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Effect of land use conversion on heavy metals and magnetic minerals on water reservoir riparian soils

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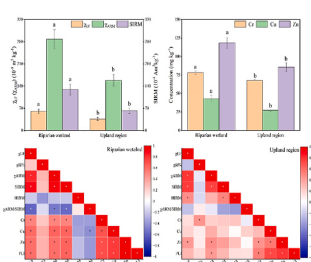
HIGHLIGHTS

- Reservoir riparian wetland soils exhibited magnetic enhancement.
- Coarse magnetic grains with metal elements present in riparian wetlands.
- χ_{LF}, χ_{ARM} and SIRM were strongly correlated with pollution load index in riparian wetlands.

GRAPHICAL ABSTRACT



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ABSTRACT

Spatial hydrological alterations seem to influence the variability of soil-bound magnetic particles and heavy metals in water reservoir riparian wetlands (RW). To date, applying geochemical analysis with magnetic techniques to assess heavy metal pollution is rarely practiced in RW soils. We studied the magnetic properties and heavy metals, including Cu, Cr, and Zn, of topsoils in RW and the adjacent upland regions (UR, as control) in the Three Gorges Reservoir, China. Potentially elevated low-frequency mass magnetic susceptibility (χ_{LF}), anhysteretic remanent magnetization susceptibility (χ_{ARM}), isothermal remanent magnetism, and all selected heavy metals were found in RW. The grain size of the magnetic minerals was coarser in RW than that in UR. The pollution load index (PLI) of the studied samples was 1.18 ± 0.12 and 1.04 ± 0.21 in RW and UR, respectively. PLI and concentrations of Cu, Cr, and Zn were positively correlated with χ_{LF}, χ_{ARM}, and isothermal remanent magnetism in RW, whereas no clear linkages were observed between PLI and isothermal remanent magnetism in UR. This finding reveals that hydrological alterations increased the magnetic enhancement and heavy metal enrichment in RW. We find that magnetic proxies of soils could trace the concentration of selected anthropogenic heavy metals and their pollution level in RW.

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1. Introduction

Reservoir riparian wetlands (RW), the interface between low- and high-water-level areas (Jiang et al. 2017), are strongly altered by dam impoundment and hydrological regulation (Liu et al. 2014; Ye et al. 2020; Zhang et al. 2021). The abundance of trace elements in RW soils is typically higher than that in the adjacent upland soils (Jiang et al. 2017; Ye et al. 2020; Yang and Xie, 2022) but lower than that in the reservoir sediments (Zhang et al. 2021). Compared with the soil before submergence, submerged soil seems to exhibit a higher concentration of trace elements (Ye et al. 2011; Liu et al. 2014). Some of the elevated elements of anthropogenic inputs prevail in the non-residual fraction (Zhu et al. 2019; Zhang et al. 2021). Excessive heavy metals in soils increase the levels of these elements in foodstuffs and, ultimately, the human body (Davodpour et al. 2019; Sabet Aghlidi et al. 2020). Thus, quantifying the abundance of soil-bound heavy metals could provide useful information for the prevention and control of soil pollution (Liu et al. 2014; Davodpour et al. 2019; Zhang et al. 2021).

The amount of soil-bound magnetic minerals in poorly drained sites is possibly influenced by the hydrological alteration frequency (Chudaničová et al. 2016) and duration (Krzywicka et al. 2017). Soil submerged for an extended period exhibit altered phases of iron-bearing minerals (Asgari et al. 2018; Huang et al. 2018) or dissolved ferrimagnetic minerals of superparamagnetic grains (Grimley et al. 2004; Chudaničová et al. 2016). The magnetic susceptibility of poorly drained sites is lower than that of well-drained areas (Valaee et al. 2016; Asgari et al. 2018; Shirzaditabar and Heck, 2021). Meanwhile, the higher values of magnetic susceptibility in poorly drained soils are linked to soil deposition or the presence of anthropogenic particles (Grimley et al. 2004). The sources of magnetic minerals in RW are possibly the bedrock, pedogenesis, and external anthropogenic loading (Zhu et al. 2014; Valaee et al. 2016). Meanwhile, the levels of magnetic minerals in RW-adjacent upland regions (UR) are slightly affected by dam impoundment and periodic hydrological regulation (Zhu et al. 2014).

Many studies have focused on the relationship between heavy metal pollution and the enrichment of magnetic particles in urban dusts (Yang et al. 2021), wetland sediments (Ma et al. 2015; Yang et al. 2019, 2022), and industrial soils (Hu et al. 2022). Anthropogenic magnetic nanoparticles enriched with certain heavy metals have been characterized as magnetic spherules, aggregates, or angular shapes (Naimi and Ayoubi, 2013; Chaparro et al. 2020; Hu et al. 2022). In fact, the presence of soil-bound iron-rich spherical shapes in RW suggests the external anthropogenic loading of iron-bearing minerals (Zhu et al. 2014). Poor linkage between metal elements and magnetic phases has been recorded in RW soils, subject to periodic wet–dry cycles. In this study, magnetic signals of magnetic minerals and the abundance of Cr, Cu, and Zn in RW and UR along the shoreline of the Three Gorges Reservoir, China, were investigated. The aims were as follows: (1) estimate the abundance of magnetic minerals in RW and UR; (2) trace the possible origins of magnetic minerals in RW; and (3) study the correlation between magnetic signal enhancement and metal element enrichment in RW.

2. Materials and methods

2.1. Study sites and sampling

The study area, the Three Gorges Reservoir (29 16′–31 25′ N, 106°–110 10′ E) is located in the main channel of Yangtze River, China (Ye et al. 2020). The experimental area is characterized by a subtropical monsoon climate, with an average temperature of 18 °C (Zhang et al. 2021). The average precipitation is 1100 mm yr^{-1} (Zhang et al. 2021), with most of the annual precipitation occurring between April and October (Ye et al. 2020). The tested soil in the study region is classified as Regosols in the FAO taxonomy (Zhang et al. 2021). Annually, the reservoir water level varies between 145 m and 175 m above sea level (Ye et al. 2020; Yang and Xie, 2022). The area of RW in the Three Gorges Reservoir is 348.92 km^2 (Ye et al. 2020).

In the summer of 2016, soil samples were collected from 20 sites in RW and 20 sites in UR along the shorelines of the Three Gorges Reservoir (Fig. 1). At each site, triplicated subsamples of 0–20 cm were collected by a wooden shovel and mixed into a composite sample (Yang and Xie, 2022). The air-dried soil samples were sieved using a 0.149 mm nylon screen in the laboratory.

2.2. Magnetic measurement

Soil magnetic susceptibility was measured using a Bartington dual frequency MS2B sensor at 0.47 kHz (χ_{LF}) and 4.7 kHz (χ_{HF}) (Dearing, 1994). The anhysteretic remanent magnetization susceptibility (χ_{ARM}) is sensitive to the magnetic size of a stable single domain (Chudaničová et al. 2016); as such, quantification using a D-2000 alternating field demagnetizer (ASC Scientific, California, USA) with a direct-current biasing field of 0.05 mT and peak alternating field of 100 mT (Yang et al. 2022) was performed. Isothermal remanent magnetism was analyzed using an IM-10-30 Molspin Pulse Magnetizer (ASC Scientific, California, USA) and JR-6A magnetometer (Agico Led., Czech Republic). The sample was imparted at 1 T (SIRM) (Chudaničová et al. 2016), −100 mT (IRM_{-100}), and −300 mT (IRM_{-300}) (Yang et al. 2022). The frequency-dependent susceptibility percentage (χ_{fd}%), hard isothermal remanent magnetization (HIRM), S_{-100}, and S_{-300} were obtained (Oldfield, 1991; Chudaničová et al. 2016) using Equations (1)–(4):

$$\chi fd\% = \frac{\chi_{LF-\chi_{HF}}}{\chi_{HF}} \times 100 \quad (1)$$

$$HIRM = \frac{IRM_{-300} + SIRM}{2} \quad (2)$$

$$S_{-100} = \frac{IRM_{-100} + SIRM}{2SIRM} \quad (3)$$

$$S_{-300} = \frac{IRM_{-300} + SIRM}{2SIRM} \quad (4)$$

2.3. Chemical analysis

Soil-bound Cr, Cu, and Zn were digested using an acid mixture of $HClO_4$, HNO_3, and HF (1:1:2) (Ye et al. 2020; Yang and Xie, 2022), and quantified using a PerkinElmer NexION 350 inductively coupled plasma mass spectrometer. Reference materials (GSS-8) were used for accuracy determination. The average recoveries for Cr, Cu, and Zn were 94.53 ± 6.12%, 97.22 ± 7.71%, and 101.81 ± 5.04%, respectively and the detection limits were 0.02, 0.05, and 0.15 mg kg^{-1}.

2.4. Assessments of soil pollution

The pollution load index (PLI) indicates the toxicity status induced by metal elements (Wang et al. 2018; Chaparro et al. 2020) and was estimated using Equation (5):

$$PLI = \sqrt[n]{PI_1 \times PI_2 \times \ldots \times PI_n} \quad (5)$$

$$PI = \frac{C_i}{B_i} \quad (6)$$

where C_i is the concentration and B_i is the baseline concentration of metal i (Cr, Cu, and Zn: 78.03, 25.00, and 69.88 mg kg^{-1}, respectively) (Tang et al. 2008).

2.5. Data processes

Statistical data analysis was performed using Pro Origin 2013. Differences in soil Cr, Cu, and Zn and magnetic signals between RW and UR were examined using the statistical *t*-test with $p < 0.05$. Spearman

Fig. 1. Map of the sampling location in Three Gorges reservoir, China.

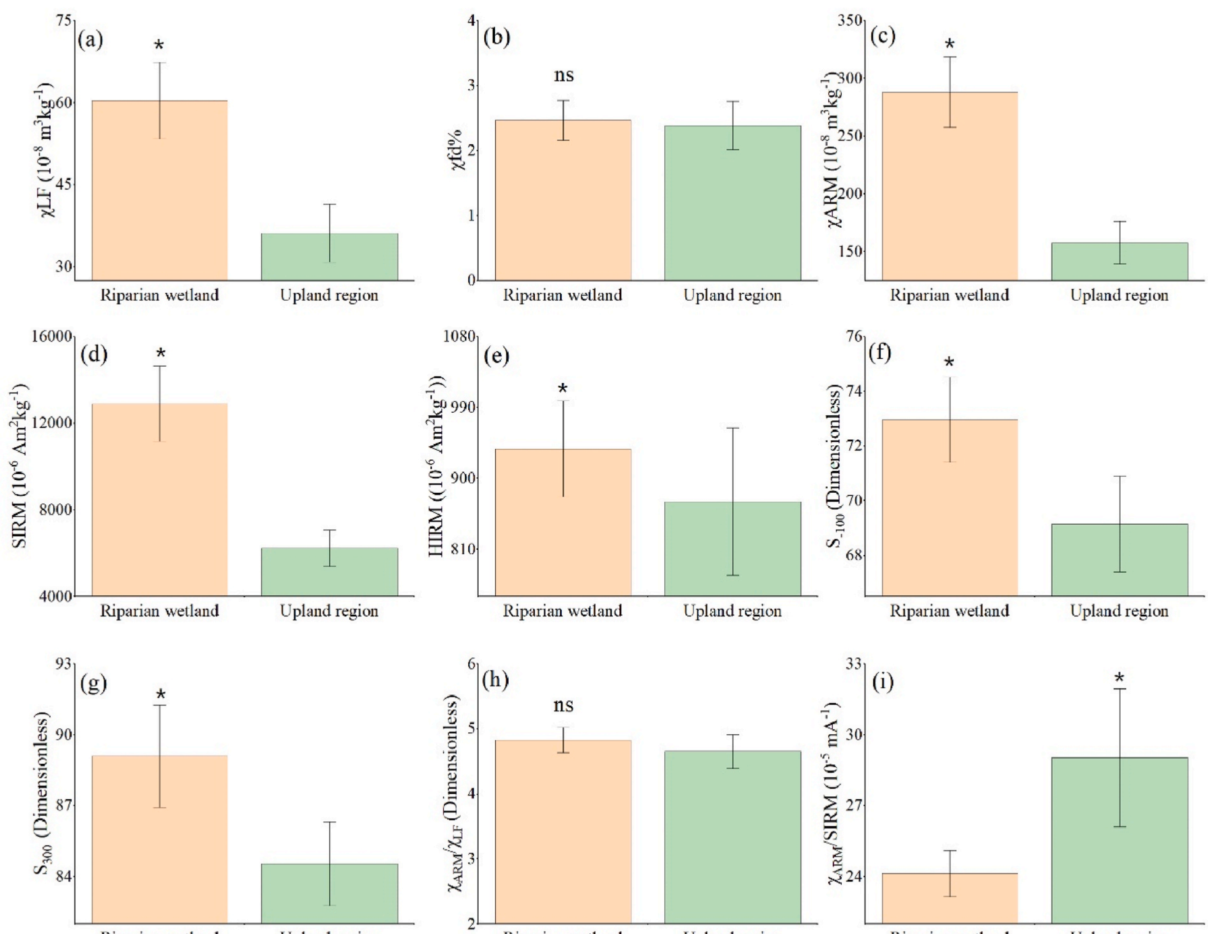

Fig. 2. Variations of magnetic signals in riparian wetland and upland regions.

correlation was used to examine the relationships between heavy metal abundance and magnetic signals.

3. Results

3.1. Distribution of magnetic parameters and heavy metals

Quantities χ_{LF}%, χ_{ARM}, SIRM, and HIRM were found to be significantly higher in RW than in UR (Fig. 2), whereas no significant difference in χ_{fd}% and χ_{ARM}/χ_{LF} between RW and UR was present. Ratio χ_{ARM}/SIRM was significantly lower in RW than in UR. Meanwhile, S_{-100}, S_{-300}, and SIRM/χ_{LF} were substantially higher in RW than those in UR. The averages of the Cr, Cu, and Zn concentrations in RW were substantively higher than those in UR (Fig. 3).

3.2. Correlations between magnetic parameters and metal element concentrations

In RW, χ_{LF}, χ_{ARM}, and SIRM were significantly correlated with Cr, Cu, and Zn. A weak correlation ($p > 0.05$) was evident between χ_{fd}% and (Cu, Zn) in RW (Fig. 4). Chromium, Cu, and Zn were negatively linearly correlated with χ_{ARM}/SIRM in RW, and there was a strong linear correlation between S_{-100}, S_{-300}, SIRM/χ_{LF}, χ_{ARM}/SIRM and the heavy metals Cr, Cu, and Zn. A significant positive correlation ($r = 0.40$–0.46, $p < 0.05$) was also found between χ_{LF} and Zn, χ_{fd}% and Cr, χ_{ARM} and Zn, and SIRM and Zn in UR (Fig. 5).

4. Discussion

4.1. Hydrological alterations increased coarse-grained magnetic minerals accumulated in RW

Our magnetic measurements indicated that the Entisols in RW in the study area showed the strongest magnetism of the studied soils. Regional studies have indicated that Entisols exhibit lower χ_{LF} (Zhu et al. 2014) and higher χ_{LF} on the surface than that in the subsurface at RW (Zhu et al. 2014). Elevated χ_{LF} values in the flooded sediments corresponded with the addition of ferrimagnetic phases during the hydrological alterations (Takesue et al. 2009). High χ_{LF} in RW is likely due to the accumulation of coarse-grained ferrimagnetic minerals resulting from water erosion, metal nanoparticle deposition, or new formation of second magnetic phases during the pedogenic process (Zhu et al. 2012, 2014). The χ_{LF} value in UR (Fig. 2) was comparable to that reported in previous studies (Zhu et al. 2012, 2014). Erosion of ferrimagnetic minerals on the upper sites is likely responsible for enhanced soil χ_{LF} on the lower sites (Saidati and Naima. 2023), as the abundance of ferrimagnetic phases in RW was higher than that in UR.

High χ_{fd}% was associated with the fine magnetic phases of pedogenic processes (Lu et al. 2012; Ayoubi et al. 2019), whereas low χ_{fd}% (<2%) was associated with the coarse magnetic phases of anthropogenic loading (Ma et al. 2015; Chaparro et al. 2020). Fine magnetic particles were dissolved owing to the long duration of submerged soil (Asgari et al. 2018). In RW and UR, χ_{fd}% of the studied samples was 2.46% ± 0.30% and 2.38% ± 0.37%, respectively (Fig. 2b), indicating that the

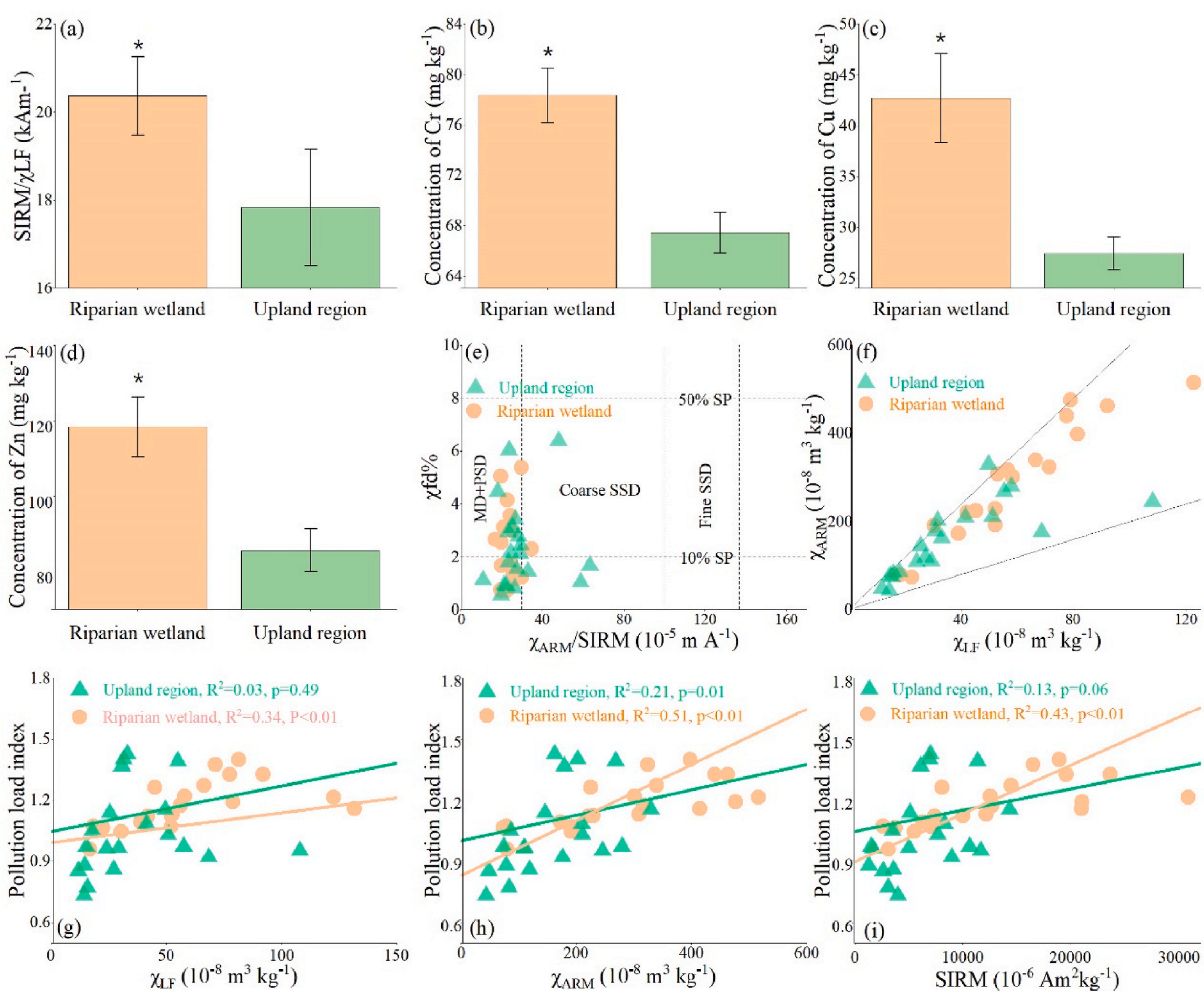

Fig. 3. Variations of SIRM/χ_{LF} (a), Cr, Cu, Zn (b–d), χ_{ARM}/SIRM *vs* $\chi_{fd\%}$, χ_{LF} *vs* χ_{ARM}, χ_{LF} *vs* PLI, χ_{LF} vs PLI and SIRM *vs* PLI.

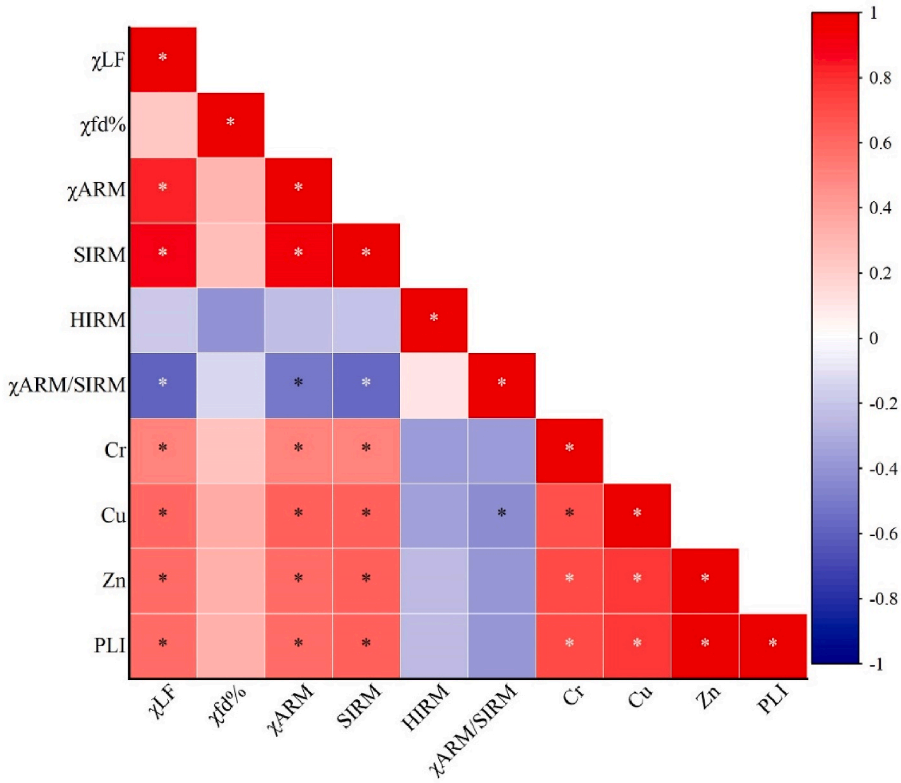

Fig. 4. Heatmap of magnetic parameters and heavy metals in reservoir riparian wetland.

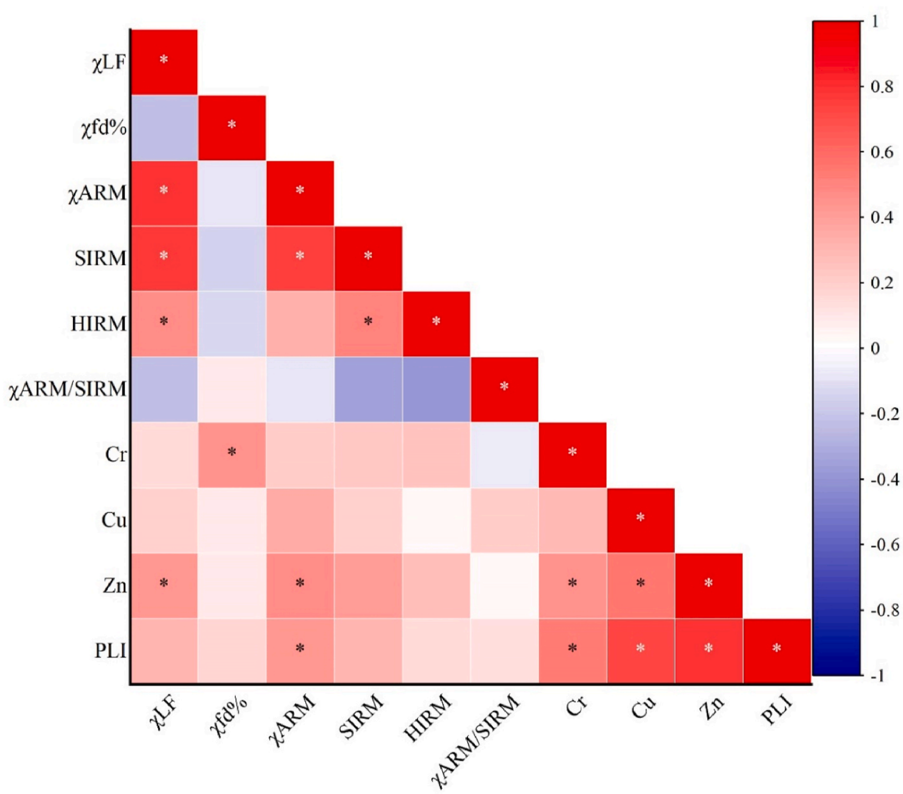

Fig. 5. Heatmap of magnetic parameters and heavy metals in upland region.

contribution of pedogenic ferrimagnetic minerals of superparamagnetic grains to χ_{LF} enhancement in UR and RW was not significantly different. The mean value for χ_{ARM}/SIRM was lower in RW than UR (Fig. 2i), and the sizes of the magnetic minerals in RW were coarser than that in UR. These results are partially in accordance with other studies (Mzuza et al. 2017a, 2017b). A plot of χ_{fd}% versus χ_{ARM}/SIRM (Fig. 3e) shows that the grain size of these magnetic minerals is within the multidomain and pseudo-single-domain ranges (King, 1982; Dearing et al. 1996). This finding indicates that the elevated χ_{LF} in RW was mainly associated with coarse-grained magnetic minerals.

S_{-300} was negatively correlated with HIRM in RW ($R^2 = 0.27$, $p = 0.02$) and UR ($R^2 = 0.14$, $p = 0.10$). S_{-300} and S_{-100} in RW were higher than that in UR (Fig. 2f and g), and large proportions of the soft coercivity phases were enriched in RW (Chudaničová et al. 2016). HIRM was substantially high in RW (Fig. 2e), and anti-ferromagnetic minerals were substantially accumulated in RW over that in UR. SIRM was insensitive to the superparamagnetic, paramagnetic, and diamagnetic phases (Chudaničová et al. 2016), and the reduction in SIRM/χ_{LF} increased with grain size from the single domain upward (Sari et al. 2016). SIRM/χ_{LF} varied from 13.70 kA m^{-1} to 26.70 kA m^{-1} (Fig. 3a), and the R^2 values between χ_{LF} and SIRM ranged between 0.65 and 0.87, further suggesting the contribution of ferrimagnetic phases to χ_{LF}.

4.2. External anthropogenic loading altered the coexistence of iron-rich magnetic particles and heavy metals

Lithological processes seem to have had a substantial contribution to the magnetic enhancement and metal enrichment in UR. The absence of correlation between magnetic variables and metal elements in metal-rich sites suggests that enhanced particular metal elements might have had little effect on magnetic signal enrichment (Yang et al. 2022). The adsorption of metals on the surface of magnetic particles is unstable (Naimi and Ayoubi, 2013; Ravisankar et al. 2018; Hu et al. 2022). Meanwhile, erosion of soil in UR promotes the non-residual fraction in soils deposited in RW or the Three Gorges Reservoir water (Zhu et al. 2019). The multi-sources of heavy metals and magnetic particles usually have poor coefficients of $\chi_{LF,}$ χ_{ARM}, SIRM, owing to their metal element concentrations (Wang et al. 2018; Yang et al. 2022). The geochemistry of the magnetic parameters of superparamagnetic size is most strongly dominated by lithological loading (Lu et al. 2012; Asgari et al. 2018). The positive relationship between χ_{fd}% and Cr concentration in UR (Fig. 5) possibly favored Cr-containing minerals, considering the natural origin of pedogenesis (Lu et al. 2012; Ayoubi et al. 2019).

Given that soil Cu, Zn, and Cr concentrations in RW were higher than those in pre-flooding sites (Ye et al. 2011) and UR (Wang et al. 2017; Zhang et al. 2019; Yang and Xie, 2022), the increased water level in RW contributed to the accumulation of metal elements (Fig. 3b–d). The positive linkages between particular metal elements and magnetic parameters in metal-enriched sites partially favored the incorporation or adsorption of these elements on the surface of iron-rich magnetic particles (Sabet Aghlidi et al. 2020; Hu et al. 2022). The addition of external iron-rich magnetic particles may cause soil magnetic enhancement and some enrichment of metal elements (Chudaničová et al. 2016; Li et al. 2021; Hu et al. 2022). Scanning electron microscopy images of anthropogenic iron-rich magnetic minerals are mainly spherical shapes (Naimi and Ayoubi, 2013; Yang et al. 2021; Hu et al. 2022), and these magnetic spheroids were also observed in RW (Zhu et al. 2014). The positive relationship with metals between χ_{LF}, χ_{ARM}, and SIRM (Fig. 4) favored the coexistence of external metal elements and magnetic phases (Li et al. 2021). Ratio χ_{ARM}/SIRM was negatively correlated with Cr, Cu, and Zn (Fig. 4), which were mostly adsorbed or combined with coarse-grained soft magnetic minerals. These results are in accordance with the findings of other studies (Yang et al. 2019; Li et al. 2021).

In 79% of the RW soil samples, PLI >1 (Fig. 3g). PLI is positively correlated with χ_{LF}, χ_{ARM}, and SIRM in RW (Fig. 4), whereas no significant correlation between isothermal remanent magnetism and PLI was observed in UR (Fig. 5). Magnetic particles are characterized by a chemical affinity for some heavy metals in the anthropogenic magnetic dust falls (Hu et al. 2022), lake sediments (Wang et al. 2018), and reservoir sediments (Ma et al. 2015); a significant positive correlation was found between PLI and magnetic parameters. The absence of a correlation between PLI and magnetic parameters might suggest variations in the degree of soil heavy metal pollution and specific circumstances (Hu et al. 2022). Thus, the magnetic parameters of soil could faithfully record the pollution of heavy metals in the water reservoir RW. Given that Cu, Zn, and Cr in RW were substantially higher than in UR in the Three Gorges Reservoir (Wang et al. 2017; Zhang et al. 2019; Qiu et al. 2021), the elevated soil Cu and Zn could enhance the toxicity of these elements to soil organisms. The sample size of RW in our study was small and concentrated in the western region, rather than covering all main channels and tributaries of the Three Gorges Reservoir. Further study is required to test whether the magnetic proxies of soil can be tracked by the possible migration pathway of heavy metals and their pollution levels in the water reservoir RW of the entire watershed.

5. Conclusions

Magnetic results revealed that χ_{LF}, χ_{ARM}, SIRM, and HIRM were significantly higher in RW than in UR. The magnetic signal of χ_{ARM}/SIRM was lower in RW than that in UR. The magnetic parameters of $\chi_{LF,}$ χ_{ARM}, SIRM, and χ_{ARM}/SIRM were found to be highly correlated with PLI and the abundances of Cr, Cu, and Zn in RW. In contrast, these magnetic signals showed a weak correlation with PLI and abundant heavy metals in UR. This finding highly indicates that Cr, Cu, and Zn are mainly associated with coarse magnetic minerals in RW. Furthermore, the magnetic-dependent concentration parameters χ_{LF}, χ_{ARM} and SIRM could semi-quantify the abundance of certain anthropogenic metal elements and their pollution levels in RW.

Credit author statement

Dan Yang: Conceptualization, Writing, Formal analysis, Funding acquisition. Xin Yang: Investigation, Writing, Formal, analysis; Na An: Data curation, Investigation, Methodology, Visualization; Zongqiang Xie: Validation, Supervision.

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.

Acknowledgments

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