03	115.19	0.36	mg kg ⁻¹
Cu	10.08	36.55	65.67	66.17	120.31	931.23	1.78	mg kg ⁻¹
Ni	6.79	19.19	20.96	24.73	32.10	56.26	0.50	mg kg ⁻¹
Pb	3.33	110.37	960.38	427.13	1352.90	18078.16	3.19	mg kg ⁻¹
Zn	31.88	198.11	662.12	736.15	1306.16	10977.15	2.24	mg kg ⁻¹
(b) Mean heavy-metal concentrations in the four spatial units (mg kg⁻¹)								
Spatial unit	n	As	Cd	Cr	Cu	Ni	Pb	Zn
Mining area	9	44.78	10.32	62.21	192.20	29.32	4867.62	2443.79
Midslope	5	27.91	3.36	54.65	54.52	18.27	368.06	649.32
Footslope	6	26.09	2.37	52.02	63.40	18.38	271.00	428.43
Plain	46	45.63	1.81	58.39	42.42	19.95	350.22	345.40
Note: Overall statistics describe the distribution of the 66 samples; spatial-process interpretation is based mainly on statistics for the four units.								

Table 2
Environmental correlations, principal component loadings and source-to-sink evidence

(a) Spearman rank correlations between heavy metals and environmental variables				
Element	pH	0–2 µm clay	Distance from mine/river	Interpretation
As	0.19	0.38	-0.06 / 0.20	Positive clay association; no consistent distance gradient
Cd	0.20	-0.48	-0.40 / -0.46	Both distance correlations negative
Cr	0.27	0.40	0.17 / 0.26	Clay association; weak distance effect
Cu	0.19	-0.32	-0.39 / -0.45	No clear distance effect
Ni	0.38	0.22	0.03 / 0.01	Both distance correlations negative
Pb	0.16	-0.51	-0.48 / -0.54	Both distance correlations negative
Zn	0.19	-0.51	-0.50 / -0.56	Both distance correlations negative
(b) Varimax-rotated principal component loadings				
Element	PC1	PC2	PC3	Interpretation
As	0.023	0.257	0.955	Mixed source / fine-particle control
Cd	0.929	0.033	0.087	Mining-river association
Cr	-0.178	0.751	0.489	Karst background / particle-size control
Cu	0.912	0.261	0.043	Mining-river association
Ni	0.279	0.885	0.181	Karst background / pedogenic process
Pb	0.941	-0.060	-0.080	Mining-river association
Zn	0.974	0.075	-0.045	Mining-river association
(c) Cross-media evidence screening				
Element	Mine/plain mean ratio	Distance criterion	PC1 criterion	Evidence-based interpretation
As	0.98	Not met	Not met	Mixed source; influenced by fine particles and local
Note: The source criteria are transparent screening rules developed for this study, not regulatory thresholds. Members of the mining-river association simultaneously meet a mine/plain mean ratio of at least 2, both distance correlations at or below – 0.30, and a PC1 loading of at least 0.70.				

(a) Spearman rank correlations between heavy metals and environmental variables				
Element	pH	0–2 µm clay	Distance from mine/river	Interpretation
				conditions
Cd	5.70	Met	Met (0.929)	Mining-river association member; requires pathway evidence
Cr	1.07	Not met	Not met (PC2 = 0.751)	Karst background and particle-size control
Cu	4.53	Met	Met (0.912)	Mining-river association member; requires pathway evidence
Ni	1.47	Not met	Not met (PC2 = 0.885)	Karst background and pedogenic process
Pb	13.90	Met	Met (0.941)	Mining-river association member; requires pathway evidence
Zn	7.08	Met	Met (0.974)	Mining-river association member; requires pathway evidence
Note: The source criteria are transparent screening rules developed for this study, not regulatory thresholds. Members of the mining-river association simultaneously meet a mine/plain mean ratio of at least 2, both distance correlations at or below – 0.30, and a PC1 loading of at least 0.70.				

Table 3
Mean concentrations and cross-media ratios of heavy metals

(a) Mean heavy-metal concentrations in river sediments and water				
Element	Sediment (mg kg⁻¹)	Surface water (µg L⁻¹)	Groundwater (µg L⁻¹)	Surface water/groundwater
As	13.29	0.59	0.35	1.69
Cd	18.15	8.66	0.27	32.07
Cr	26.92	0.11	0.41	0.27
Cu	92.67	0.36	0.43	0.84
Ni	20.03	1.72	0.64	2.69
Pb	1106.03	1.17	0.30	3.90
Zn	4164.40	737.05	24.94	29.55
(b) Sediment/surface-soil mean ratios				
Element	Sediment/soil ratio	Element	Sediment/soil ratio	Interpretation
As	0.31	Cd	5.78	Zn and Cd show clear sediment enrichment
Cr	0.46	Cu	1.41	Cu is slightly above the overall soil mean
Ni	0.96	Pb	1.15	Ni is close to 1; Pb is slightly above 1
Zn	6.29			Zn shows clear sediment enrichment
Note: Cross-media ratios are descriptive comparisons of means and do not represent transport fluxes. Discharge, sediment loads, and paired rainfall-event samples were not available.				

4 Discussion

4.1 Scope and applicability of the cross-media evidence framework

The main contribution of this study is methodological transparency rather than a new receptor model. When the required data are available, PMF, self-organizing maps, geographic detectors, isotopic tracing, and uncertainty analysis can support stronger quantitative source apportionment (Li et al. 2022b; Wang et al. 2021; Fei et al. 2020). The present dataset came from a single spatial survey. Although it included multiple media, it lacked source end-members, isotope ratios, event-scale discharge, and sediment fluxes. Direct application of a quantitative source model could therefore produce a level of precision not

supported by the data. Our framework links each inference to an explicit evidence domain and labels process steps that were not directly observed as hypotheses.

4.2 Distinguishing the mining-river association from karst background

Cd, Cu, Pb, and Zn satisfied all three prespecified solid-phase criteria. A high mine/plain mean ratio indicates a strong source-area contrast; negative distance gradients indicate decreasing influence with increasing distance from the mine and river corridor; and high PC1 loadings indicate co-variation among the four elements in the soil dataset. Converging evidence is stronger than any single result and is consistent with a mining-river association in mine-impacted catchments (Pan et al. 2024; Qiao et al. 2023). However, the results cannot convert historical deposits, mine drainage, bank erosion, and flood deposition into separate contribution fractions. The more defensible conclusion is a source-pathway association that still requires quantitative validation. The Cr-Ni grouping should not be interpreted as mining-source evidence simply because the elements co-occur in the same samples. Their positive association with fine particles, weak distance gradients, and PC2 loadings are more consistent with lithological background and pedogenic redistribution. Karst studies indicate that high geological backgrounds can overlap with anthropogenic inputs and that total concentrations alone may overestimate anthropogenic contributions (Huang et al. 2021; Li et al. 2022a; Zhang et al. 2021; Zhang et al. 2022). The source of As is more difficult to resolve: its highest PC3 loading and positive clay association are consistent with fine-particle retention, but the absence of a clear spatial gradient means that geological and local-transport explanations cannot be excluded. Source fractions would require source end-members, isotopic tracing, or a receptor model with uncertainty evaluation.

4.3 River-corridor storage and subsurface transfer

The sediment results support a dual role for the river corridor as both a temporary sink and a potential secondary source. Sediment fingerprinting studies emphasize the need for conservative tracers, particle-size correction, and preferably mixing-model validation for source discrimination (Collins et al. 2005; Laceby et al. 2017; Sellier et al. 2021; Xiao et al. 2021). Because these measurements were not conducted here, high sediment/soil ratios can only be interpreted as enrichment evidence and cannot establish source mass fractions. Reviews of risk evaluation and remediation also emphasize that metal mobility and treatment depend on metal form, site conditions, and the regulatory context (Wang et al. 2022). The high Zn and Cd surface-water/groundwater ratios are compatible with attenuation during subsurface transfer. This interpretation is consistent with studies of elemental partitioning and surface-water/groundwater exchange in karst systems, but total concentrations alone cannot resolve the mechanisms (Liu et al. 2019; Qin et al. 2021). Figure 2 summarizes the inferred relationships among the source area, transport pathways, and receiving units.

4.4 Validation needs, management implications, and limitations

A calibrated SWAT-HM model could test whether erosion, runoff, deposition, resuspension, and burial processes reproduce the observed distribution, but such calibration requires continuous hydrological, sediment, and metal observations (Meng et al. 2018). Similarly, source apportionment across flow conditions requires comparison of different discharge events and may require tracer injections or high-frequency sampling (Onnis et al. 2023). Vertical-transport simulations show that soil texture, sorption affinity, and rainfall intensity jointly affect metal movement through soil profiles, confirming that a surface-soil spatial survey cannot replace profile and event observations (Xu et al. 2024b). The framework identifies priority observations for future work: source end-members from mine wastes and parent materials, dissolved and particle-bound metals, rainfall-event runoff, sediment loads, groundwater-surface-water exchange, and repeated profiles along the river corridor. With these additions, the qualitative pathway model could be developed into a mass-balance or uncertainty-evaluated analysis. The available evidence supports staged management: the mining area should be treated as a priority source-control zone; the river corridor should be monitored as a transport and sediment-storage zone; and the plain should be screened for local high-value patches rather than represented by one overall mean. This zoning logic is consistent with combining spatial source apportionment and process-based transport analysis (Qu et al. 2020; Wang et al. 2021). The main limitations are the cross-sectional survey design, unbalanced sample sizes among the four units, lack of speciation and isotope data, and absence of event-scale fluxes. We therefore do not assess health risks or report quantitative source percentages. Instead, we provide a reproducible evidence-based zoning framework for the next stage of sampling and modelling. Future evaluations of stabilization and disposal should also include post-treatment metal speciation and material safety (Chen et al. 2024).

The cross-media evidence framework for heavy-metal source-pathway-sink processes was synthesized from spatial-unit contrasts, distance gradients, PCA groupings, and cross-media ratios (Fig. 2).

5 Conclusions

1. Comparisons among the four spatial units showed marked metal heterogeneity. Pb, Zn, Cu, and Cd were most enriched in the mining area, with mine/plain mean ratios of 13.90, 7.08, 4.53, and 5.70, respectively. The plain still contained local high-value samples.
2. Cd, Cu, Pb, and Zn showed clear mine/plain contrasts, decreasing gradients with distance from the mine and river, and high PC1 loadings, forming a converging mining-river association. Cr-Ni were more consistent with karst-background-pedogenic control, whereas As should retain a mixed-source interpretation.
3. Zn and Cd were enriched in channel sediments relative to the overall surface-soil means, and their surface-water concentrations were 29.55 and 32.07 times the groundwater concentrations, respectively. These contrasts support hypotheses of sediment storage and subsurface attenuation but cannot replace flux estimates.
4. The cross-media evidence framework converts a single spatial survey into a transparent source-to-sink screening result, separates qualitative inference from quantitative source apportionment, and

identifies priority observations for isotope tracing, event monitoring, and calibrated transport modelling.

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Author Contribution

Author Contributions Statement K.L. prepared the original draft and revised the manuscript. Q.L. collected and verified the data. J.D. reviewed and edited the manuscript and supervised the research. K.B. revised and updated the manuscript. Z.W. provided access to the analytical instruments and guidance on their operation. D.F. reviewed the manuscript and coordinated the project. All authors contributed to the interpretation of the results and critically revised the manuscript for important intellectual content. All authors approved the version to be published and agree to be accountable for all aspects of the work.

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Data availability:

The datasets generated and/or analyzed during the current study are available from the corresponding author on reasonable request.

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Figures

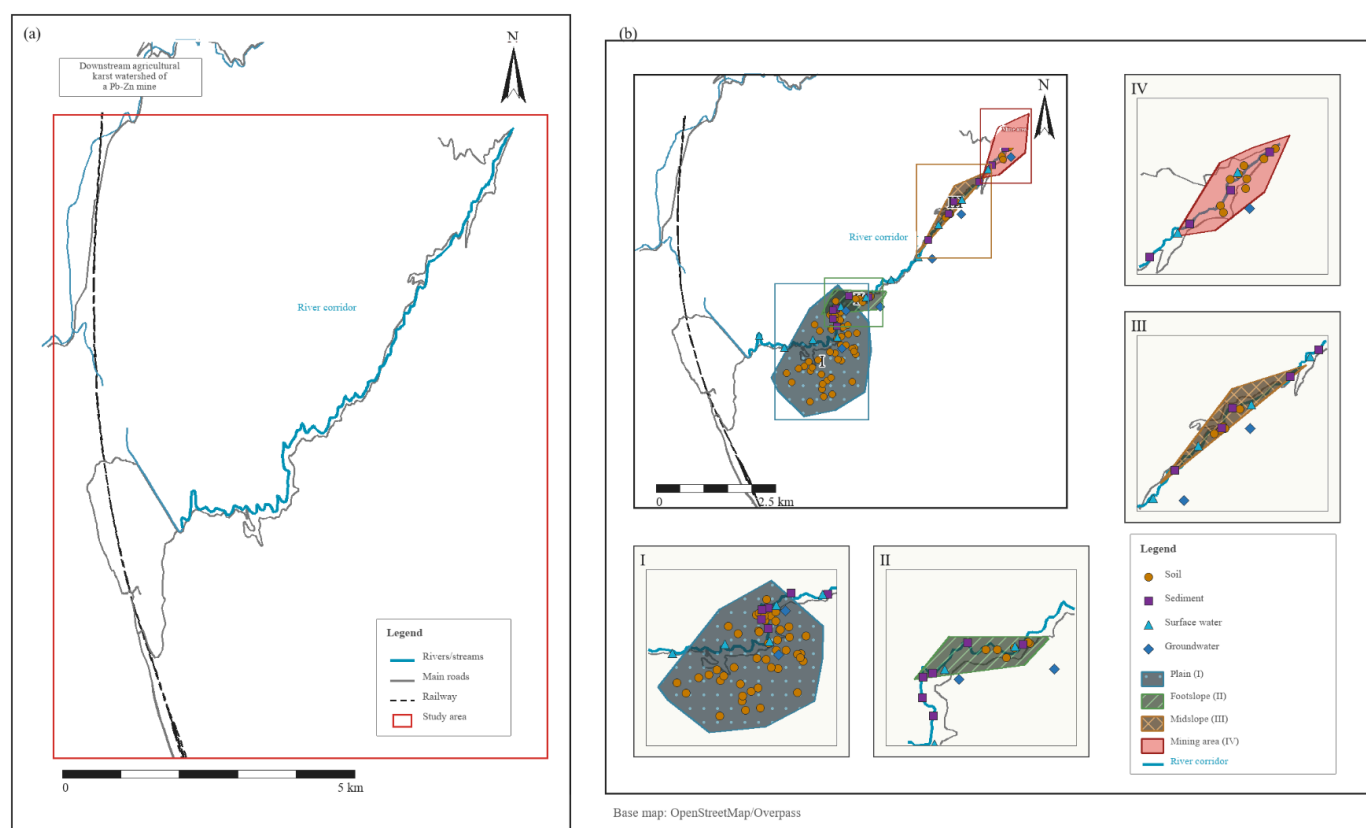


Figure 1

Study area and spatial sampling units

Conceptual Model of Heavy-Metal Source-Pathway-Sink Processes

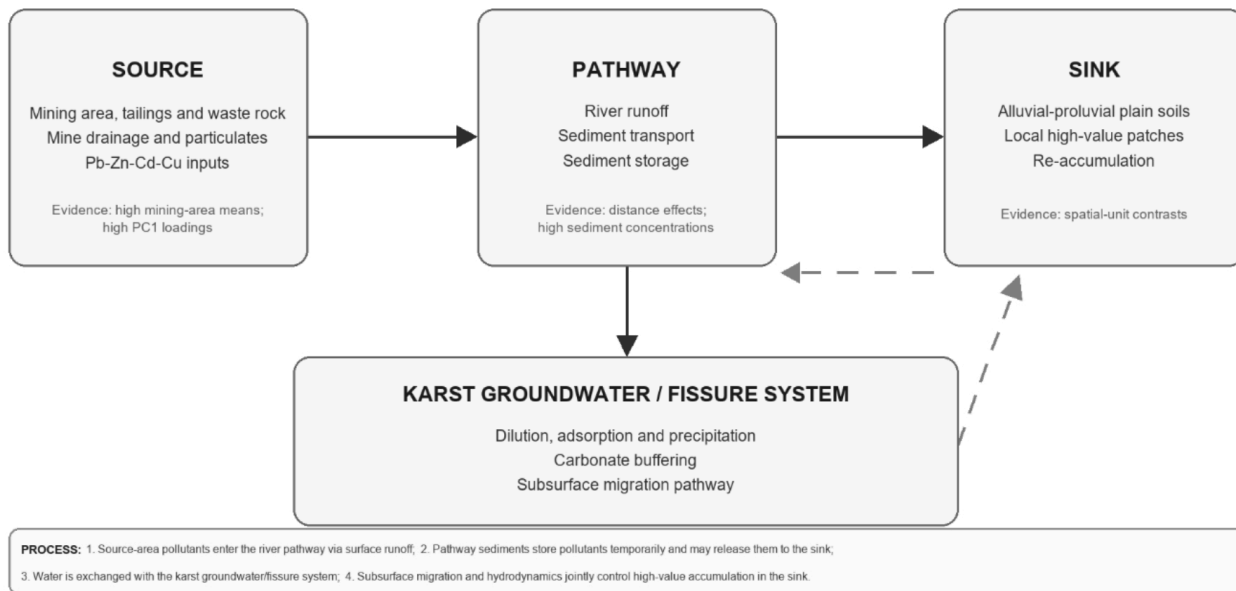


FIGURE LOGIC: Spatial-unit contrasts, distance effects, PCA groupings and multi-media responses jointly support the source-pathway-sink interpretation.

Figure 2

Cross-media evidence framework for heavy-metal source-to-sink processes