Phytomanagement improves soil aggregation and ecological security near tailings

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Abstract

Aims

Revegetation is an effective measure to improve soil structure and nutrients in erosion-prone areas. However, little is known about the impact of diverse phytomanagement of revegetation on soil quality and ecological security near tailings.

Methods

We investigated the water-stability and soil aggregate nutrients and assessed the associated risk of heavy metal contamination under multiple phytomanagement (natural grassland, artificial forest, and artificial forest mixed with shrubs and herbs) near abandoned tailings on the Loess Plateau, comparing with the adjacent bare land.

Results

The results showed that phytomanagement significantly enhanced soil aggregate stability, as demonstrated by the rise in soil macro-aggregate proportion (> 0.25 mm, 10.5–22.4%) and geometric mean diameter (16.3–44.0%). Furthermore, the soil organic carbon content (SOC), glomalin-related soil protein (GRSP), aromatic-C, and alkene-C in macro-aggregates increased alongside aggregate stability enhancement. The increased stability of soil aggregates following phytomanagement could reduce the risk of heavy metal leaching, but the increased concentration of heavy metals in the aggregates. In addition, the management of artificial forests mixed with shrubs and herbs greatly reduced the ecological risk of heavy metal pollution compared to other phytomanagement. SOC and GRSP were identified as key factors influencing the risk of heavy metal contamination in soil aggregates following phytomanagement.

Conclusion

Our study highlights that revegetation of tailings improves soil quality and ecological security by increasing the stability of soil structure as well as SOC and GRSP within the aggregates. Artificial forests mixed with shrubs and herbs could be an optimal phytomanagement to achieve environmental sustainability in tailings areas.

Introduction

Mineral tailings are one of the most daunting environmental challenges faced by the global mining industry. Improper remediation of tailings can lead to various environmental issues, such as surface erosion, excessive concentrations of heavy metals, and nutrient deficiency (Cai et al. 2023; Kumar et al. 2018). Abandoned tailings are subject to strong wind and rain erosion, and their surface nutrients and heavy metals are transferred with runoff and sediments, destabilizing the surrounding ecosystem (Djukic et al. 2016; Karaca et al. 2018; Zhang et al. 2015). Additionally, the prolonged abandonment of tailings
can exacerbate the degradation of the surrounding pristine ecosystem and pose a threat to human health (Alvarez-Rogel et al. 2022; Gutierrez et al. 2016). It is imperative to take effective measures to minimize the environmental risks associated with heavy metal pollution and to improve soil quality in tailing areas.

Phytoremediation of tailings by establishing vegetation is widely considered to be a sustainable approach for remediating tailings, because it is more environmentally friendly and cost-effective compared to traditional techniques (Ju et al. 2019; Wang et al. 2017). Revegetation has been shown to enhance soil quality by improving soil structural stability and nutrient levels, and can serve as a phytomanagement strategy to modify the transfer and accumulation of heavy metals in tailings soil and reduce environmental risks associated with heavy metals (Alvarez-Rogel et al. 2022; Cai et al. 2023; Karaca et al. 2018). In addition, the fate of heavy metals in soil depends on the plant species and soil environment. Co-planting of multiple plant species has more plant diversity, biomass and plant cover than monoculture planting and is considered a promising phytomanagement for improving soil environmental quality (Alvarez-Rogel et al. 2021; Erktan et al. 2016; Zhao et al. 2022). For example, co-planting *Brassica napus* and *Salix nigra* had better results on soil structure and copper stabilization compared to planting alone (Massenet et al. 2021). Multiple tree-herb symbiosis treatments can effectively mitigate the toxic effects of metal ions on soil enzyme activity and microbial communities and improve the biological quality of heavy metal contaminated soils (Zeng et al. 2019). However, further clarification is needed on the effectiveness of diverse phytomanagement measures in improving soil quality and ecological security in tailings areas.

Soil aggregation and structure are fundamental properties in ecosystems, and can serve as physical characteristics of soil quality indicators (Bronick and Lal 2005; Rillig et al. 2002). The stability of soil structure depends on the presence of stable aggregates and is essential for maintaining soil productivity and reducing soil erosion and degradation (An et al. 2013; Menon et al. 2020; Six et al. 2000). Determination of soil aggregation may involve assessing the proportion of water-stable aggregates (WSA) and the geometric mean diameter (GMD). Furthermore, an increase in physical binder soil organic carbon (SOC) and biological binder glomalin-related soil protein (GRSP) within soil aggregates enhances soil aggregation (Bronick and Lal 2005; Holatko et al. 2021). The C-related functional components of soil aggregates determined from transform infrared spectroscopy can predict soil aggregate stability (Liu et al. 2021). GRSP is not only one of the main sources of organic carbon in soils, but also affects the transport of heavy metals as well as their bioavailability (Chen et al. 2022; Gujre et al. 2021). In addition, the particle size of soil aggregates is an important factor in determining the concentration, mobility and transformation of heavy metals (Deng et al. 2018; Huang et al. 2020; Neagoe et al. 2012). It has been suggested that heavy metals are more stable in soil macroaggregates than in microaggregates, thus increasing the proportion of macroaggregates can reduce the leaching of heavy metals from the soil (Chen et al. 2021). However, the degree of heavy metal contamination and ecological risk caused by the presence of multiple heavy metals in aggregates of different particle sizes has not yet been addressed. Therefore, understanding soil aggregate-associated structural stability and heavy metal pollution risks is essential for assessing the improvement of soil ecological quality through revegetation of tailings.
In China, more than 70% of coal mining activities occur in the semi-arid Loess Plateau, resulting in the destruction of its delicate ecosystem and exacerbating soil erosion and land degradation (Wang et al. 2021; Zhang et al. 2021). In the past 20 years, the Loess Plateau has implemented large-scale phytomanagement measures such as returning farmland to forests and grasses, artificial afforestation and natural restoration, which have greatly alleviated the problems of soil erosion and land degradation (An et al. 2013; Liu et al. 2020; Wang et al. 2021). However, past research has tended to focus solely on either the soil structure of the Loess Plateau or the level of heavy metal contamination from tailings (Duan et al. 2022; Yuan et al. 2022; Zhang et al. 2015; Zhang et al. 2021), with few studies considering both factors together. As a result, it is essential to comprehensively evaluate the effects of revegetation on soil quality and heavy metal pollution risks on the Loess Plateau.

The Baiyin is an industrial resource-based city located in the western part of China's Loess Plateau, rich in mineral resources and with a large number of smelters and chemical plants (He et al. 2019). The development of long-term and large-scale mining and metal smelting activities has seriously damaged the local ecological environment. In this study, we investigate the effects of revegetation conducted by multiple phytomanagement on soil structural stability and risk of heavy metal contamination near an abandoned tailings area in Baiyin. The main objectives of this research are (1) to investigate the stability of soil aggregates and the influencing factors under phytomanagement, and (2) to evaluate the degree and risk of heavy metal contamination in soil aggregates and to determine the optimal phytomanagement measures for the site. Here, we propose two hypotheses: (1) phytomanagement can improve environmental quality in soil aggregates, which is associated with the increase of nutrients and GRSP; (2) multiple combinations of plant species may be the optimal phytomanagement to improve the ecological quality of tailings on the Loess Plateau.

**Materials and methods**

Study sites and soil sampling design

The study site is located in southeast of Baiyin City, Gansu Province, China, near an abandoned mine tailing (Fig. 1). A Pb-Zn smelter is located in the northeastern part of the tailings. The area is located on the Loess Plateau and has an arid and semi-arid climate with an average annual precipitation of 300 mm. The prevailing wind direction in the area is north and the soil type is calcareous (He et al. 2019). The study area is located in the Loess Plateau, where the soil is extremely vulnerable to water and wind erosion. In addition, frequent mining activities have led to a high risk of soil erosion at the site (Tong and Shi 2020). The study sites had four different land-use types, including bare land without vegetation (BL) as a control and three phytomanagement treatments after revegetation (Table S1). The three phytomanagement treatments were divided into natural grassland with herbs and shrubs (NG), artificial forest with poplar trees (AF), and artificial forest mixed with shrubs and herbs (FSH). These three phytomanagement measures have been in place for approximately five years. The dominant herbs and shrubs in the GL treatment were *Setaria viridis* (L.) Beauv., *Taraxacum mongolicum* Hand.-Mazz., *Bothriochloa ischaemum* (L.) Keng, and *Artemisia argyi* Levl. et Van. The poplar trees grown in the AF
treatment were *Populus* L.. The poplar trees in the FSH treatment were *Populus* L. and the shrubs and herbs were mainly *Setaria viridis* (L.) Beauv., *Bothriochloa ischaemum* (L.) Keng, *Artemisia gmelinii* Weber ex Stechm, and *Artemisia argyi* Levl. et Van.. The different land use types at the four study sites had similar elevations, slopes, and aspects, without interference from watering, fertilizing, tillage, and other measures.

Soil samples from the four sampling sites were collected in September 2018. Five sampling points were selected in each sampling site through an “S”-shaped random-sampling method. Each sampling point was separated by approximately 10 m. The litter horizons were removed before soil samples were collected at all points. Soil cores were collected from the top 10 cm of the soil profile at each point using aluminum containers (10 cm high and 10 cm diameter) to reduce sampling disturbance to the soil structure. Soil samples were then transferred to the laboratory and dried naturally. The air-dried soil samples from each sampling site were mixed through an 8-mm sieve into a composite sample before the determination of soil aggregates.

### Determination of soil aggregate size distribution and stability

Four aggregate size classes were isolated using the classical wet sieving method (Elliott 1986) as follows: large macroaggregates (> 2 mm, LMA), macroaggregates (2–0.25 mm, MA), microaggregates (0.25–0.053 mm, MI), and silt + clay (< 0.053 mm, SC). Briefly, approximately 100 g air-dried and undisturbed soil samples were moistened in distilled water for 30 min and subsequently transferred to three different size sieves (2, 0.25 and 0.053 mm). The sieves were then moved vertically in the water (5 cm amplitude at a frequency of 40 times min\(^{-1}\) over 2 min). After oven drying at 60°C, the four classes of aggregates were stored at room temperature for future analysis of the physicochemical characteristics. The mass percentage (%) of each aggregate fraction was calculated by weighing. In addition, the macro-aggregates mentioned below represent aggregates > 0.25 mm and micro-aggregates represent aggregates < 0.25 mm.

The water-stable aggregates (WSA, %) and geometric mean diameter (GMD, mm) indices were used to assess the water stability of soil aggregates. The value of WSA was calculated by the proportion of soil particles > 0.25 mm. The value of GMD is calculated using the following equation:

\[
GMD = \exp\left(\frac{\sum_{i=1}^{n} w_i \ln x_i}{\sum_{i=1}^{n} w_i}\right)
\]

where \(n\) is the number of aggregate size fractions, \(w_i\) is the mass proportion of the aggregate fraction \(i\), and \(x_i\) is the mean diameter of the aggregate fraction \(i\).

The soil structural stability index (SSI, %) was calculated using the following equation proposed by Pieri (1992) and Reynolds et al. (2009):
where \( SOM(\%) \) is the content of soil organic matter that is calculated from the SOC content (\%) and the conversion coefficient of 1.724; \( SC(\%) \) is the mass proportion of the aggregate fraction silt + clay. An SSI > 9% indicates stable soil structure, 5% < SSI ≤ 9% indicates soil structure at risk of degradation, and SSI ≤ 5% indicates structurally degraded soil. Note that since SSI is based on SOC and texture, it does not relate directly to the porosity aspects of soil structure, but rather to the “resilience” of the structure (Reynolds et al. 2009).

**Determination of soil nutrients, metals and GRSP content and FTIR analysis**

The dried soil aggregate samples underwent a pre-treatment procedure that involved grinding and acidification. Subsequently, the soil organic carbon (SOC) and total nitrogen (TN) contents were quantified using a FlashSmart Elemental Analyzer (Thermo Fisher Scientific, USA). The total phosphorus (TP) content was measured utilizing the molybdenum blue method with the aid of an ultraviolet spectrophotometer (UV3200, Shimadzu Corporation, Japan). Approximately 0.200 g of soil was digested in an \( \text{HNO}_3 \) and \( \text{HClO}_4 \) and \( \text{HCl} \) \( (3:1:1, \text{v/v/v}) \) solution (Ju et al. 2019) and then tested for total heavy metal concentrations using a flame atomic absorption spectrophotometry (PinAAcle 900F, PerkinElmer, Germany). The tested heavy metals included cadmium (Cd), copper (Cu), lead (Pb), and zinc (Zn).

The content of glomalin-related soil protein (GRSP) in soil aggregates was determined after extraction according to the procedures described by Wright and Upadhyaya (1998). Easily extractable GRSP \( (E_{\text{GRSP}}) \) and total extractable GRSP \( (T_{\text{GRSP}}) \) were extracted from air-dried soils using an autoclave method. This method obtains \( E_{\text{GRSP}} \) by autoclaving 1 g of soil samples in sodium citrate buffer \( (8 \text{ mL}, \text{pH} = 7.0) \) at 121 °C for 60 min and then centrifuging the solution at 10,000 \( \times \text{g} \) for 5 min to obtain the supernatant. \( T_{\text{GRSP}} \) was obtained by repeatedly autoclaving 1 g of soil sample in sodium citrate buffer \( (8 \text{ mL}, 50 \text{ mM}, \text{pH} = 8.0) \) at 121°C for 5 times, each time lasting 60 min, followed by centrifugation at 10,000 \( \times \text{g} \) for 5 min to obtain pooled supernatants. Finally, the extracts were added to Coomassie Brilliant Blue and colorimetric at 595 nm using a spectrophotometer (UV3200, Shimadzu Corporation, Japan) with bovine serum albumin (BSA) as a standard.

Soil components were investigated by using attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR) (Dhillon et al. 2017). A Bruker Nicolet iS10 FTIR spectrometer equipped with a deuterated triglycine sulphate (DTGS) detector was used for ATR-FTIR spectra acquisition of soil samples. After cleaning the ATR crystal with absolute ethanol, the soil sample was pressed onto the ATR crystal for spectral scanning. The spectra were collected by scanning over a spectral range of 4000 – 400 \text{ cm}^{-1} \) with a resolution of 4 \text{ cm}^{-1} and calibrated against the ambient air background. In addition, we conducted semi-quantitative analysis of organic C composition (Liu et al. 2021; Solomon et al. 2007). Each peak in the spectrogram was identified and classified into the corresponding C composition, and the
relative peak areas were calculated by performing a multi-peak fit analysis using OriginPro 2022 (OriginLab Corporation, Northampton, MA, USA) to estimate the relative proportions of C composition.

Assessment of heavy metal pollution in soil aggregates

Grain size fraction metals loading (GSF) refers to the heavy metal loading in each particle size of soil aggregates, which was used to evaluate the contribution of heavy metals in different particle sizes (Sutherland 2003). The value of GSF is calculated using the following equation:

\[
\text{GSF} = \frac{\sum_{i=1}^{n} (\text{metal}_i \times w_i)}{\sum_{i=1}^{n} (\text{metal}_i \times w_i)} \times 100
\]

where \( n \) is the number of aggregate size fractions, \( w_i \) is the mass proportion of the aggregate fraction \( i \), and \( \text{metal}_i \) is the concentration of heavy metals in the corresponding aggregate fraction \( i \).

The Pollution Load Index (\( PLI \)) was used to assess the level of heavy metal contamination in soils as a way to demonstrate the deterioration of soil conditions due to the accumulation of heavy metals (Kowalska et al. 2018; Rinklebe et al. 2019). \( PLI \) is calculated as a geometric average of Single Pollution Index (\( PI \)) based on the following equation:

\[
\text{PLI} = (\text{PI}_1 \times \text{PI}_2 \times \text{PI}_3 \ldots \times \text{PI}_n)^{1/n}
\]

\[
\text{PI} = \frac{C_S}{C_{\text{RefS}}}
\]

where \( C_S \) is the content of heavy metal in soil, \( C_{\text{RefS}} \) is reference content in pristine soils, \( PI \) is the contamination factor of each metal, and \( n \) is the number of analyzed heavy metals. The reference concentrations used in this study were according to the local soil elemental geochemical background values (\( C_{\text{RefS\_GB}}, PL_{\text{GB}}, PLL_{\text{GB}} \)) and the contamination thresholds in national soil environmental quality standards (\( C_{\text{RefS\_NS}}, PL_{\text{NS}}, PLL_{\text{NS}} \)), respectively. The geochemical background content of Cd, Cu, Pb, and Zn in the soil in Gansu was 0.146, 21.7, 24.5, and 71.8 mg kg\(^{-1}\) (Chen et al., 2015). The contamination threshold of Cd, Cu, Pb, and Zn in the soil was 0.60, 100, 170, and 300 mg kg\(^{-1}\), obtained from Soil Environmental Quality Risk Control Standard for Soil Contamination of Agricultural Land of China (GB 15618 – 2018). Single metal pollution classification assessed by \( PI \): no pollution (\( PI < 1 \)), low pollution (\( 1 < PI < 2 \)), moderate pollution (\( 2 < PI < 5 \)), and severe pollution (\( PI > 5 \)). \( PLI < 1 \) represents that the level of heavy metal pollution in soil is not significant; \( PLI > 1 \) represents significant pollution.

Potential ecological risk (\( RI \)) is an index applicable for the assessment of the degree of ecological risk caused by heavy metal concentrations in the soil, and determines the sensitivity of biological
communities in large contaminated areas (Kowalska et al., 2018). It takes into account the $PI$ of metals, their potential ecological risk factor ($E_r$) and the toxicological response factor ($T_r$) (Hakanson, 1980). It is calculated by the following equation:

$$RI = \sum_{i=1}^{n} E_r^i = \sum_{i=1}^{n} T_r^i \times PI^i$$

where $E_r^i$ is the potential ecological risk factor of a single metal, $T_r^i$ is the toxicity response coefficient of a single metal, $PI^i$ is calculated values for the Single Pollution Index, and $n$ is the number of analyzed heavy metals. Soil ecological risk classification assessed by $RI$: low risk ($RI < 90$), moderate risk ($90 < RI < 180$), strong risk ($180 < RI < 720$), and severe risk ($RI > 720$).

**Statistical analysis**

Prior to statistical analysis, normality and homogeneity of variance were assessed for all data using the "car" package in the R environment. One-way analysis of variance (ANOVA) with Tukey's HSD test ($P < 0.05$) was employed to determine the impact of phytomanagement on various variables, including soil aggregate particle size, WSA, GMD, SSI, nutrients, GRSP, heavy metals, $PLI$, and $RI$. The values reported were the means ($n = 3$) ± the standard deviation ($sd$). Two-way ANOVA was performed to analyze the effects of phytomanagement, aggregate size, and their interactions on various variables. Line and box plots were created using the "ggplot2" package to visualize nutrient, GRSP, and heavy metal contents in soil aggregates. Circular bar plots were constructed using the "ggplot2" and "tidyverse" packages to show the GSF, $PLI$, and $RI$ of heavy metals in soil aggregates.

A principal component analysis (PCA) was conducted using the "FactoMineR" package to explore the correlation of various variables in soil aggregates. The linear regression relationship of $PLI$ and $RI$ of heavy metals with nutrients and GRSP was analyzed using the "ggpmisc" package. Variation partitioning analysis (VPA) in the "vegan" package was utilized to identify the contribution of interferences (phytomanagement practices and aggregate particle size), nutrients, and GRSP and their interactions to the variation of the $PLI$ and $RI$ of heavy metals in soil aggregates. We utilized a random forest model (RFM) implemented using the "randomForest" package to identify key predictors affecting the $PLI$ and $RI$ of heavy metals. The importance of these predictors was estimated using the percentage increase in MSE (Mean Squared Error). The significance of RFM and predictors were analyzed using the "rfPermute" and "rfUtilities" packages, respectively. The key predictors identified by the RMF were further incorporated into a partial least squares path modeling (PLS-PM) to determine the direct and indirect paths by which the predictors affected $PLI$ and $RI$. PLS-PM was implemented by using the "plspm" package. All statistical analyses were performed in the R environment (v.4.1.3, https://www.r-project.org/).

**Results**
Soil aggregate size distribution and stability

The size distribution and stability of soil aggregates varied significantly between land use types (Fig. 2). LMA was the dominant soil aggregate in bare land, accounting for 49.1% of the total, followed by SC at 31.9% (Fig. 2a). Phytomanagement decreased the proportion of LMA (25.9–57.2%) and SC (31.3–53.0%), but increased the proportion of MA (181–324%). In comparison to the BL treatment, soil WSA (> 0.25 mm) was significantly increased by 19.3% and 22.4% in the AF and FSH treatments, respectively (P < 0.05, Fig. 2b). The GMD of soil aggregates increased by 16.3–44.0% after phytomanagement, with the highest value observed in the AF treatment (Fig. 2c). Similarly, phytomanagement significantly increased the soil SSI, with the highest SSI was found in the NG treatment (25.6), followed by the AF (20.0) and FSH (12.8) treatments (Fig. 2d).

Aggregate-associated nutrients and heavy metals

The implementation of multiple phytomanagement practices led to an increase in the SOC content and C:N:P stoichiometry of the bulk soil, while decreasing the TP content (Table S2). The heavy metal (Cd, Cu, Pb, Zn) content of the bulk soil was generally higher in the NG treatment as compared to the BL treatment. Phytomanagement and aggregate size and their interactions significantly impacted the distribution of nutrients and heavy metals in soil aggregates (Fig. 3 and Table S3, P < 0.05). In general, phytomanagement increased SOC content and decreased TP content in soil aggregates, as compared to bare land (Fig. 3a-c). The highest SOC content in soil aggregates was found in the NG treatment, which was 1.8–4.4 times higher than that in the BL treatment. Relatively low levels of TN and TP in soil aggregates were found in the AF treatment. Additionally, the nutrients were found to primarily accumulate in aggregates greater than 0.25 mm in the NG and FSH treatments and in aggregates less than 0.053 mm in the AF treatment. The average values of C:N, C:P and N:P in soil aggregates in all treatments were found to range from roughly 10–30, 10–75 and 1–3, respectively (Fig. 3d-f). The C:N and C:P values in soil aggregates increased after plant management, with the highest values observed in the NG treatment. The levels of N:P in < 2 mm aggregates increased after plant management, while decreasing slightly in LMA. Furthermore, we found that the heavy metal content in LMA generally decreased after plant management, whereas the heavy metal content in aggregates of other sizes increased (Fig. 3g-j). The Cd, Cu, Pb, and Zn contents in LMA were the lowest in the AF treatment, which were 57.7%, 38.1%, 45.9%, and 49.8% lower than that in the BL treatment, respectively. With regards to phytomanagement treatments, the order of heavy metal content in soil aggregates was NG > AF > FSH, with the heavy metal content being higher in MA and lower in MI.

Soil components and GRSP content

In general, the absorbance of the soil ATR-FTIR spectra was comparable among different treatments as well as different aggregate sizes, as shown in Fig. 4a. We summarized the three main absorption peak characteristics corresponding to functional groups or soil components. The intense vibrational bands observed at 1680 cm⁻¹ and 1520 cm⁻¹ were likely attributed to C = C vibration of alkenes and aromatic
compounds, respectively. The band observed at 1005 cm$^{-1}$ was assigned to the Si–O stretch from both quartz and clay minerals. The relative proportions of alkenes and aromatics in soil aggregates were higher in the phytomanagement treatments than in the BL treatment, while the proportions of quartz and clay minerals were lower (Table S4). The dominant components of soil aggregates were aromatics in the AF and FSH treatments and minerals in the BL treatment. Quartz and clay minerals were the dominant components of aggregates > 0.25 mm in the NG treatment, whereas aromatics were dominant in aggregates < 0.25 mm.

The content of GRSP (i.e. T_GRSP and E_GRSP) in soil aggregates showed a significant increase after plant management (Fig. 4b-d). The highest T_GRSP and E_GRSP contents were observed in the NG treatment, being 56–156% and 209–519% higher, respectively, than those in the BL treatment. The lowest GRSP content was observed in MI across all treatments. The E_GRSP/T_GRSP of soil aggregates in the phytomanagement treatments was found to be higher compared to the BL treatment. The value of E_GRSP/T_GRSP was higher in SC than that in other aggregate sizes.

Risk of heavy metal pollution in soil aggregates and its influencing factors

We observed that the heavy metal pollution load index and ecological risk calculated using the national soil pollution standards ($PLI_{NS}$ and $RI_{NS}$) were lower than those calculated using the elemental geochemical background content of the area ($PLI_{GB}$ and $RI_{GB}$) (Fig. 5). Based on the estimated bulk soil $PLI$ and $RI$, the FSH treatment showed the lowest level of heavy metal pollution and ecological risk compared to the other treatments (Fig. 5a). Phytomanagement had a significant impact on the heavy metal loading in each particle size of soil aggregates (Fig. 5b), with an increasing contribution of heavy metals in MA and a decreasing contribution in LMA for all phytomanagement treatments compared to the BL treatment. The level of heavy metal pollution, as indicated by the $PLI$, exhibited significant variation among the various soil aggregate sizes in the BL, NG, and AF treatments (Fig. 5c). Specifically, the highest $PLI$ values were observed in the LMA for the BL treatment, and in the MA for the NG and AF treatments. Notably, the discrepancy in $PLI$ values among the different aggregate sizes was mitigated in the FSH treatment. Moreover, the $RI$ results for heavy metals in soil aggregates were congruent with those of the $PLI$ (Fig. 5d).

The results of the linear regression analysis showed significant and positive correlations between the $PLI$ and $RI$ of heavy metals in soil aggregates with the contents of SOC, TN, and GRSP, as well as nutrient stoichiometry (Fig. S1). Additionally, the PCA analysis revealed positive correlations between $PLI$ and $RI$ with nutrients, GRSP, and their stoichiometry (Fig. S2). The RFM and VPA were employed to identify the key factors influencing heavy metal pollution load and ecological risk in soil aggregates (Fig. 6). Our findings showed that soil GRSP, C, and N contents were key factors that predicted changes in $PLI$ and $RI$ (Fig. 6a and b). Interferences including phytomanagement practices and aggregate particle size were also non-negligible factors affecting the $PLI$ and $RI$. Results from the VPA showed that interferences, soil nutrients, and GRSP, and their interactions explained 67.6% and 61.6% of the variation in $PLI$ and $RI$, respectively (Fig. 6c and d). Soil nutrients alone explained the greatest variation in $PLI$ and $RI$, followed by
interferences and soil GRSP. The interaction between GRSP and nutrients was also the key player in the changes in \( PLI \) and \( RI \). We used PLS-PM analysis to further decipher the processes by main factors influence the heavy metal pollution load and ecological risk in soil aggregates (Fig. 7).

Phytomanagement practices and aggregate particle size had negative direct effects on \( PLI \) and \( RI \), while soil nutrients (SOC and TN), TN:TP, and GRSP had positive direct effects (Fig. 7a and c). Soil TN:TP had the greatest direct impact on \( PLI \) (0.41) and \( RI \) (0.50). Overall, soil nutrients had the strongest and positive total effects on \( PLI \) and \( RI \), followed by TN:TP and GRSP, while aggregate particle size had a negative total effect on \( PLI \) and \( RI \) (Fig. 7b and d).

**Discussion**

Effects of phytomanagement on soil aggregation

We found that the WSA and GMD indices were higher in revegetation-managed land than in bare land, suggesting that phytomanagement improved the stability of soil aggregates. The results of SSI also indicated that the implementation of phytomanagement near tailings with severe soil erosion can stop soil degradation and improve soil structural stability. Revegetation as an effective phytomanagement measure has been shown to improve soil physical properties and anti-erodibility in vulnerable ecological mining regions (Alvarez-Rogel et al. 2021; Yuan et al. 2022; Zhang et al. 2015). We also found that forests were more effective than natural grasslands in improving the water stability of soil aggregates, possibly due to the fact that tree plantings produce more WSA than herbs and shrubs. The stability of soil aggregates can increase along the successional gradient of severely eroded ecosystems, being thrice higher in tree-dominated communities as compared to grass-dominated communities, which is related to soil characteristics, plant cover and root traits (Erktan et al. 2016).

The distribution of aggregate particle size is a crucial factor in determining soil aggregate stability, with > 0.25 mm macro-aggregates (WSA) being a more sensitive indicator of differences in soil structural stability among various land use types (Menon et al. 2020; Six et al. 2000). We used RFM to reveal the main factors influencing the proportion change of WSA under phytomanagement and found that soil TN, GRSP, and SOC contents were the primary contributors to the stability of soil aggregates (Fig. S3). The influence of N content on the proportion of WSA in the soil is likely because N is more readily distributed in macro-aggregates (Ju et al. 2023; Lu et al. 2021)d accumulation increases soil macro-aggregates (Simansky et al. 2019). Furthermore, N can influence the binder SOC and GRSP (Rillig et al. 2002; Zhang et al. 2023). In addition, heavy metals in soil affected WSA, because the particle size of aggregates under land use can affect the distribution of metal ions (Chen et al. 2022; Huang et al. 2020). Metal ions can also affect the chelation with organic matter and GRSP, the production of GRSP by AMF, and the turnover of nutrients by microorganisms (Gujre et al. 2021; Ju et al. 2019; Riaz et al. 2021).

The GRSP and SOC were found to be key factors indicating the stability of heavy metal-contaminated soil aggregates under multiple phytomanagement, which is consistent with the mechanism of soil aggregate stabilization (Holatko et al. 2021; Lehmann et al. 2007; Rillig et al. 2002; Six et al. 2000). The GRSP
content in soil aggregates increased after the implementation of plant management, indicating that phytomanagement can stabilize soil aggregates by exerting the binding effect of GRSP. The contents of T_GRSP and E_GRSP in soil aggregates were higher in the NG treatment than in the AF and FSH treatments, which may be due to the higher plant diversity, biomass and shallow fine roots in the grassland that promote the turnover of AM hyphae in the soil surface and produce more GRSP (Holatko et al. 2021; Rillig et al. 2002). E_GRSP is considered as a more reactive and unstable C, and its difference with T_GRSP (E_GRSP/T_GRSP) is used to assess the GRSP accumulation potential (Liu et al. 2020). The higher E_GRSP and GRSP accumulation potential in SC in plant management, especially in the AF and FSH treatments, coupled with a relatively high proportion of WSA, we speculate that soil aggregate formation in forests may be a bottom-up model in which micro-aggregates further form macro-aggregates by increasing C and GRSP (Haddix et al. 2016; Lehmann et al. 2007; Wilpiszeski et al. 2019). In addition, the change pattern of GRSP in soil aggregates was similar to that of SOC, which may imply that phytomanagement can achieve C accumulation in heavy metal-contaminated soils by altering the conversion of GRSP (Gujre et al. 2021).

All three phytomanagement measures in heavy metal-contaminated areas increased the SOC content of soil aggregates, and the SOC content was generally higher in macro-aggregates than in micro-aggregates. Furthermore, we found that phytomanagement increased the contents of alkene-C and aromatic-C in soil aggregates and decreased the contents of quartz and clay minerals (Table S4). Aromatic-C is produced by the oxidation of lignin and remains relatively stable in the soil (Verchot et al. 2011). Aromatic-C and alkene-C are mainly distributed in macro-aggregates and constitute most of C, playing a key role in the formation of macro-aggregates (Liu et al. 2021; Xue et al. 2019). Aromatic-C and alkene-C in macroaggregates were higher in AF and FSH treatments than in NG treatments, thus explaining the higher water-stability of soil aggregates in artificial forest than in naturally restoration grassland. In addition, the higher SOC, aromatic-C, and alkene-C were found in SC in the AF treatment, indicating that artificial forests can store OC persistently in the soil relative to other plant management. This is because C in < 0.053 mm aggregates persist longer in the soil as mineral-associated organic carbon (MAOC), which is protected from decomposition by binding to minerals (Cotrufo et al. 2019; Lavallee et al. 2020; Lehmann et al. 2007). Therefore, SOC components are also key factors indicating the stability of soil aggregates under plant management in heavy metal-contaminated areas, in addition to SOC associated with macro-aggregates.

Environmental risks of heavy metal contamination in soil aggregates

The mean values of Cd, Cu, Pb, and Zn in soil aggregates at the studied site were 1.42, 33.8, 46.6 and 86.5 mg·kg$^{-1}$, respectively (Fig. S4). These values were 9.7, 1.6, 1.9, and 1.2 times of their background values (Chen et al. 2015). Based on the risk control standards for agricultural soil contamination in China, only Cd exceeded the standard threshold (0.60 mg·kg$^{-1}$) in this area. Generally, phytomanagement increased heavy metal content in surface soil aggregates near the tailings, when compared to bare land. This phenomenon could be attributed to the vulnerability of bare land to water and wind erosion, leading to the easy transfer of metals from the surface of bare land through wind and water forces (Djukic et al.
Although establishing vegetation on mine tailings could decrease wind and surface erosion, it may also reduce the leaching and movement of heavy metals from the soil surface and increase the accumulation of heavy metals. Our results support the notion that the implementation of revegetation near tailings could reduce the ecological risk of heavy metal leaching by immobilizing and stabilizing soil heavy metals through plant roots and exudates as well as aggregate characteristics and stability (Chen et al. 2021; Zhao et al. 2022). Moreover, natural grasslands allowed more heavy metals to accumulate on the soil surface than artificial forests.

Previous research has indicated that smaller soil aggregates can accumulate more heavy metals than larger ones due to their greater surface area, presence of organic matter, and iron-manganese oxides (Deng et al. 2018; Quenea et al. 2009). However, in this investigation, heavy metals were mainly distributed in larger soil aggregates. Heavy metals were primarily distributed in 2–0.25 mm aggregates under phytomanagement, while the distribution in 0.25–0.053 mm aggregates was negligible. The GSF of heavy metals in 2–0.25 mm aggregates did not differ significantly between grasslands and artificial forests. Additionally, the mass loadings of heavy metals were higher in macro-aggregates as compared to micro-aggregates. This is because > 0.25 mm macro-aggregates account for a large proportion in the whole bulk soil, and more binding substances in macro-aggregates, such as SOC and GRSP, can combine with metal ions to form stable organic-metal complexes (Chen et al. 2021; Huang et al. 2020; Neagoe et al. 2012). One of our previous studies also found that metal loading in macro-aggregates was also higher than in micro-aggregates during in situ phytoremediation at a mining site (Chen et al. 2022). While phytomanagement resulted in increased levels of SC-associated heavy metals, the proportion of SC was relatively small, and its contribution to the overall heavy metal mass load remained limited. Furthermore, the distribution of heavy metals among different soil aggregate particle sizes is contingent upon various factors such as metal species, soil type, land use, and human activities.

Phytomanagement has been shown to increase heavy metal pollution levels (PLI) and ecological risks (RI) in soil aggregates, particularly in the 2–0.25 mm aggregates, due to increased heavy metal content. The pollution levels and ecological risks of heavy metals in the AF and FSH treatments were found to be lower than in the NG treatment, suggesting that tree planting may effectively reduce the risk of heavy metal contamination in the soil at this site (Alvarez-Rogel et al. 2022; Ettler et al. 2014; Fang et al. 2017). Moreover, the pollution levels and ecological risks of heavy metals in the artificial forest mixed with shrubs and herbs were the lowest among the four land use types. This is consistent with our second hypothesis that multi-plant species such as woody-herb combinations are more likely to improve soil ecological security near tailings. The RFM, VPA, and SEM analyses elucidated the soil properties affecting PLI and RI in soil aggregates, indicating that soil nutrients and GRSP strongly influenced heavy metal pollution levels and ecological risks. Among these, SOC and E_GRSP had strong positive effects on heavy metal pollution levels and ecological risks in soil aggregates. Previous studies have demonstrated that higher GRSP and SOC levels in soils can increase the stability of heavy metals and reduce their leaching risk by adsorbing metal ions (Gujre et al. 2021; Holatko et al. 2021; Neagoe et al. 2012; Riaz et al. 2021). Because of the low levels of SOC and GRSP, the ecological risks of artificial forest mixed with shrubs and
herbs was relatively lower than that of natural grassland and artificial forest without grasses. Therefore, the levels of SOC and GRSP can also indicate the pollution level and ecological security of tailings soils.

**Conclusion**

Revegetation-based phytomanagement has been found to improve the structural stability of tailings soils on the Loess Plateau by increasing the nutrients, GRSP, WSA, and GMD of soil aggregates. The content of aromatic-C and alkene-C in macro-aggregates also increased with the improvement in aggregate stability. Furthermore, the improved stability of soil aggregates reduced the risk of heavy metal leaching within the aggregates. The mass loadings, pollution loadings, and ecological risks of heavy metals were higher in macro-aggregates than in micro-aggregates. Phytomanagement could improve soil quality and enhance ecological security by increasing soil structural stability, SOC and GRSP. In addition, the study found that an artificial forest mixed with shrubs and herbs exhibited higher soil aggregation stability, nutrients and ecological security, compared to natural restoration grassland and artificial forest. Therefore, a combination of woody and herbaceous plants could be the optimal land use strategy to mitigate ecological risks caused by heavy metal contamination in soils near mining sites.

**Declarations**

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**Declarations**

**Competing Interests** The authors have no relevant financial or non-financial interests to disclose.

**Data availability**

Data will be made available on request.

**References**


Figures

Figure 1

Map of study sites and sampling locations. BL, bare land without vegetation, as a control; NG, natural grassland with herbs and shrubs; AF, artificial forest with poplar trees; and FSH, artificial forest mixed with shrubs and herbs.
Figure 2

Soil aggregate size distribution and stability under multiple phytomanagement. WSA, water-stable aggregates; GMD, geometric mean diameter; SSI, soil structural stability index. BL, bare land; NG, natural grassland; AF, artificial forest; and FSH, artificial forest mixed with shrubs and herbs. LMA, large macroaggregates, >2 mm; MA, macroaggregates, 2–0.25 mm; MI, microaggregates, 0.25–0.053 mm; and
SC, silt+clay, <0.053 mm. Different lowercase letters indicate statistical differences (Tukey's HSD test, P<0.05) between different sites.

Figure 3

The distribution of nutrients and heavy metals in soil aggregates. SOC, soil organic carbon; TN, total nitrogen; TP, total phosphorus; Cd, cadmium; Cu, copper; Pb, lead; and Zn, zinc. BL, bare land; NG, natural grassland; AF, artificial forest; and FSH, artificial forest mixed with shrubs and herbs. LMA, large
macroaggregates, >2 mm; MA, macroaggregates, 2–0.25 mm; MI, microaggregates, 0.25–0.053 mm; and SC, silt+clay, <0.053 mm. Different lowercase letters indicate statistical differences (Tukey’s HSD test, $P < 0.05$) between different sites for the same sized aggregates.

**Figure 4**

The carbon components and glomalin-related soil protein content in soil aggregates. (a), soil carbon components were investigated by using attenuated total reflectance Fourier transform infrared spectroscopy (ATR-FTIR); (b), $T_{GRSP}$, total extractable glomalin-related soil protein; (c), $E_{GRSP}$, easily extractable glomalin-related soil protein; (d), $E_{GRSP}/T_{GRSP}$, ratio of easily extractable GRSP to total
extractable GRSP. BL, bare land; NG, natural grassland; AF, artificial forest; and FSH, artificial forest mixed with shrubs and herbs. LMA, large macroaggregates, >2 mm; MA, macroaggregates, 2–0.25 mm; MI, microaggregates, 0.25–0.053 mm; and SC, silt+clay, <0.053 mm. Different lowercase letters indicate statistical differences (Tukey’s HSD test, \( P < 0.05 \)) between different sites for the same sized aggregates.

Figure 5

Heavy metal pollution loads and ecological risks in soils under multiple phytomanagement. (a), \( PLI \) and \( RI \) of heavy metals in the estimated bulk soil; (b), GSF of Cd, Cu, Pb, and Zn in soil aggregates; (c), \( PLI \) of heavy metals in soil aggregates; (d), \( RI \) of heavy metals in soil aggregates. The estimated bulk soil \( PLI \)
and \(R_I\) are the values calculated from the percentage of aggregates and their \(PL_I\) and \(R_I\). GSF, grain size fraction metals loading; \(PL_I\), Pollution Load Index; \(R_I\), potential ecological risk; GB, the local soil elemental geochemical background values; and NS, the contamination thresholds in national soil environmental quality standards. BL, bare land; NG, natural grassland; AF, artificial forest; and FSH, artificial forest mixed with shrubs and herbs. LMA, large macroaggregates, >2 mm; MA, macroaggregates, 2–0.25 mm; MI, microaggregates, 0.25–0.053 mm; and SC, silt+clay, <0.053 mm.

Figure 6

Potential drivers of variation in the level and ecological risk of heavy metal contamination in soil aggregates under multiple phytomanagement. Random forest model (RFM) predicts the importance (percentage of increase in MSE) of variables as drivers for \(PL_I\) (a) and \(R_I\) (b) of heavy metals in soil aggregates. Variation partitioning analysis (VPA) showing the contribution of interference and soil nutrients and GRSP to variation in \(PL_I\) (c) and \(R_I\) (d). Pollution Load Index (\(PL_I\)) and ecological risk (\(R_I\)) are used to assess the level and ecological risk of heavy metal contamination, respectively. The \(PL_I\) and \(R_I\) here are calculated based on the contamination thresholds in national soil environmental quality standards. Phytom., phytomanagement measures; aggre., aggregate particle size; SOC, soil organic carbon; TN, total nitrogen; TP, total phosphorus; T_GRSP, total extractable glomalin-related soil protein;
and E_GRSP, easily extractable GRSP. * denotes significance at $P < 0.05$; ** denotes significance at $P < 0.01$.

**Figure 7**

The major pathways by which key factors influence the level and ecological risk of heavy metal contamination. Partial least squares path modeling (PLS-PM) shows the direct and indirect effects of phytomanagement measures, aggregate particle size, soil nutrients (SOC and TN), TN:TP, and GRSP (T_GRSP and E_GRSP) on PLI (a and b) and RI (c and d). The key factors are determined by the random forest model. Pollution Load Index (PLI) and ecological risk (RI) are used to assess the level and ecological risk of heavy metal contamination, respectively. GOF denotes the goodness-of-fit index of PLS-PM. Solid and dashed lines denote positive and negative effects of causality, respectively. Numbers on arrow lines denote standardized path coefficients; * denotes significance at $P < 0.05$; ** denotes significance at $P < 0.01$; *** denotes significance at $P < 0.001$. Standardized total effects are direct plus indirect effects.

**Supplementary Files**
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