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Research Paper

The effects of microplastics on heavy metals bioavailability in soils: a meta-analysis

Qiuying An [a,b], Tong Zhou [a,b], Ce Wen [a,b], Changzhou Yan [a,*]

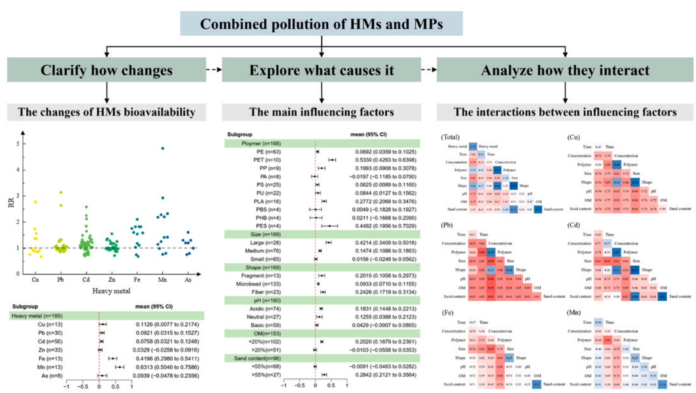
[a] Key Laboratory of Urban Environment and Health, Fujian Key Laboratory of Watershed Ecology, Institute of Urban Environment, Chinese Academy of Sciences, Xiamen 361021, China

[b] University of Chinese Academy of Sciences, Beijing 100049, China

HIGHLIGHTS

- PE, PET, PP, PS, PU, PLA, and PES might improve the bioavailability of Cu, Pb, Cd, Fe, and Mn, whereas PA had no impact.
- The effect of hydrolyzed MPs containing nitrogen elements on the bioavailability of HMs is relatively weak.
- Larger size MPs and acidic, low organic matter and high sandy soil can increase the bioavailability of HMs in soils.
- There is an interaction between MPs characteristics and soil physicochemical indexes on the bioavailability of HMs.

GRAPHICAL ABSTRACT



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ABSTRACT

The combined pollution of heavy metals and microplastics is common in natural soil environments. Here, we collected 790 data sets from 39 studies to investigate the effects of microplastics on heavy metal bioavailability. The results showed that microplastics could increase the bioavailability of Cu, Pb, Cd, Fe, and Mn. The heavy metal bioavailability was positively correlated with microplastic size, soil sand concentration, and exposure time, but negatively correlated with soil pH and organic matter. The bioavailability of heavy metals can be promoted by microplastics of all shapes. Hydrolysable microplastics, which contain N, might have less influence. Furthermore, the size of microplastics and soil organic matter were positively correlated with the acid-soluble and reducible fractions of heavy metals, while the microplastic concentration, soil pH, and exposure time were positively correlated with the oxidizable fractions of heavy metals. The interaction detector results indicated that there was an interaction between microplastic characteristics, especially polymer types, and soil physicochemical indexes on the bioavailability of heavy metals. These findings suggested that long-term combined pollution of microplastics and heavy metals might increase heavy metal bioavailability in soils, thereby extending their migratory and hazardous range and bringing further risks to the environment and public health safety.

* Corresponding author.
E-mail address: czyan@iue.ac.cn (C. Yan).

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1. Introduction

Plastics are used extensively in various fields due to their portability, excellent performance, and low cost [1,2]. In each of the previous five years, more than 300 million tons of plastic were produced worldwide. With the COVID-19 pandemic, this number increased to 390.7 million tons in 2021 [3]. The amount of plastic garbage in the natural environment is predicted to reach 12,000 million tons by 2050 [4]. Among them, plastics in the soil are 4–23 times more abundant than in the ocean [5], and it can be transported to other ecosystems via groundwater and surface runoff [6]. Therefore, soil as a source and sink of plastic pollution requires widespread attention.

Soil is the basis of nearly all territorial environments and is inextricably linked to human production and life [7]. Due to industrial production, agricultural production, transportation, etc., soil is usually polluted with heavy metals (HMs) [8,9]. Plastics were degraded to microplastics (MPs) in soils with diameters less than 5 mm by photodegradation, biodegradation, and weathering mechanisms, which can persist for hundreds of years [10,11]. This implies that MPs and HMs would coexist and pollute the soil for a long period of time. Actually, previous research has discovered that the distribution of MPs pollution in China coincides with that of HMs [12]. The combined pollution phenomenon has also been discovered in the Persian Gulf in southwest Iran [13,14], North Rhine-Westphalia in western Germany [15], Faisalabad in northeast Pakistan [16], Cooch Behar in eastern India [17], Moknine in east-central Tunisia [18], and eastern Sohag governorate in Egypt [19]. As can be seen, the combined pollution of MPs and HMs has become a pressing issue in the global environmental field, but there have been few in-depth studies (Fig.S1a blue area, and Fig.S1b).

Microplastics are frequently thought as effective transporters of other contaminants because of their hydrophobicity and huge surface area [20]. By physical adsorption [21], chemical coprecipitation [22], and formation of complexes with hydrous oxides [23], it can directly adsorb HMs and alter the distribution of HMs in soils [24]. In addition, MPs may have an impact on microbial communities and soil physicochemical indexes (pH, organic matter, porosity, etc.), which may have an indirect impact on the chemical speciation of HMs [25]. According to Yu et al. [22], MPs could increase the organic-bound HMs fraction in soil while lowering their bioavailability. Li et al. [26] discovered the opposite phenomenon, in which it allowed them to move more easily in terrestrial environments. However, the toxicity of HMs to terrestrial organisms is intimately correlated with their mobility and bioavailability [27]. Hence, it is crucial to clearly investigate how HM bioavailability was affected by MPs under the combined pollution condition in order to understand the environmental risks.

The interaction of MPs and HMs in soils was extremely complex and frequently affected by a variety of factors, which can be divided into MPs characteristics, soil physicochemical indexes and microbial community. Among them, MPs polymer type and size, soil organic matter (OM) and sand content received the most attention (Fig.S1a red area). Besides that, MPs concentration and shape, as well as soil pH and exposure time, could also influence HMs bioavailability in combined pollution environments. A high concentration of polyethylene (PE) could enhance HMs bioavailability [26]. Polystyrene (PS) microbeads could decrease Cd bioavailability [28], whereas the fragments can increase it [29]. By changing the surface charge amount of soil particles, the pH can alter the HMs availability [30]. Meanwhile, exposure time was also crucial to HMs bioavailability. Yu et al. [31] discovered that within 45–120 d, soil HMs bioavailability significantly changed. Although some studies have demonstrated that HMs bioavailability was more sensitive to MPs at smaller aggregate fractions [22,31], as far as we were aware, the influence of soil texture had not been thoroughly investigated. The addition of MPs and changes in soil physicochemical indexes can also alter the HMs bioavailability by affecting the composition of soil microbial communities [32]. Several meta-analysis on the distribution of MPs in soils and their impacts on soil physicochemical indexes, as well as the toxic effects of microorganisms, plants, and animals, have previously been published [33,1,34,35]. However, they did not take into account the effects of MPs on HMs bioavailability under the condition of MPs and HMs combined pollution in soils and the interactions between different influencing factors.

In order to effectively integrate and analyze the relevant data of HMs bioavailability under the condition of MPs and HMs combined pollution from the current literature, a systematic procedure for literature collection, data screening and statistical analysis was carried out in this study [36]. The purpose of formulating and implementing this procedure were: (1) to clarify the impact of MPs on the bioavailability of different HMs in soils; (2) to explore the main factors that affect soil HMs bioavailability under the condition of MPs and HMs combined pollution; (3) to analyze the interaction relationship between these influencing factors. To our knowledge, this is the first investigation on the effects of MPs on HMs bioavailability in soils and their influencing factors by using meta-analysis.

2. Literature and methods

2.1. Literature collection

As shown in Fig.S1a, the primary pollutants associated with MPs in soils were HMs and persistent organic pollutants. Our study mainly focused on the combined pollution of HMs and MPs in soils, and the relevant literatures are mainly concentrated in 2021 and 2022 (Fig.S1b). All articles published before March 30, 2023, in the Web of Science Core Collection database were searched using the keywords “microplastic”, “heavy metal”, “soil”, “terrestrial environment”, and “terrestrial system”. The combination of keywords was: (microplastic) AND (“heavy metal”) AND (soil OR “terrestrial environment” OR “terrestrial system”). Finally, 362 articles were selected, and the number of published articles has continuously increased since 2014, reaching a high (33.6%) in 2021 and continuing now (Fig.S1c). The research primarily focused on nonbiodegradable MPs (PS, PE, PVC) and HMs (Cd, Pb) (Fig.S2). Furthermore, the primary research directions were the polluted environment and ecological risks, while HMs bioavailability was comparatively poorly studied (Fig.S2). According to the analysis of major publication journals, these studies primarily focused on environmental sciences, ecology, toxicology, and engineering technology (Fig.S1d).

To further increase the relevance and comparability of the data, the articles were screened based on the following criteria (Fig. 1): (1) The experimental group (added MPs) and the control group (no MPs) need to coexist, and the samples need to have more than three replicates. (2) Experiments must be conducted in soil to exclude the effect of soil containing MPs on HMs in overlying water. (3) Pollutants are only MPs and HMs, and combined pollution with other pollutants is excluded. (4) Articles that do not include HMs bioavailability (chemical morphology or bioavailability) or only measure the adsorption of HMs by MPs without measuring soil should also be excluded. (5) The soil used must be collected from natural field environments, excluding studies using artificial soil. Meanwhile, if multiple articles use the same sampling site soil, only the one with the earliest publication date will be retained. Finally, a total of 39 studies (12 combined pollution, 27 bioavailability), 190 study groups, and 790 data sets were chosen.

2.2. Pre-treatment of data

For all articles, the means and standard deviations of the experimental and control groups were mentioned, and for data that could not be obtained directly from the original text, GetData Graph Digitizer (v.2.26) was used to extract the graph. All pairs were treated independently in each study for different MPs concentration, polymer type, size, and shape. In order to more thoroughly investigate the effects of various factors on the HMs bioavailability, the size of MPs in soil was categorized into large (≥500 μm), medium (10–500 μm) and small (≤10 μm), according to Liu et al. [1]. The soil itself: pH was categorized into acidic (≤6.5), neutral (6.5–7.5) and basic (≥7.5); OM was categorized into ≤ 20% and > 20%; Sand content was categorized into ≤ 55% and > 55%. The polymer types and shapes of MPs were recorded in their real conditions. There was no heterogeneity among the subgroups, thus the exposure time and concentration of MPs were not grouped (Table S1).

2.3. Meta-analysis

If standard error (SE) was used when extracting the data, it is transformed to standard deviation (SD) (Eq.1); If just the mean was provided without the SD, the SD should be calculated at 10% of the mean.

$$SD = SE \times \sqrt{n} \tag{1}$$

where n is the number of samples.

The random-effects model meta-analysis was conducted using MetaWin 2. The research data were matched according to the experimental and control groups, the response ratio (RR) was used as the effect size for comparison, and the logarithm was obtained for further analysis (Eq. 2 and Eq. 3).

$$RR = \frac{\bar{x}_e}{\bar{x}_c} \tag{2}$$

$$\ln RR = \ln\frac{\bar{x}_e}{\bar{x}_c} \tag{3}$$

where $\bar{x}_e$ and $\bar{x}_c$ stands for the means of the experimental and control groups, respectively.

The variance (V) and 95% confidence interval (CI) corresponding to lnRR were calculated by Eq. 4 and Eq. 5, respectively. If the 95% CI did not overlap with zero, the effect size was regarded as statistically significant ($p < 0.05$).

$$V = \frac{SD_e^2}{n_e\bar{x}_e^2} + \frac{SD_c^2}{n_c\bar{x}_c^2} \tag{4}$$

$$95\%CI = \ln RR + 1.96 \times \sqrt{V} \tag{5}$$

where SD_e and SD_c were the SD of the experimental and control, respectively, while n_e and n_c were the sample numbers for the experimental and control, respectively.

To compare the heterogeneity of each effect size among groups, the between-group Q test was also performed. Significant Q values (Q_M) indicated significant differences in effect size among groups ($p < 0.05$). The publication bias of the articles that were included was verified using Rosenthal's method. Due to Rosenthal's fail-safe number was substantially bigger than 5 n + 10 (n is the number of observations), the results were unaffected by the potential publication bias in some variables, and there was no discernible publication bias in the majority of the variables (Table S2).

2.4. The interaction between influencing factors

To measure the contributions of each factor and reveal how they interact, the Geodetector analysis model was employed (http://www.geodetector.cn/). In compared with common correlation analysis and regression analysis, it had the advantage of quantifying the influence and interaction of each factor on the dependent variable without making linear assumptions [36]. The index q value for evaluating the driving force or explanatory force of the independent variable on the dependent variable ranges from 0 to 1. The stronger the influence of the independent variable on the dependent variable, the higher the q value.

3. Result and Discussion

3.1. Situation and source of HMs and MPs in combined polluted soils

As shown in Fig. 2a, HMs such as Cr, Cd, Pb, Cu, Ni, As, Zn, Fe, and Mn were discovered in sediments and farmland, while other metals (Hg, Al, and Mg) which were only mentioned in a single paper had not yet been subjected to analysis. With the exception of Cd and As, the concentration of HMs in farmland was greater than that in sediments. The major causes of HMs contamination in farmland were farming operations, natural processes, and atmospheric deposition [37]. Chemical fertilizers and pesticides frequently include HM elements including Cd, Zn, Cu, and As, and using them inappropriately may be one way to pollute soil [38]. Second, industrial and residential sewage, which frequently contained high levels of HMs, was mostly utilized to irrigate agriculture at the present time, causing the HMs to progressively accumulate in the soil [39]. Some soils had significant background levels of HMs, and natural weathering would release HMs from the parent material into the soil [37]. In addition, automobile exhaust emissions, waste incineration, and dust generated by the metallurgical industry will further aggravate soil HMs pollution through atmospheric deposition [40].

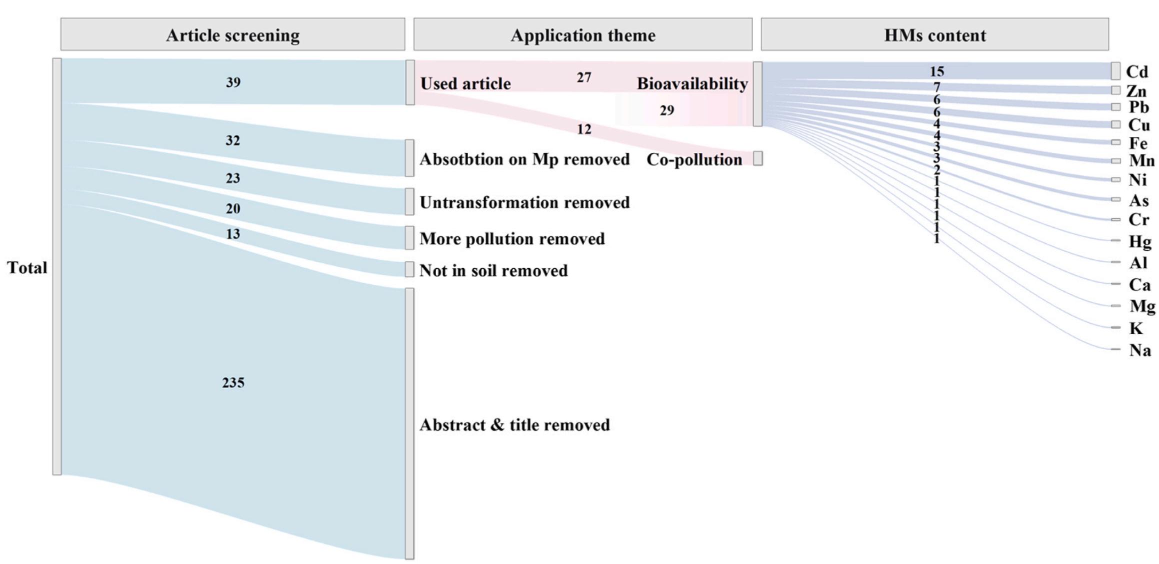

Fig. 1. Data collection and quality control framework in meta-analysis.

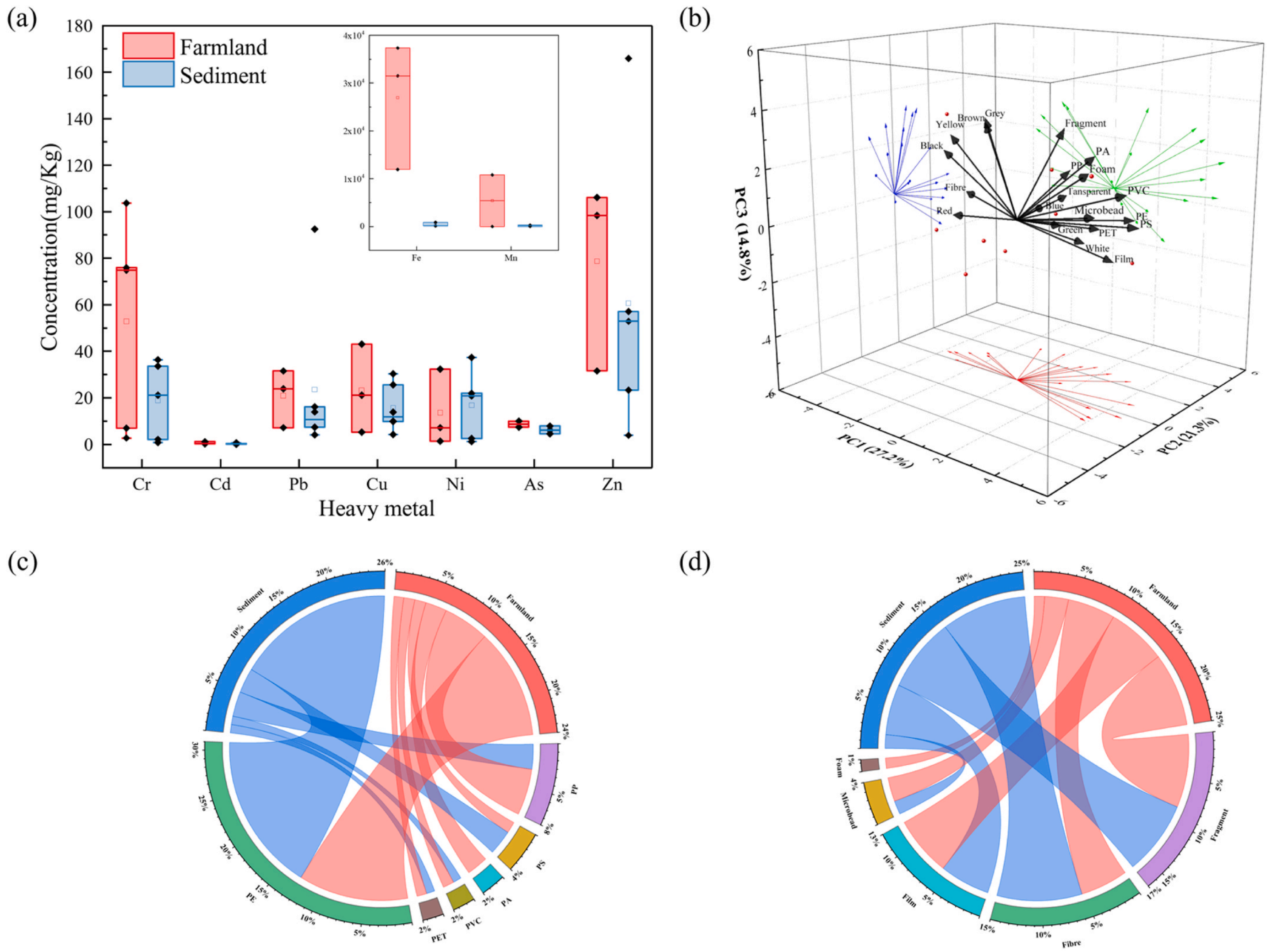

Fig. 2. Characteristics and source of HMs and MPs in combined pollution soils. (a): Concentrations of HMs in soils. (b): PCA analysis of MPs characteristics on current research. (c-d): polymer type and shape characteristic of MPs.

Microplastics were also detected in soils containing HMs. The abundance of MPs in the soil of China, Egypt, Iran, Germany, Tunisia, Pakistan, and India ranged from 8 to 220,000 items/kg (Fig.S3a), suggesting that MPs and HMs combined pollution levels differed throughout the world. The dominant particle size of MPs was smaller than 500 μm (Fig.S3b), which brought more ecological risks [41]. As shown in Fig. 2c, PE had the highest amount of MPs in farmland and sediment, accounting for 43.44% and 67.90%, respectively. PA was a particular MP in agriculture, accounting for 9.23%, which may be explained by the fact that it enters the soil during semi-automated agricultural production [42]. In both farmland and sediment, fragments larger than 40% was the most predominant MP shape (Fig. 2d). Sediment has more fibers, whereas farmland has more films, microbeads, and foams. After mechanical wear and natural weathering, fragments were the primary shape of plastic bottles, bags, and other packaging materials; hence, they were frequently found in natural environments [43]. Since films were the primary shape of plastic film disintegration, it was common in agricultural soil [44,9,45]. Microbeads and foams were more likely to be found in terrestrial environments since they were mainly used in cosmetic packaging and everyday decoration [46]. The majority of the fibers were discovered in domestic and industrial wastewater, and a significant amount of fibers were also produced by the fishing nets employed in fisheries, which led to its relatively high concentration in sediment [47]. Transparent was the most prevalent color in both farmland and sediment (Fig.S3c). The majority of disposable plastic items were transparent, which made them easily accessible in a variety of environments [48].

A principal component analysis was performed based on the polymer type, color, and shape characteristics of MPs in the environment to further evaluate the main sources of them (Fig. 2b), which detailed results were shown in Table S3. There was no statistically significant difference between the MPs characteristics of farmland and sediment, according to the 63.3% interpretation of the results. Thus, the grouping discussion was not conducted. The variance contribution rate of the first principal component was 27.2%, with PVC (0.36), PS (0.35), and foam (0.30) having the largest loads. The most common foam plastics were PVC and PS, which had strong buffering, low water absorption, good moisture, mildew, and heat insulation qualities and were mostly utilized as materials for packing and transportation [49]. As a result, it was assumed that the first principal component had to do with how a product was transported and packaged. The variance contribution rate of the second principal component was 21.3%, and the highest loading rates were transparent (0.39), blue (0.38), PE (0.36), and film (0.33). PE film has the ability to improve soil temperature, retain moisture, encourage plant development, and boost yield [50]. In addition to being transparent, color film has gained popularity in recent years, with blue film being the most popular to control crop growth [51-53]. Therefore, it was determined that the second principal component was linked to MPs created by the breaking and degrading of a lot of agricultural films. The variance contribution rate of the third principal component was 14.8%, with fragment (0.41) and grey (0.39) having the largest loads. Fragment was one of the common shapes of MPs, but gray was less commonly seen, and it may mainly come from storage and transportation processes [54]. Therefore, the third major component is considered to be related to warehousing and transportation.

3.2. *Effects of MPs on HMs bioavailability*

3.2.1. *Changes in bioavailability of HMs*

The RR value can more accurately depict the difference between them since it is the ratio between the experimental group and the control group. When RR> 1, it means that MPs made HMs more bioavailable, R= 1 means there was no impact, and RR< 1 means the bioavailability was decreased. Studies on how MPs affect the identical HMs bioavailability produced a variety of results (Fig. 3a). Studies have demonstrated effects that were stimulating (above the reference line), inhibitive (below the reference line), or exhibited no effect (on the reference line). At the same time, different HMs also had different changes in bioavailability. For example, most investigations showed that MPs enhanced the bioavailability of Fe, Mn, and As more significantly than Cu, Pb, Cd, and Zn, because there are more data with RR> 1.3 (Fig. 3a). Meta-analysis was employed to further investigate the impact of MPs on the various HMs bioavailability in order to quantify and interpret the inconsistent results in numerous investigations.

After being categorized by HMs, the findings of the meta-analysis were displayed in Fig. 3b. Microplastics have no discernible impact on Zn and As, but they can improve the bioavailability of Cu, Pb, Cd, Fe, and Mn, particularly Fe and Mn. Fe and Mn are sensitive soil elements that react quickly to changes in the soil environment [55]. Additionally, due to the abundance of high affinity reaction sites and vast surface area of their oxides, they can directly influence the effective forms of other HMs by forming complexes and adsorbing on the surface [56,57]. Zinc was a common HM found in polluted soils [58]. The capacity of soil and MPs to adsorb HMs will affect their bioavailability. If soil and MPs have strong adsorption capacity for HMs, it is not conducive to the increase of their bioavailability. Hodson et al. [59] had found that Zn as more challenging to desorb from MPs. However, studies on Zn adsorption in soil were contradictory, which may be due to the competitive adsorption of HMs such as Cu and Pb with similar chemical properties [60]. Research by Jalali and Moharrami [60] found that competition between HMs influenced their adsorption in soils, regardless of changes in pH and solid-liquid ratios, with Zn having higher adsorption than Cu and Pb. However, other studies have shown that the relative mobility of HMs in soils follows the trend of Zn>Pb>Cu [61]. These conflicting results indicated that the competitive adsorption process between HMs in soils was very complex and frequently subject to other factors. The uncertainty of Zn adsorption in soil will lead to the uncertainty of its bioavailability change, which may be the reason why the change of Zn bioavailability was not obvious. Furthermore, it is important to note that Zn was often measured as an accompanying metal in research. Currently, there is no specific study on the effect of MPs on Zn bioavailability, and further exploration of its mechanism is necessary. Arsenic is a kind of polyvalent metal, and the amount of Fe-Mn oxides in soil is frequently closely related to the form of it [62]. Theoretically, MPs should have an impact on the bioavailability of As, but the results of the meta-analysis revealed no significant effect. Firstly, this might be because there were currently only three studies on the effect of MPs on the bioavailability of As, which made the results of the analysis insufficiently representative. Secondly, arsenic existed in soils in various forms as a variable valence metal [63]. The mechanism of MPs impacted on As bioavailability may be more complex than that of other metals, and it was difficult to obtain a consistent trend of change, which might also be the reason why the results were not statistically significant. As a result, since the effect of MPs on the bioavailability of Zn and As was not significant, and the reasons ware still debated, it would not be discussed in detail in the following study. These results showed that MPs have different effects on the bioavailability of different HMs in soil.

3.2.2. *Major influencing factors of HMs bioavailability*

The polymer type, concentration, size, and shape of MPs all affect soil physicochemical indexes, such as pH and OM. The chemical form of HMs was frequently impacted by changes in soil physicochemical indexes, which altered their bioavailability [1]. Furthermore, exposure time and soil texture may also be important [34]. According to Fig.S4, the HMs bioavailability in the presence of MPs in soils can be affected by all of the aforementioned factors, and there was no publication deviation in the articles included by each influencing factor (Table S2), suggesting that the analysis results had a certain validity. Therefore, investigating how these factors affect the HMs bioavailability is crucial. As shown in Fig. 4a, the HMs bioavailability increased with prolonged exposure time, indicating a positive correlation between exposure time and RR. Since the RR with pH and OM were negatively correlated, alkaline conditions and a high OM content would inhibit the HMs bioavailability. However, the HMs bioavailability was not significantly impacted by the concentration or size of MPs. This could be because the data about the concentration and size of MPs was primarily focused on small areas. As a result, further study and categorization of the influencing factors were required (Fig. 4b).

The results showed that PE, PET, PP, PS, PU, PLA, and PES might improve the HMs bioavailability, whereas PA, PBS, PHB and PES had no impact. Feng et al. [64] also discovered a connection between the polymer type in MPs and the HMs bioavailability. Among them, the data of PBS, PHB and PES were all from only one article, so it would not be discussed. Heavy metals are often less able to be absorbed by MPs than by soil, leading to a "dilution effect" that eventually increases their mobility and availability [65]. Because they all include chemical bonds

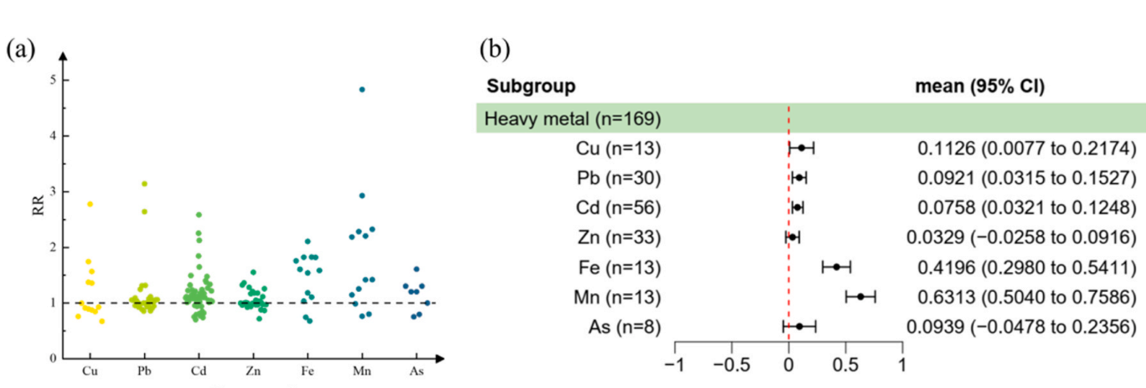

Fig. 3. Scatter plot (a) and forest plot (b) of HMs bioavailability in soil. The points and error lines in the forest plot represent the mean and 95% confidence intervals, respectively. When the 95% confidence interval contains 0, MPs is considered to have no significant effect on HMs bioavailability.

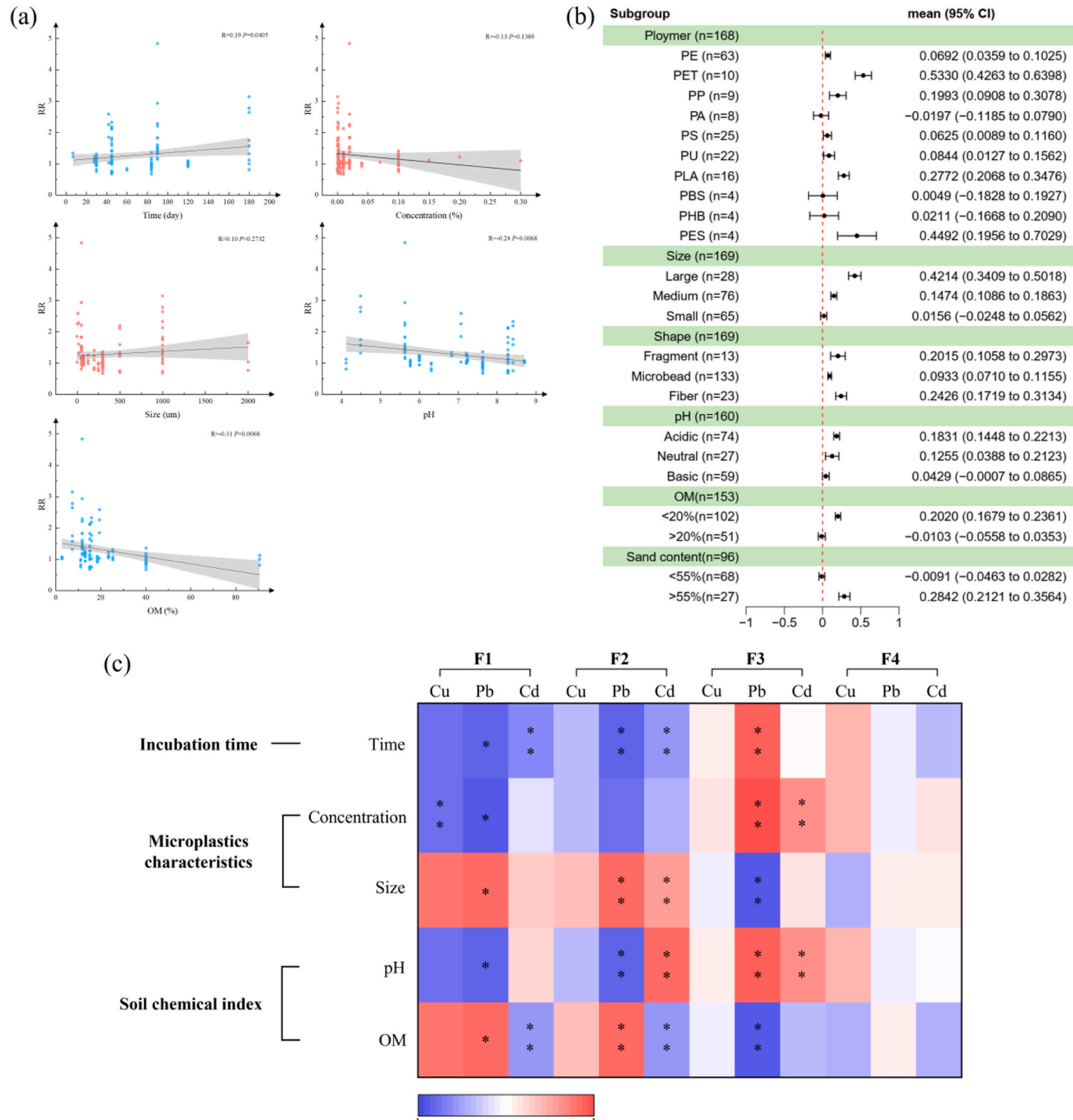

Fig. 4. Relationship between RR and effecting factors for the HMs bioavailability in soil. (a): Scatter plot of the relationship between RR and various effecting factors. (b): grouped forest plot. (c): Heat map of correlation coefficient between chemical forms of HMs and influencing factors. The blue dot indicates that the fit line is statistically significant, while the red is not. OM, organic matter. PE, polyethylene. PET, polyethylene terephthalate. PP, polypropylene. PA, polyamide. PS, polystyrene. PU, polyurethane. PLA, Polylactic acid. PBS, Polybutadiene styrene. PHB, polyhydroxybutyrate. PES, polyether sulfone. F1, acid soluble fraction of HMs. F2, reducible fraction of HMs. F3, oxidizable fraction of HMs. F4, residual fraction of HMs.

(-NH-C═O-/-COO-), PA, PU, PLA, and PET may all be hydrolyzed by biological enzymes [66]. The result of categorization indicated that hydrolysable MPs have a greater potential to increase the HMs bioavailability than non-hydrolysable (Fig.S5), such as PE, PP and PS. Interestingly, despite the fact that -NH-C═O- is more difficult to hydrolyze than the -COO-, PA has no significant impact on HMs bioavailability. PA had an amide bond, which allowed it to provide N, an element that was helpful for microorganism reproduction, in addition to C, H, and O for the growth of microorganisms [64,67]. This is conducive to the degradation of PA into low molecular weight OM and increases the amount of OM in soil [64,68]. High OM had no impact on the HMs bioavailability, which may be the reason PA is unable to alter the HMs bioavailability. In contrast to PET and PLA without N element, PU also contains N element, which has a lesser impact on the HMs bioavailability. This suggested that the HMs bioavailability was not significantly affected by hydrolysable MPs which containing N. This might be because microorganisms prefer N-containing materials for reproduction, reducing the influence of MPs. However, further study is required to verify this hypothesis. Additionally, it was intended to categorize MPs based on their biodegradability. However, PLA was the only biodegradable MP with analytical value, so the categorization was not compared. Future studies on the effects of biodegradable MPs on HMs bioavailability should be strengthened in order to further compare the effects of biodegradable and non-biodegradable MPs. Small MPs (≤10 μm) exhibited no discernible impact on the HMs bioavailability, but as MPs get larger, the boosting effect became stronger. MPs with small sizes had more sites to absorb HMs and a bigger specific surface area than MPs with large sizes [69]. Similar findings were reached by Zhao et al. [57], who demonstrated that smaller MPs had a lower concentration of available Cd in soil than larger ones. Relatively speaking, there was no difference in how the shape of MPs affected the HMs bioavailability. Their bioavailability was improved by fragment, microbead, and fiber. The HMs bioavailability was enhanced in neutral (6.5–7.5) and acidic (≤6.5) soils, but not in alkaline (>7.5) soils. High pH caused soil particles and MPs to have more charged sites on their surfaces, which effectively promoted the adsorption of HMs [70]. As pH steadily decreased, hydrogen ions will rise and compete with HM ions for adsorption sites, resulting in the reduction of HMs adsorption on soil particles and MPs [71]. Furthermore, Fe and Mn were present in soils as amorphous and microcrystalline oxides, and their surfaces had a large number of adsorption sites, which can be combined with other metal ions [72,56,73]. when the pH decreased, Fe and Mn (hydroxide) oxides dissolved, releasing more HMs ions and enhancing the HMs bioavailability [74]. Therefore, changes in their morphological structure often affect the bioavailability of other metals. Therefore, the HMs bioavailability increased under acidic conditions. At conditions of high OM content (>20%), the HMs bioavailability was unaffected, while conditions of low OM content (≤20%) demonstrated a promoting impact. Organic matter in soils is a supramolecular mixture composed of various biological residues, biological secretions, and humus, which has abundant surface functional groups [75]. The carboxyl groups on the OM surface often formed complexes with HMs ions [76], increasing the quantity of the organic composite form of HMs while decreasing the amount of the insoluble chemical form and reducing HMs bioavailability [77]. According to the results of the soil sand content grouping, high sand content (>55%) had a promoting impact, whereas low sand content (≤55%) had no discernible influence on the HMs bioavailability. Similar results were reached by Yu et al. [22], who discovered that HMs responded more strongly in coarse particle components, which may differ from the distribution ratio of MPs in various aggregates. In conclusion, the HMs bioavailability may be increased by MPs with large size and soils with acidic, low OM, and high sandy content.

The bioavailability of soil HMs often depends on their distribution and soil properties. The chemical speciation of HMs helps predict whether they will be bioavailable as free ions, organic complexes, or mineral binding components [78]. Based on the BCR method recommended by the European Community Standards Office, soil HMs were separated into four fractions: acid-soluble (F1), reducible (F2), oxidizable (F3), and residual (F4) [79]. While the residual fraction (F4) was inert to the environment, acid-soluble (F1), reducible (F2), and oxidizable fraction (F3) had environmental characteristics of bioavailability [80]. The contents of their four chemical forms, which had an impact on the HMs bioavailability in soil, were constantly subject to variations with soil characteristics [81]. Therefore, the relationship between various HMs chemical speciation and influencing factors has been further explored (Fig. 4c). Iron and manganese oxides are often the key factors affecting the bioavailability of other HMs, which often lead to changes in the reducible fraction of HMs [82]. However, only the bioavailability of Fe and Mn was determined in the studies collected, not their chemical speciation. Due to the lack of data, the chemical speciation of Fe and Mn were not analyzed. The results revealed that the oxidizable fraction (F3) of HMs was mostly positively connected with exposure time, MPs concentration, and soil pH, whereas the acid-soluble (F1) and reducible (F2) fractions of HMs were primarily positively correlated with MPs size and soil OM. On the other hand, the residual fraction (F4) did not alter appreciably. The addition of MPs increased soil pH and was exacerbated over time by reducing soil respiration [83]. Xu et al. [84] have found that the oxidizable fraction of Cu, Pb and Cd increased with the increasing of pH, which may be because high pH could induce the formation of metal hydroxide. This phenomenon was likely to intensify with increasing MPs concentrations. At the same time, the addition of MPs also increases soil aeration and porosity, which increasing the redox potential and OM [85]. This induces the oxidation of sulfides in the organic form of HMs, which releases large amounts of HMs that subsequently exist in acid-soluble and reducible fractions [86]. The increase of MPs size would aggravate a series of problems caused by soil aeration and porosity increase [85]. It can be seen that the MPs characteristics can indirectly change the HMs bioavailability by affecting the soil physicochemical indexes. It indicated that there was interaction between MPs characteristics and soil physicochemical indexes, which need to be further explored. In addition, soil microorganisms also play a crucial role. It was important to note that, contrary to the result of the categorized forest plot, the acid soluble (F1) and reducible fraction (F2) of Cu and Pb were positively linked with soil OM content. According to research by Korshin et al. [87], the accumulation of charge on their surface might be the mechanism that caused the increase in OM and speeded up HMs release.

In conclusion, the susceptibility of different HMs to influencing factors varies. Additionally, because these influencing factors frequently coexist and interact with one another in the natural environment, it is important to investigate how they interact in order to more accurately assess and forecast how combined pollution will affect the HMs bioavailability.

3.2.3. The interaction among influencing factors

The Geodetector model was used to investigate the impacts of influencing factors on HMs bioavailability in combined polluted soils and their interactions (Fig. 5). The HMs bioavailability was significantly influenced by concentration (0.53), pH (0.52), and OM (0.45), as shown in Fig. 5a, but not by shape (0.01), polymer type (0.07), or sandy content (0.05). It was important to note that different HMs (0.15) responded to the influencing factors in different ways, and that the pH (0.79), OM (0.79), concentrations (0.79), and sizes (0.81) in the soil all exhibited significant interactions with the HMs. As a result, the effects of influencing factors on different HMs were individually examined (Fig. 5b–f).

The main factors impacting all HMs were still concentration, pH, and OM. When compared to other HMs, the size and concentration had negligible impacts on Mn and Cd bioavailability, respectively. The bioavailability of Pb and Cd was greatly influenced by exposure time, whereas Fe and Mn were greatly influenced by polymer type, and Pb and Cu were greatly influenced by the sand content. The interaction detection can divide the interaction between factors into none-weaken, Univariable weaken, Bi-variable enhance, Independent and Nonlinear-enhance (Table S4). In order to further investigate the interaction between MPs characteristics and soil physicochemical indexes, interactive detectors of Geodetector model were used. As shown in Fig. 5a, there was a nonlinear enhancement relationship between HMs and other factors (Table S5), which suggested that different HMs may affect each other. Therefore, the interaction between the MPs characteristics and soil physicochemical indexes was subsequently investigated by classifying them according to HMs (Fig. 5b-f). The results revealed that the majority of the effecting factors showed bi-variable enhancement or nonlinear enhancement on the HMs bioavailability (Table S5). This indicated that there was a clear interaction between the MPs characteristics (concentration, polymer type, size) and the soil physicochemical indexes (pH, OM, sand content) on HMs bioavailability.

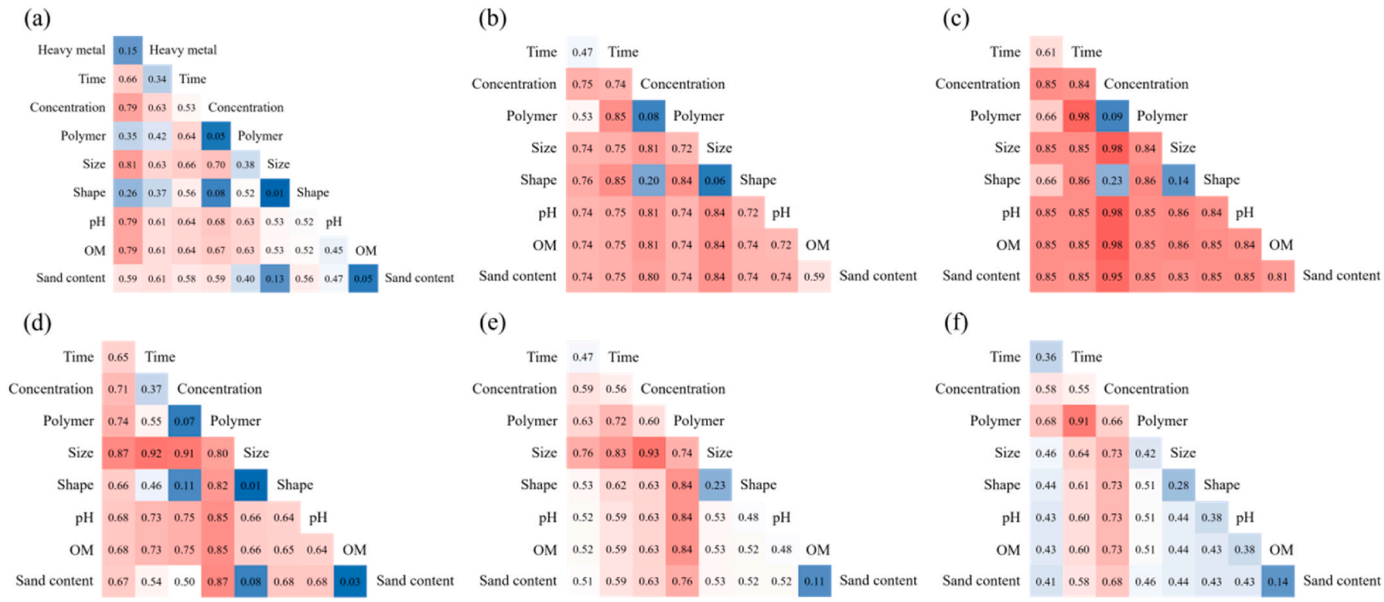

Fig. 5. The effecting factors of Total HMs (a), Cu (b), Pb (c), Cd (d), Fe (e) and Mn (f) bioavailability of in combined pollution soils.

Microplastics indirectly affected the HMs bioavailability by changing soil physicochemical indexes, which might be the reason for the interaction between them. There were also Bi-variable and nonlinear enhancement relationships between soil physicochemical indexes, in which the interaction between pH and OM was the strongest. This may be due to the fact that pH could indirectly affect OM by altering soil enzyme activity [88]. Furthermore, the sensitivity of different HMs to the change of soil physicochemical indexes was different, so the interaction between the MPs characteristics and soil physicochemical indexes on the HMs bioavailability was also different. Among them, the interaction between MPs polymer type and soil physicochemical indexes was the strongest, which may be related to the different functional groups on the surface of each polymer type. Compared with other HMs, the number of nonlinear enhance relationships among the influencing factors of Fe and Mn was less, indicating that the interaction of the influencing factors on the bioavailability of Fe and Mn was relatively weak. Numerous researchers had shown that the presence of MPs could influence the soil physicochemical indexes and had a certain correlation with MPs characteristics [89,80,31,22], but the mechanism was complicated and required further studies. Furthermore, various other pollutants (pesticides, antibiotics, persistent organic pollutant, etc.) as well as plants and animals in the soil can all have an impact on soil physicochemical indexes. Because this paper only includes articles about MPs and HMs, the subject of multi-pollutant combined pollution will not be discussed further. However, it is worth noting that combined pollution of numerous pollutants exists and should be attention, particularly in agricultural soil. Plants and animals in soil can also influence soil physicochemical indexes through growth and metabolism, which may be influenced by plant type [72,73]. Therefore, the effect of MPs on the HMs bioavailability in soils containing plants or animals may also be affected. Due to data limitations, only a subgroup analysis has been performed on the bioavailability of Cd with or without plants/animals. The results showed that the effect of MPs on the bioavailability of Cd was not significantly altered by the presence or nonexistence of plants/animals (Fig. S6). However, studies have shown that MPs can affect soil-plant systems by altering soil microbial communities, thereby promoting the bioavailability of Cd [32]. These conflicting results are worth further investigation. In conclusion, MPs can enhance the bioavailability of Cu, Pb, Cd, Fe and Mn, which has certain environmental risks. In complex natural environments, the risks may rise accordingly.

3.3. *Environmental impacts and limitations*

A comprehensive analysis of the prior studies on the impact of MPs on HM bioavailability in soil was conducted in this research using meta-analysis. The findings shown that the bioavailability of Cu, Pb, Cd, Fe, and Mn could be improved by PE, PP, PS, PET, PU, and PLA. This suggested that the presence of MPs enhanced the bioaccessibility of HMs, increasing their mobility and hazards, as well as potential risks to the environment and ecology. The main influencing factors were MPs concentration, soil pH and OM content. The impact of MPs on the HMs bioavailability was strengthened when MP size, soil sand content, and exposure time increased. On the bioavailability of HMs, the effect of hydrolysable MPs containing N element was only marginally significant. It provided a certain reference value for the development of MPs pollution management measures and remediation technologies. A significant interaction between MP characteristics and soil physicochemical indexes on the impacts of HMs bioavailability was also discovered by the study, but the mechanism was still unclear. This provides a new perspective for the investigation of how MPs affect the bioavailability of HMs in the future.

However, it was discovered through the data collecting and statistical processes that the existing investigation into the combined pollution of MPs and HMs was insufficient and mostly concentrated in China and a small number of other countries. The majority of the land polymer types were found in agricultural soils. Moreover, there were few investigations on the other HMs, and the influence of MPs on the HMs bioavailability was also mostly focused on the Cd. The MPs polymer types used were also mostly non-biodegradable, and there were few studies on biodegradable MPs, making it impossible to predict which MPs polymer types will have a greater impact on HMs bioavailability. At the same time, it was not able to investigate how different MPs effected the HMs bioavailability based on a single HM. Furthermore, the results showed that soil microorganisms were directly or indirectly involved in the process of MPs affecting the bioavailability of soil HMs. This made it play an important role in the interaction influence of MPs characteristics and soil physicochemical indexes on the HMs bioavailability. Unfortunately, microbial community indicators (such as Shannon, Chao1, Simpson and ACE indexes) were not fully available at the time of data collection. In conclusion, it was required to strengthen the study on the effects of MPs on the HMs bioavailability in soils in order to more precisely quantify the impact of MPs and HMs combined pollution on ecological risks.

4. Conclusion and future prospects

The combined pollution phenomenon of HMs and MPs was widespread, which increased the HMs bioavailability and also had an impact on their chemical speciation. The main influencing factors were MPs concentration, soil pH, and OM, and there was a clear interaction between MPs characteristics and soil physicochemical indexes on the HMs bioavailability. MPs size, soil sand content, and exposure time all had positive correlations with HMs bioavailability, while pH and OM had a negative correlation with them. All of these suggested that there was a risk to the environment from the combined pollution of MPs and HMs in soils through increasing the HMs bioavailability. To the best of our knowledge, this is the first time a meta-analysis about the impact of combined pollution of MPs and HMs on HMs bioavailability has been conducted. In the background of ongoing global soil combined pollution of MPs and HMs, our findings will help to develop pollutant discharge and land management systems. They will also serve as a theoretical foundation and a basis for reference to rational control of MPs and HMs combined pollution and help prevent environmental ecological and public health problems. However, in the meta-analysis process, we believe the following aspects require further research:

1. Although combined pollution has been shown to improve the bioavailability of heavy metals, the types of HMs, MPs, and land use types involved were relatively simple. Therefore, it is necessary to further expand the scope of research on the above factors.
2. Research on the impact of HMs bioavailability now focuses mostly on one particular factor. For example, targeting only certain MPs characteristics or soil physicochemical indexes of soil. There are few studies on the interaction of their effects on the HMs bioavailability. However, this interaction might raise the ecological risk. Therefore, it is urgent to conduct multi-factor interaction research. In addition, the role of soil microorganisms in this process was also worth further investigation.
3. Based on the increased HMs bioavailability and the nature of bioaccumulation and biomagnification of HMs, it can enhance toxicity through the food chain and its range of influence. Therefore, with reference to the concept of life cycle assessment, it is very necessary to conduct research on the impact intensity of MPs pollution at different periods on the HMs bioavailability and the health risk assessment.

Environmental Implications

The toxicity of heavy metals to terrestrial organisms is intimately correlated with their bioavailability. However, research results on the effects of microplastics on heavy metal bioavailability seemed contradictory. This study employed meta-analysis to determine the heavy metals whose bioavailability was increased by microplastics, identify the primary influencing factors that led to the increased in bioavailability, and assess whether there was interaction among each factor. This will help to evaluate the potential environmental risks caused by combined pollution of microplastics and heavy metals in soils and has certain reference value for the development of MPs pollution management measures and remediation technologies.

CRediT authorship contribution statement

Qiuying An: Data curation, Formal analysis, Visualization, Writing – original draft. **Tong Zhou:** Software. **Ce Wen:** Visualization. **Changzhou Yan:** Conceptualization, Writing – review & editing, Supervision, Funding acquisition.

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.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.132369.

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