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1 Plant Litter as a Heavy Metal Migration Strategy Following Application of Sewage Sludge to
2 Subtropical Forest Soils

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Abstract

The environmental risks of migration of heavy metals (HMs) following applications of sewage sludge (SS) to forest soils are garnering increased attention. Plant litter at the forest floor may modify HM migration pathways through impacts on soil aggregates and water/soil erosion; however, HM migration responses to plant litter are poorly understood. The aim of this study was to determine the effects of plant litter cover on HMs migration, and water and soil erosion following the application of SS to subtropical forest soils. Effects of addition of SS along and SS plus plant litter at 0.75 or 1.5 kg m⁻² on the migration of cadmium, chromium, copper, nickel, lead, and zinc in surface runoff, soil interflow, and sediments were quantified across nine simulated rainfall events in a laboratory experiment and following natural intense rain events in a field experiment. Addition of SS elevated HM concentrations in surface runoff by 38.7 to 98.5%, in soil interflow by 48.3 to 312.5%, and in sediment by 28.5 to 149.4%, and increased the production of sediment aggregates <0.05 mm that led to greater cumulative migrations of HMs in surface runoff and sediment; sediment accounted for 89.5% of HM migrations. Addition of plant litter reduced cumulative migration of HMs by 87.1–97.27%; however, the higher rate of plant litter led to a decrease in surface runoff and sediment yield, and an increase in soil interflow. Addition of plant litter shifted the main pathway of HM migration from sediment to surface runoff and soil interflow. The potential ecological HM risk index was “low” for each treatment. We found consistency in HM concentrations and migrations via surface runoff between the field and laboratory experiments. Overall, the addition of plant litter with SS mitigated soil erosion and reduced total migration of HMs, resulting in a 88.7–97.3% decrease in the ecological risk index of the six HMs. We conclude that the addition of plant litter may provide a management strategy for the mitigation of HM risks to environmental safety for the disposal of SS in subtropical forest systems.

Keywords: Sewage sludge; Plant litter; Forest soil; Heavy metal; Migration pathway; Ecological

risk

1. Introduction

Sewage sludge (SS) is widely used as a soil amendment that reduces soil bulk density and shear strength of the surface layer, while increasing soil porosity, structural coefficient, permeability, and water-holding capacity (Zoghلامي et al., 2020), and acts as an exogenous source of nitrogen (N) and phosphorus (P) (Leila et al., 2017; Hoang et al., 2022), enhancing soil fertility and accumulation of plant biomass (Chu et al., 2017; Chu et al., 2023). Forest land is increasingly utilized for the disposal of SS (Abreu-Junior et al., 2017; Badzmierowski et al., 2021; Li et al., 2024), resulting in greater tree height, diameter, and biomass, including timber yield (Chu et al., 2017), due to greater soil content of N, P, potassium, and organic matter (Chow and Pan, 2020). However, forest inputs of SS are associated with greater content of heavy metals (HMs), such as copper (Cu), and zinc (Zn) (Chow and Pan, 2020), within the soil profile (Marguí et al., 2016; Chu and He, 2021; Li et al., 2024). The migration of HMs through soils mediated by surface water runoff and soil interflow pathways poses a potential threat to the environmental safety of rivers and groundwater (Koyuncu, 2022).

While soil adsorption and surface runoff migration of soluble or exchangeable state HMs tend to be negatively associated with soil particle size (Deng et al., 2018; Huang et al., 2018; Zhu and Cheng, 2022) and time since SS application (Rostagno and Sosebee, 2001), respectively. Studies have reported contrasting effects of SS application on HM migration in surface runoff. For example, Joshua et al. (1998) showed increases in rainwater soil infiltration under SS addition led to reductions in surface water runoff and sediment yield, and a lower associated risk of HM migration in surface runoff water, whereas Yang et al. (2023) found that applications of SS increased the risk of HM migration in the surface runoff, soil interflow, and sediment pathways, where risk was greatest through smallest aggregate fraction in sediment flow.

Dynamics of these soil pathways vary with ecosystem; for example, the greater biomass of plant litter at the soil surface of forests than in agricultural systems mitigates soil erosion by raindrops, leads to greater stability of soil aggregates (Li et al., 2014) and reduces the velocity of surface runoff, resulting in lower associated sediment transport (Zhu and Cheng, 2022). Thus, there is a need to understand the effects of litter cover on migration risks of SS HMs in forest soils, across forest soil flow pathways, to improve the environmental risk assessment and mitigation of potentially toxic soil amendments (Chu et al., 2021).

In this study, we quantified effects of SS application under contrasting levels of plant litter on concentration and cumulative migration of HMs in three soil flow pathways following field ambient and laboratory simulated rainfall events to assess the ecological risk of SS HMs. The objectives of the study are to: (1) test for effects of level of plant litter cover on forest surface water runoff, interflow, and sediment following SS addition; (2) test for relationships between of level of plant litter cover and HM migration in the three soil flow pathways; (3) check for consistency in HM runoff migrations between field and laboratory experiments; and, (4) provide a theoretical basis for the management of forest soil HM pollution following SS application.

2. Materials and methods

2.1 Ambient rainfall experimental design

The field ambient rainfall experiment was located in a *Eucalyptus urophylla* plantation created with tree spacings of 3×2 m in 1998 at Jiuzhuan Lake in the Dalingshan Forest Park, Guangdong Province, China ($22^{\circ}51'18.99''\text{N}$, $113^{\circ}45'22.44''\text{E}$), where soils comprise a sandy loam derived from granite (56.7% sand; 27.7% silt; 15.6% clay). In July 2021, we selected twelve 10×2 m experimental plots with comparable site conditions (east-facing, 15° slope) and equal tree density that were oriented with the longer sides aligning with slope direction; herbaceous plants were removed and irregular terrain was smoothed. The experimental plots comprised a surrounding

boundary wall, runoff collection pool, and water diversion channel. The 20-cm high boundary wall was constructed from cement bricks to prevent surface runoff to adjacent plots, while the runoff collection pool ($1 \times 1 \times 1$ m) situated at the down-slope plot edge and water diversion channel were covered with a PVC plastic board and gridded PVC film, respectively, to exclude rainwater and falling plant debris/litter, respectively.

2.2 Simulated rainfall experimental design

We simulated rainfall intensity of the climatic conditions of southern China, to emulate scenarios of heavy rainfall and intense soil erosion. The simulated rainfall experiments were carried out at the Institute of Eco-environment and Soil Sciences, Guangdong Academy of Sciences using soils collected from depths of 0–10, 10–20, and 20–30 cm in the *E. urophylla* plantation at Jiuzhuan Lake in November 2020. The soils were air-dried at room temperature in the laboratory for 2 weeks, when organic residues, roots, and large sand grains were manually extracted; then, the soils were passed through a 10-mm nylon sieve and mixed by profile depth, to ensure uniformity of distribution in soil aggregates within each layer.

The experiment comprised 12 stainless-steel soil flumes (1 m long $\times$ 0.3 m wide $\times$ 0.3 m high; 15° slope) filled with the collected soils arranged by layer and fitted with a water tank (1.2 m long $\times$ 0.4 m wide $\times$ 0.12 m high) with 150 holes (0.8 mm diameter) distributed across its base. Between March and August 2022, nine simulation rainfall events were conducted with a two-week interval between each simulated rainfall, where each event was administered at $0\text{--}5 \text{ L}\cdot\text{min}^{-1}$ for 60 min ($120 \text{ mm}\cdot\text{h}^{-1}$) using a variable power pump. A channel was fixed to the front of the flumes to collect surface runoff and accompanying sediment, and a hole at the lowest downstream point of the flumes facilitated the collection of interflow. Full details of the experimental design are reported by (Yang et al., 2023).

2.3 Treatments

Four treatments, comprising the addition of $6 \text{ kg} \cdot \text{m}^{-2}$ of SS at litter cover levels of 0, 0.75, and 1.5 kg m^{-2} , plus a control of zero SS and zero litter (CK), were applied to three replicates of the simulated rainfall soil flumes and ambient rain field plots. “0, 0.75, 1.5, CK” and “0F, 0.75F, 1.5F, CKF” were used to represent these treatments in the laboratory and field experiments, respectively. We sourced SS from the Guangzhou Datansha Wastewater Treatment Plant, Guangzhou City, Guangdong Province, China that was subjected to anaerobic composting for 60 days, followed by air-drying and sieving through a 10-mm nylon screen. The SS was mixed thoroughly, before it was incorporated with the top 10-cm soil profile and spread across the surface of the field plots and flumes. Dry plant litter was collected from a *E. urophylla* plantation in the arboretum of South China Agricultural University (23°15'56"N, 113°26'05"E) and applied it to the surface of experimental plots and flume, as appropriate; for field plots, plant litter levels were maintained at monthly intervals.

2.4 Sample collection and analysis of heavy metals

For the field experiment, we collected 12 samples of surface runoff in 200-mL polyethylene plastic bottles from the collection pools on the day following heavy rainfall events, from July 2021 to August 2022; immediately prior to collection, the runoff water in the collection pools was thoroughly mixed. Water samples were stored at 4 °C with 1 mL of HNO_3 to inhibit microbial activity and prevent the precipitation of heavy metal ions prior to analysis.

For the rainfall simulation experiments, we collected total surface runoff and soil interflow following each simulated event. Surface runoff and soil interflow were collected separately and left undisturbed for 30 min; then, the suspensions were transferred into 200-mL polyethylene plastic bottles, to which 1 mL of HNO_3 was added to inhibit microbial activity and prevent the precipitation of heavy metal ions. The treated samples were then stored at 4 °C prior to analysis.

Sediment deposited in the runoff collection pool was collected and then segregated into four size classes of aggregate (>1 mm; 1–0.25 mm; 0.25–0.05 mm; <0.05 mm) using the wet sieve method (Xiao et al., 2016; Huang et al., 2018); half of each size class sample was dried, weighed, and pulverized, while the remaining half was combined with the others as a composite sample of total sediment per plot.

The surface runoff, soil interflow and sediment samples were digested using the tri-acid (nitric-hydrofluoric-perchloric acid) method and subjected to closed microwave dissolution (CEM MARS6, Matthews, NC, USA), before concentrations of cadmium (Cd), chromium (Cr), nickel (Ni), lead (Pb), Cu, and Zn were determined using a Plasma atomic emission spectrometer (Leeman prodigy7 model, Teledyne LABS, OKC, USA) (Fang et al., 2017; Qi et al., 2020; Zuo et al., 2021). We calculated cumulative mass migrations of HMs from individual and cumulative rainfall events across the three soil flow pathways. We pulverized and sieved (<0.15 mm) samples of the experimental SS and soils prior to analysis of pH using a pH meter (AB33PH-F pH Meter, Ohaus, NH, USA) and total concentrations of Cd, Cr, Ni, Pb, Cu, and Zn (Table S1) that were determined. Similarly, we analyzed pH and HM properties of the litter (Table S2).

2.5 Potential ecological risk assessment

We used a modified ecological risk index (*RI*) (Li et al., 2022a) to evaluate the potential ecological risk to the surrounding environment posed by the release of HMs from SS applications, based on the ratio of the total amount of HMs released from SS during the nine simulated rainfall events to the total amount of background HMs in the control (Chu et al., 2021; Yang et al., 2023) for single and multiple soil flow pathways. Thus, *RI* was calculated as:

$$Ei = Ti \times \frac{Ci}{Co} \quad (1)$$

$$RI = \sum_{i=1}^n Ei \quad (2)$$

where E_i represents the risk factor of HM i ; T_i is the toxic-response factor for HM i , based on $Cu = Cr = Pb = Ni = 5$, $Zn = 1$, and $Cd = 30$ (Håkanson, 1980; Liu et al., 2021); C_i is the total amount of HM i in surface runoff, soil leachate or sediment; and, C_0 is total amount of background HM in the control.

2.6 Statistical analysis

All statistical analyses were performed in R 4.1.3 (R Core Team, 2021). Given a lack of normality of some data collected from the 0.75 and 1.5 kg plant litter m^{-2} treatments, we conducted a permutational-based multivariate analysis of variance (PERMANOVA; setseed:1234), using Kruskal-Wallis and Dunn's test in the "PMCMRplus" package. We used Bonferroni's post-hoc test to test for within-pathway treatment differences in volume of surface runoff, interflow, and sediment yield, and in concentrations and cumulative migrations of the HMs. Aggregate size class and soil flow pathway differences in relative abundance of cumulative HM migrations were visualized using a stacked bar alluvial chart using the "ggplot" and "ggalluvial" packages. Treatment effects were tested at $p < 0.05$ and presented data are means $\pm$ SE.

3. Results

3.1 Soil flow pathways under laboratory conditions

The volumes of surface runoff and soil interflow, and particularly sediment yield, were affected more by plant litter cover than addition of SS, where there were no differences in runoff and interflow volumes between the CK and 0 kg m^{-2} of plant litter treatment across individual and combined simulated rain events (Fig. 1a, 1b). Compared with the 0 kg m^{-2} of plant litter treatment, the volume of surface runoff under the 0.75 and 1.5 kg m^{-2} of litter treatments decreased by 19.3 and 85.2%, respectively, while the volume of soil interflow increased by 224.6 and 1624.3%, respectively (Fig. 1b).

Volume of sediment yields increased during the first three simulated rain events with the addition

of SS alone and decreased during events six to nine (Fig. 1c). While there were no effects of SS in the absence of plant litter on total sediment yield, there were increases in yield of sediment aggregates <0.05 mm and decreases in yields of aggregate classes 0.05–0.25 mm and 0.25–1 mm (Fig. 1d). Addition of SS with plant litter at 0.75 and 1.5 kg m⁻² reduced yields of all size classes of sediments, leading to almost zero sediment flow (Fig. 1e).

3.2 Sediment content of HMs under laboratory conditions

The concentrations of Cd, Cr, Cu, and Ni in the <0.05 mm and 0.05–0.25 mm aggregate fractions were notably higher than those in the 0.25–1 mm and >1 mm ones (Fig. S1). The addition of SS in the absence of plant litter increased total sediment concentrations of HMs by 28.5 to 149.4% (Fig. S1). Concentrations of Pb and Zn in the >1 mm aggregate fraction increased by 97.0 and 259.7% respectively, representing a 2–3 fold increase compared to other fractions, while there were increases in Cd concentration in 0.05–0.25 mm and 0.25–1 mm fractions (113.5 and 116.4%, respectively) and increases in Ni concentration in the 0.05–0.25 mm fraction (81.6%), representing 1.5–2.3 increases over other fractions (Fig. S1).

Cumulative migrations of the HMs increased with the addition of SS alone, particularly Cd and Zn that increased by 80.4 and 121.9%, respectively, primarily due to increases in content in the <0.05 mm fraction (Fig. 2a, Fig. S2). The cumulative migrations of Cd, Cr, Cu, and Ni in the smallest fraction under addition of SS alone accounted for approximately 50% of the total HM migration in sediment; in contrast, addition of SS alone led to a decrease in cumulative HM migration in the 0.05–0.25 mm and 0.25–1 mm fractions (Fig. 2b). Addition of SS decreased cumulative migrations of Cr, Cu, and Pb in the 0.05–0.25 mm fraction (Fig. S2).

3.3 Soil flow pathway migration of HMs under laboratory conditions

There were effects of addition of SS and plant litter cover on migration of HMs via surface runoff, interflow, and sediment (Fig. 3). From the second simulated rainfall event, there were increases in

cumulative migrations of the six HMs in surface runoff and sediment under addition of SS alone and addition of SS and plant litter led to reductions in HM migration in runoff, with zero HMs detected in sediment (Fig. 3). Addition of SS and plant litter increased cumulative interflow migrations of the HMs, with greatest cumulative migrations under 1.5 kg of litter m^{-2} following each simulated rain event; from the seventh simulated rainfall event, there was a slight increase in the cumulative migration of Cd and Cu under the addition of SS alone (Fig. 3). Migration velocity of the six HMs remained relatively constant in the runoff and interflow pathways, while in the sediment, it steadily slowed in the absence of SS and litter (CK) and under SS alone under ongoing rainfall events (Fig. 3).

Cumulative migration of the HMs was 21.5–113.1% greater under the addition of SS alone compared with the CK, while addition of 0.75 and 1.5 kg litter m^{-2} along with SS led to 87.1–97.2% decreases in cumulative HM migrations compared with the addition of SS alone, primarily due to a decrease in sediment migration of HMs (Fig. 4a). Sediment was the dominant pathway for the migration of the HMs in the absence of litter, accounting for 89.5–97.1% of migration and exceeding that of runoff and interflow by an order of magnitude (Fig. 4b). In contrast, soil interflow was the main pathway for the migration of Cd and Zn in the presence of litter, where it accounted for 95.8 and 92.8% of migrations, respectively, under 1.5 kg of litter m^{-2} , whereas runoff and interflow were the main Cr, Cu, Ni, and Pb migration pathways under 0.75 and 1.5 kg of litter m^{-2} , respectively; there was no contribution to the cumulative migration of HMs from the sediment pathway (Fig. 4b).

3.4 Consistency in surface runoff HMs between field and laboratory

Field and laboratory data showed addition of SS and plant litter led to a decrease in the volume of surface runoff (Fig. 1, Fig. S3), while there was an order of magnitude larger level of cumulative migration of HMs in the laboratory experiments than in the field experiments (Fig. 5a, Fig. 3) and

similar levels of runoff concentration of the six HMs between the two experiments (Fig. 5b, Fig. S4). There was an increase in runoff HM concentrations under addition of SS alone and, with the exception of Cr and Cu in the field and Zn in the laboratory, there were decreases in runoff HM concentration under the addition of SS plus plant litter (Fig. 5b, Fig. S4).

Following 12 heavy rain events in the field experiment, the cumulative migration of Cd, Cu, Ni, and Pb in surface runoff was greater with the addition of only sewage sludge (0%-F) compared to the control (CK-F). However, adding 0.75 to 1.5 kg/m² of litter along with the SS resulted in a 14.2–56.9% reduction in cumulative HM migration compared to SS alone (Fig. 5a), aligning with the laboratory experiment results (Fig. 3). Since the heavy rainfall events in August 2021, the migration rates of six HMs in surface runoff increased under SS addition in the field experiments, but then gradually slowed, consistent with the trends observed in the laboratory experiments (Fig. 3).

3.5 Risks of HM migration under laboratory conditions

Ecological risk assessment revealed a higher *Ei* for Cd migration in runoff, soil interflow, and sediment (“moderate”) under addition of SS alone, a higher Cd interflow *Ei* (“considerable”) under addition of SS plus litter, and higher Cu interflow *Ei* (“considerable”) under addition of SS and litter at 1.5 kg m⁻² (Table 1). Across the three pathways, HM *Ei* under addition of SS alone was higher for Cd (“moderate”) than the other HMs (“low”) (Table 1).

Risk assessment of interflow migration of all HMs under addition of SS and litter at 0.75 and 1.5 kg m⁻² showed *RI* of “moderate” and “considerable”, respectively, primarily due to the high risks of Cd and Cu (Table 1). In contrast, *RI* for runoff and sediment HM migrations tended to be “low” across treatments, and zero for sediment pathways with the addition of litter (Table 1). The *RI* for migration of all HMs across the three pathways was “low” across treatments (SS alone > CK > 0.75 and 1.5 kg litter m⁻²).

4. Discussion

4.1 Effects of SS and litter cover on soil and water erosion

Surface runoff and sediment yield tend to be reduced by the presence of leaf and woody litter at the soil surface (Li et al., 2014; Zhu and Cheng, 2022), due to the interception and retention of rainfall that slows the drainage process (Nainar et al., 2021) and reduces detachment and splash erosion of soil aggregates (Li et al., 2014; Felix et al., 2023). We found decreases in surface runoff under the addition of SS and litter in the field and laboratory experiments; however, surface runoff in the field was an order of magnitude lower than in the laboratory, possibly due to interception of rainfall by the canopy in the field (Inokoshi et al., 2023).

Reductions in the velocity and volume of surface runoff increase levels of soil interflow that may further contribute to lower levels of sediment yield (Sun et al., 2022; Yang et al., 2023). Indeed, data from our simulated rainfall experiments showed simultaneous reductions in surface runoff and increases in soil interflow volume with the addition of litter, particularly at the rate of 1.5 kg m⁻². Thus, these findings indicate that plant litter facilitates the redistribution of water within the soil profile, promoting deeper infiltration (Jourgholami et al., 2021).

We observed that the application of SS initially increased sediment yield, before decreasing with time, possibly due to its combination with soil that may have led to a collapse in aggregates, clogging of water-conducting pores, and subsequent sealing of the soil surface (Mamedov et al., 2014). Previous research has shown that the clogging of soil pore spaces following a series of simulated rain events diminishes the anti-erosion effects of the addition of SS (Bruggeman and Mostaghimi, 1993), likely resulting in increased fragmentation of aggregates and rapid migration of <0.05 mm aggregates from the topsoil; however, there may be subsequent improvements in soil structure and stability as SS becomes integrated into the soil (Maria et al., 2007; Da Silva et al., 2021), thereby reducing sediment yield.

4.2 Effects of SS and litter cover on sediment HMs

Our observation that smaller sediment aggregates (<0.05 mm and $0.05\text{--}0.25$ mm) contain higher concentrations of HMs (Cd, Cr, Cu, Ni) than larger aggregates is consistent with previous research (Li et al., 2022; Yang et al., 2023), due to higher levels of soil organic matter and larger surface area-to-volume ratios in smaller aggregates that facilitate HM accumulation via co-precipitation, occlusion, sorption, and complexation (Huang et al., 2014; Fang et al., 2023). We found increases in total concentration of sediment HMs following the addition of SS, likely due to variations in HM adsorption capacity with sediment aggregate particle size (Huang et al., 2014; Liu and Han, 2020). For example, initial adsorption rates of Cu and Zn in red paddy soils increased with decreasing aggregate particle size, except for the $0.05\text{--}0.002$ mm fraction, where Cu adsorption amounts were 2–3 times higher than Zn in the same equilibrium concentration range (Huang et al., 2014; Liu and Han, 2020).

We found the addition of SS led to an increase in the cumulative migrations of sediment HMs, particularly of Cd and Zn, primarily due to the highest concentration and proportional migration of the smaller fraction aggregates (<0.05 mm), as reported previously (Huang et al., 2018; Yang et al., 2023). We found that SS concentrations of Cd and Zn were 3–4 times higher than those of forest soil and, given the higher background levels of Cd content of subtropical forest soils in China than farmland and bare land (Deng et al., 2018), SS applications to this type of forest may pose high environmental risks of Cd pollution via water and soil losses (Chu et al., 2023). The increase in proportion of cumulative HM migrations in the smallest aggregate fraction (<0.05 mm) following the addition of SS contrasted with decrease in the $0.05\text{--}0.25$ mm and $0.25\text{--}1$ mm aggregate fractions, indicating the key role of smaller aggregates in the environmental risk of SS HM migrations (Yang et al., 2023).

4.3 Effects of litter cover on soil flow pathway HM migrations

Our field and laboratory experiments showed that the application of SS increased surface runoff concentration and cumulative migrations of HMs, supporting previous studies (Peyton et al., 2016; Chu et al., 2021; Yang et al., 2023), where the addition of plant litter mitigated these rises, likely due to the physical barrier formed by the litter and its water-holding capacity that reduce surface water runoff (Pandey and Raghubanshi, 2022), and the total amount of Cd adsorbed by plant litter constitutes an extremely small fraction of the overall HM migration (Table S2).

Data from the field experiment show the velocity of HM migration through surface runoff escalated markedly under intense rainfall, particularly under the addition of SS, whereas there was such no increase with the co-addition of plant litter. Given these findings indicate that heavy rainfall events may trigger rapid increases in surface runoff HM migration, we suggest additional experiments are required to ascertain the combined impact of rainfall intensity and litter cover on HM migration under SS applications (Zhang and Wang, 2020), particularly in the subtropical regions of China where heavy rainfall events are becoming increasingly frequent (Zhou et al., 2011).

We found that levels of cumulative surface runoff migrations of HMs were markedly lower in the field than in the laboratory, likely due to effects of rainfall interception by the tree canopy (Zhou et al., 2016). In contrast, we found that addition of plant litter with SS under simulated rainfall events led to increases in interflow migration of HMs, with highest levels recorded under 1.5 kg of litter m⁻², likely as a result of greater water infiltration and percolation, despite simultaneously reducing interflow concentrations of HMs (Perez-de-Mora et al., 2007). The consistency of HM runoff migrations between field and laboratory experiments, as well as the consistency of HM migration patterns in sediment and soil interflow, need further detailed study.

The dominant pathway for HM migration was found to vary depending on the addition of SS, as reported by (Yang et al., 2023), along with rate of plant litter addition, where we found that

sediment was the main HM migration pathway under SS addition alone, likely due to the high HM content and erodibility of exposed sludge-amended soils. In contrast, there was virtually no sediment production under the addition of plant litter, and the main HM migration pathways were surface runoff and soil interflow. These results underscore the effectiveness of plant litter in the reduction of HM migration via the sediment pathway as a promising strategy for HM risk management (Wu et al., 2018).

Under the addition of SS, we found surface runoff and interflow were the main migration pathways for Cr, Cu, Ni, and Pb under the co-addition litter at 0.75 and 1.5 kg m⁻², respectively, indicating HM variation in migration responses and greater potential risks of groundwater pollution under the addition of plant litter. Thus, we suggest further research to understand HM migration pathway responses to litter to improve HM pollution management and policy making. And the changes of chemical speciation in HM migration need further study to enhance our understanding of potential reaction pathways and migration mechanisms (Chu et al., 2023).

4.4 Effects of SS and litter cover on HM migration risk

This study has shown that the addition of SS escalated the ecological risk of Cd migrations via surface runoff, soil interflow, and sediment, supporting previous studies have identified Cd, along with Cu and Ni, as the primary elements in SS of high ecological risk to soils (Fang et al., 2017; Qi et al., 2020; Zuo et al., 2021). Although Cd represents the lowest HM concentration in SS, its ions pose a high level of risk to groundwater quality through interflow or confluence with surface runoff, due to high levels of aggregation, persistence, and environmental mobility (Notten et al., 2005; Tóth et al., 2016). For example, the primary risk of Cd pollution is migration from soils to water bodies via surface runoff and underground leaching (Yang et al., 2018; Liu et al., 2022), where greater amounts enter water in runoff as suspended solids, particulate matter, and sediment than through aqueous migration (Demirak et al., 2006). While our study demonstrates plant litter

mitigation of ecological risks of Cd by reducing surface runoff and sediment migration pathways, HM risks through the soil interflow pathway were increased and estimated to be “moderate” and “considerable” under litter at 0.75 and 1.5 kg m⁻², respectively, primarily due to the contribution of Cd and Cu.

The *RI* for the six HMs across the three soil flow pathways indicated that the addition of SS alone and with litter pose a low level of ecological risk, primarily because plant litter reduces sediment yield. Thus, addition of litter in combination with SS to subtropical forest soils may mitigate HM migrations, but it may also increase the risk of Cd migration, depending on soil pathway, highlighting the importance of Cd risk assessment and management in SS soil amendment programs (Chu et al., 2021; Wang et al., 2023).

Conclusion

Our experiments quantified HM migrations via surface runoff, interflow, and sediments following addition of SS alone and in combination with contrasting rates of plant litter. While the addition of SS increased HM concentrations and migrations in surface runoff, soil interflow, and small-sized sediments, and increased sediment yield, the co-addition of plant litter mitigated these impacts and changed the Cd migration pathway from surface runoff to interflow that further reduced the risk of HM contamination. These findings underscore the environmental risks associated with the application of SS, while also highlighting the potential of plant litter cover as a management strategy for the mitigation HM pollution. Concentrations of the six HMs in surface runoff were similar between the field and laboratory experiments, although cumulative runoff migrations were higher in the laboratory. We suggest further comprehensive field experiments are needed to assess the risk of HM pollution caused by soil interflow and sediment under SS applications to subtropical forest soils.

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378 **Tables**

379 **Table 1. Estimation of ecological risks of heavy metals by pathway and sewage-litter treatment in the laboratory**
 380 **experiments.**

Pathway	Litter rate (kg m ⁻²)	Ei						RI	
		Cd	Cr	Cu	Ni	Pb	Zn	Value	Risk level
Surface runoff	CK	30.00	2.00	5.00	5.00	5.00	1.00	48.00	Low
	0	63.12	3.98	6.72	7.26	9.14	1.41	91.64	Low
	0.75	33.66	2.64	4.11	4.44	3.90	1.03	49.78	Low
	1.5	5.28	0.45	0.62	1.04	0.65	0.21	8.25	Low
Soil interflow	CK	30.00	2.00	5.00	5.00	5.00	1.00	48.00	Low
	0	47.52	1.40	9.67	5.79	4.36	1.31	70.04	Low
	0.75	108.49	4.40	22.69	11.91	11.65	2.68	161.83	Moderate
	1.5	170.73	16.94	66.59	25.65	25.65	6.54	312.10	Considerable
Sediment	CK	30.00	2.00	5.00	5.00	5.00	1.00	48.00	Low
	0	54.13	2.54	6.05	7.41	7.02	2.22	79.37	Low
	0.75	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-

	1.5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-
Multi-Pathway	CK	30.00	2.00	5.00	5.00	5.00	1.00	48.00	Low
	0	54.37	2.61	6.07	7.39	7.10	2.13	79.68	Low
	0.75	6.15	0.15	0.17	0.22	0.32	0.16	7.17	Low
	1.5	7.01	0.07	0.22	0.23	0.30	0.22	8.05	Low

381

382 Classification of *Ei* risk: low < 40; $40 \leq$ moderate < 80; $80 \leq$ considerable < 160; and. $160 \leq$ high < 320. Classification of *RI*

383 risk: low < 150; $150 \leq$ moderate < 300; $300 \leq$ considerable < 600; high $\geq$ 600.

Figure captions

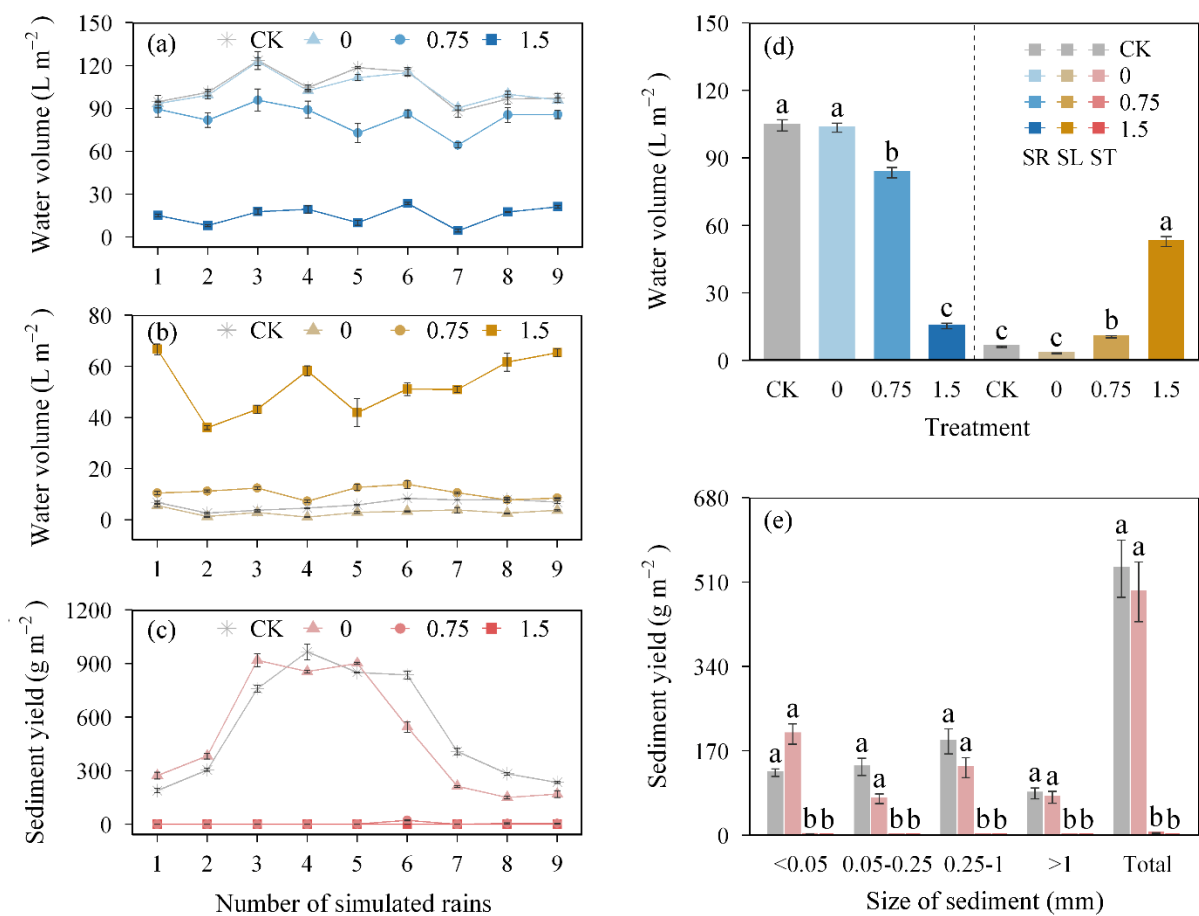
Fig. 1. Effects of treatment on volumes of surface runoff (SR) (a), soil interflow (SI) (b), and sediment yield (ST) (c), across simulated rainfall events (SR and SI) (d), and sediment size classes (ST) (e). Data are means $\pm$ SE; different lowercase letters indicate treatment differences at $p<0.05$.

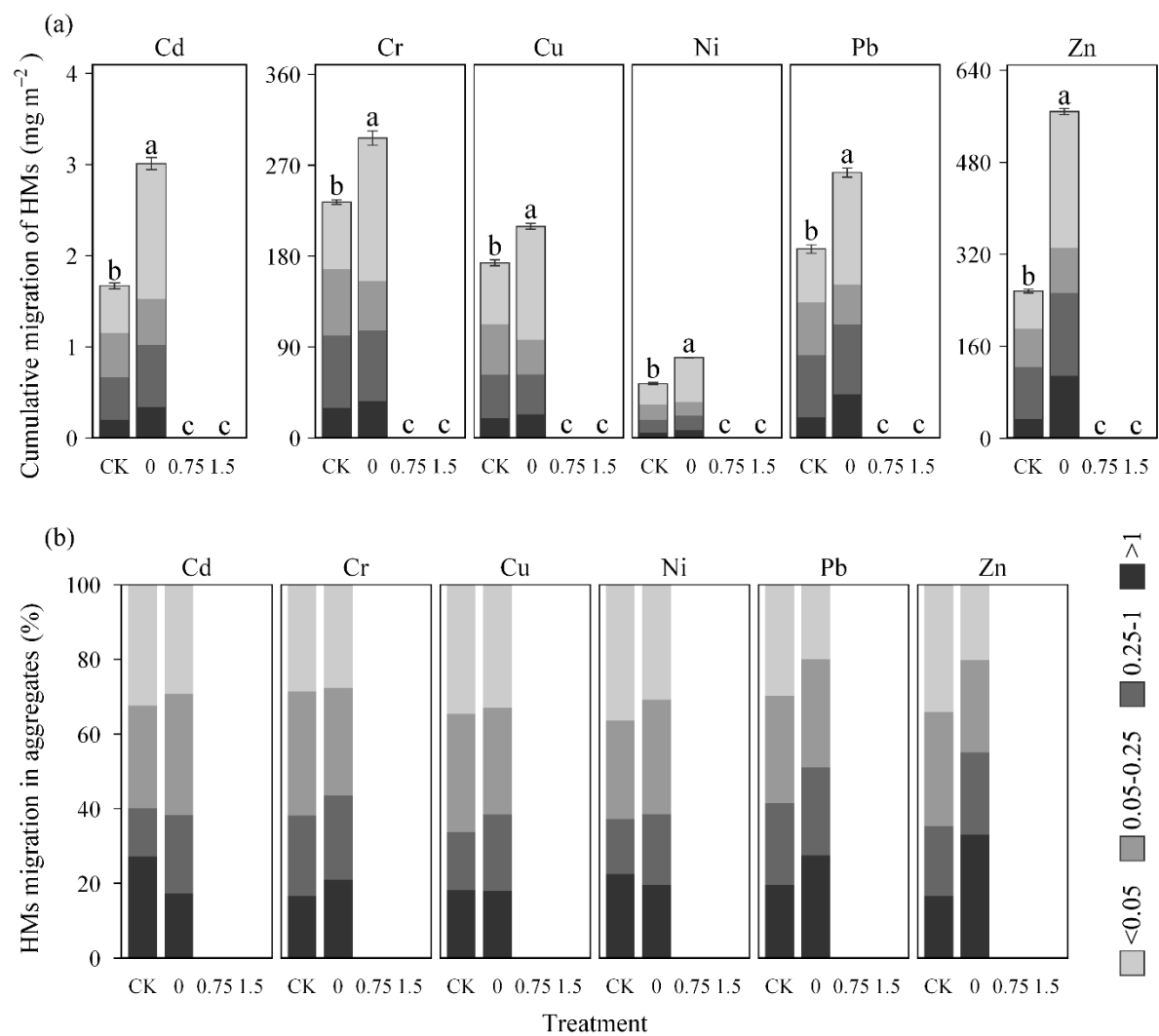
Fig. 2. Effects of treatment on cumulative heavy metal migrations (a) and proportion of heavy metals in aggregate size classes (b) under laboratory conditions. Data are means $\pm$ SE; different lowercase letters indicate treatment differences at $p<0.05$.

Fig. 3. Effects of treatment on cumulative migrations of six heavy metals in surface runoff, soil interflow, and sediment across simulated rainfall events. Data are means $\pm$ SE.

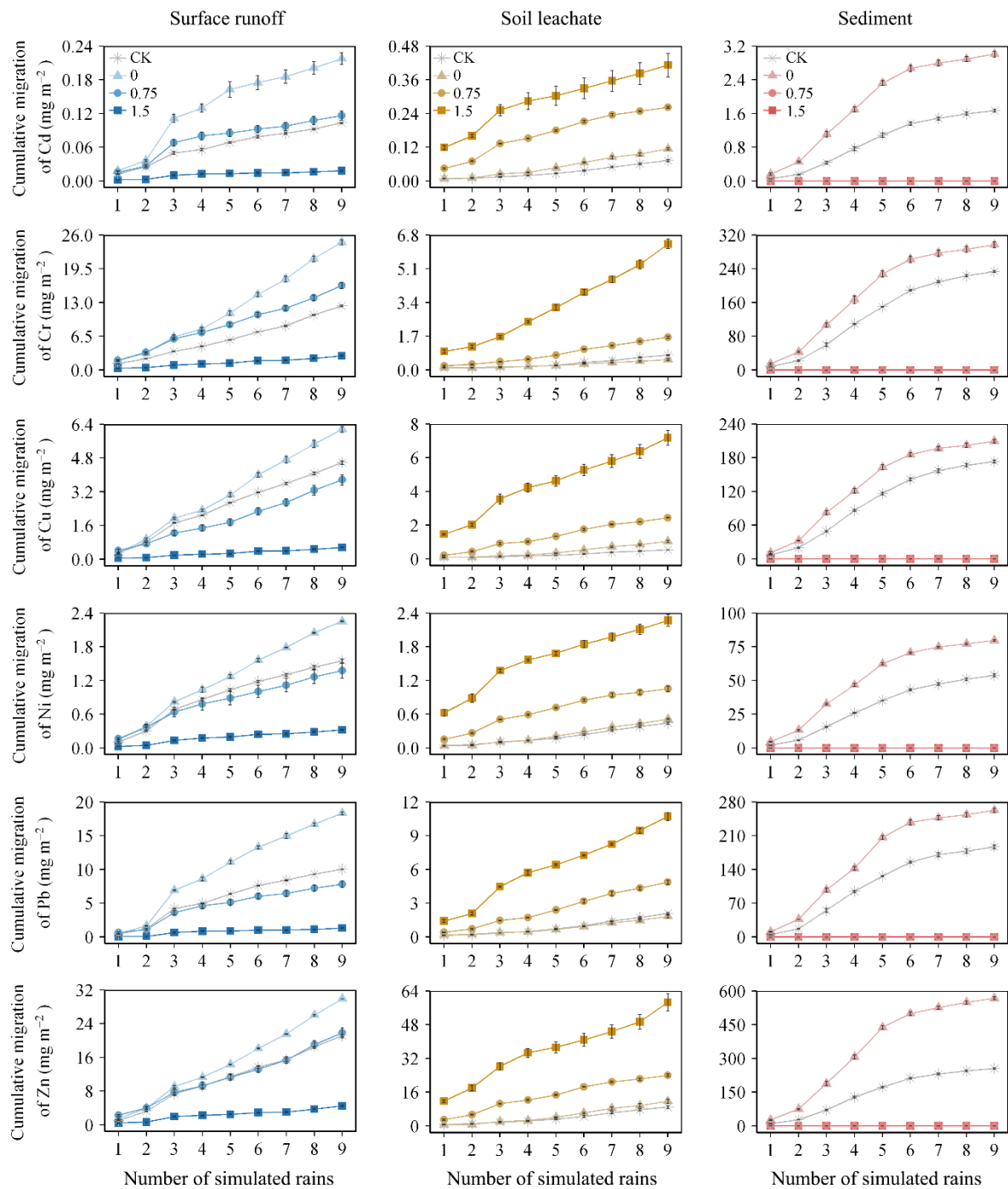
Fig. 4. Effects of treatment on cumulative heavy metal migrations (a) and proportion of heavy metals (b) in surface runoff (SR), soil interflow (SI) and sediment (ST) under laboratory conditions. Data are means $\pm$ SE; different lowercase letters indicate treatment differences at $p<0.05$.

Fig. 5. Effects of treatment on cumulative heavy metal migrations (a) and heavy metal concentrations under field conditions. Data are means $\pm$ SE; different lowercase letters indicate treatment differences at $p<0.05$.





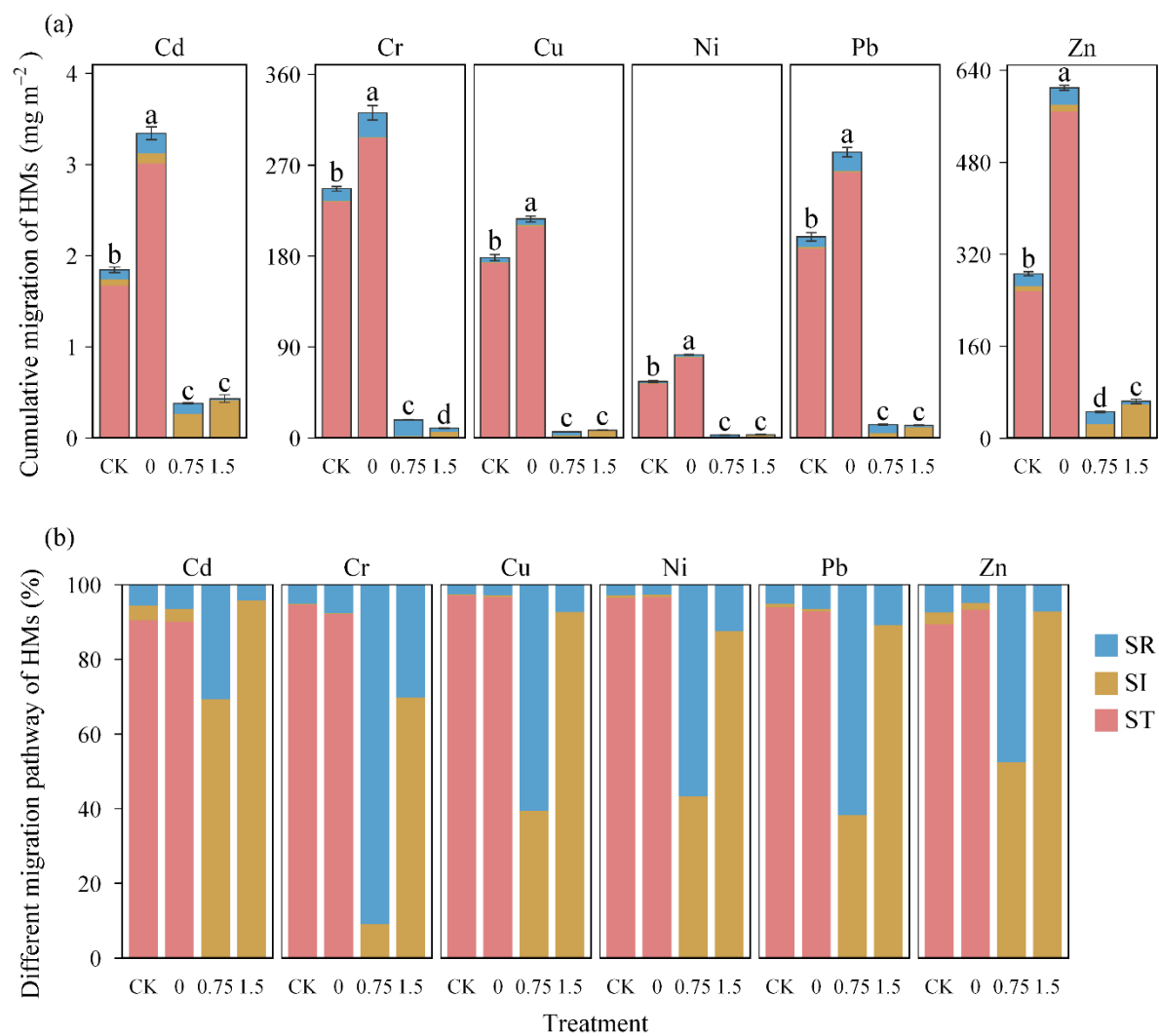
401 **Fig. 3**



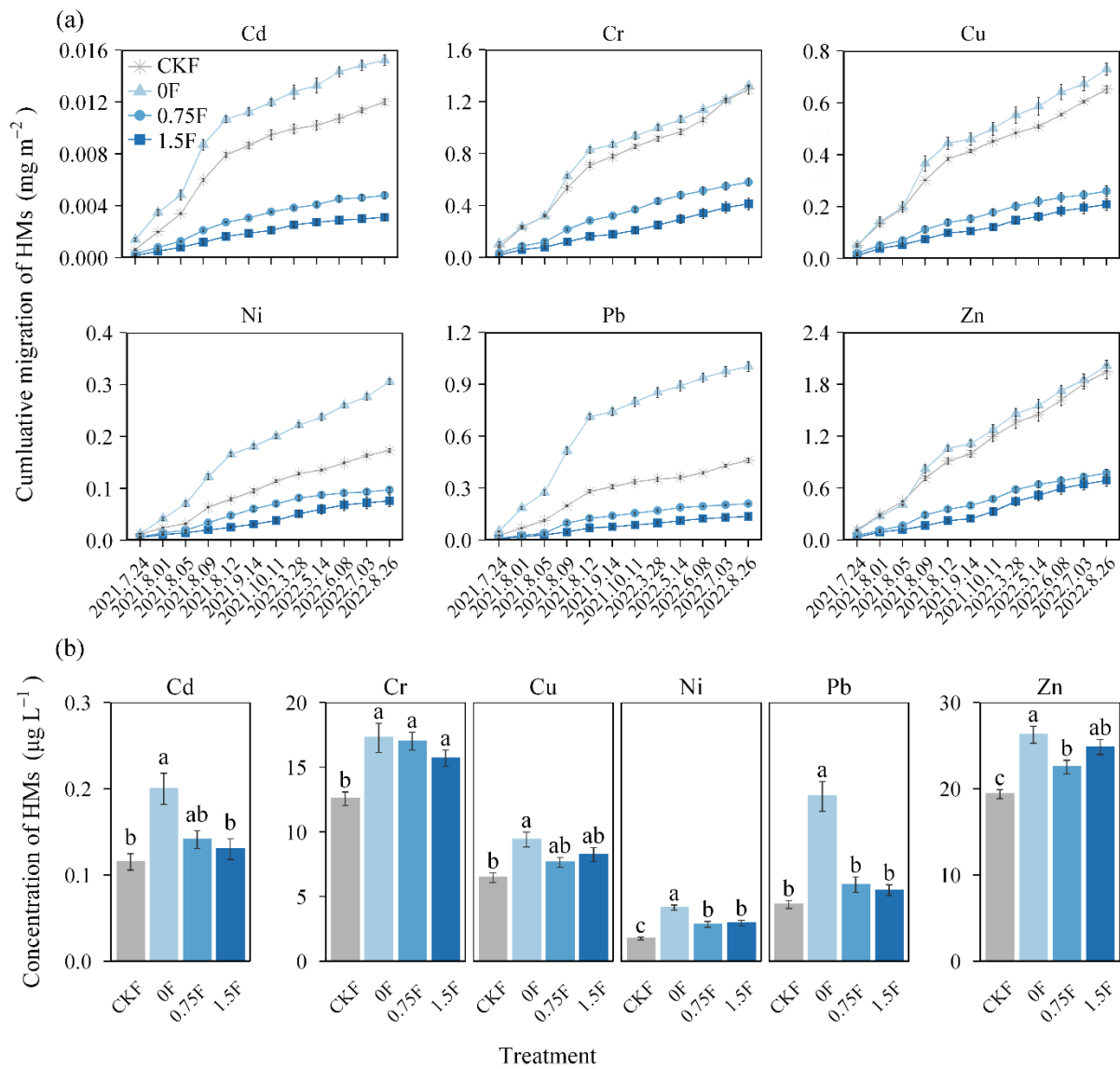
402

403

404 **Fig. 4**



405



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