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# Macro- and micro-plastic accumulation in soils under different intensive farming systems: A case study in Quzhou county, the North China Plain<sup>☆</sup>

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## ABSTRACT

The macroplastics (MaPs) and microplastics (MiPs) polluting agricultural soils raise great concerns. Unfortunately, scientists know little about the occurrence of MaPs/MiPs in soil among different farming systems. In this study, we analyzed MaPs/MiPs in soils (0–30 cm) collected from six different farming systems (wheat-maize rotations, cotton, vegetables, permanent orchards, greenhouses with and without mulching) in Quzhou county, the North China Plain, by using fluorescence microscope and micro-Fourier transform infrared spectroscopy. The results showed that the abundance of MaPs and MiPs ranged from 0.2 to 46.8 kg ha<sup>-1</sup>, and 4.1 × 10<sup>3</sup>–3.7 × 10<sup>4</sup> items kg<sup>-1</sup>, respectively. The prominent colors of the MaPs were white and black. The predominant shape, size and chemical composition of soil MiPs were fragments (45–62%), <1 mm (98–99%), and polyethylene (38–43%), respectively. MaPs were mainly detected in the 0–10 cm soil layer. MiP abundance in the 0–10 cm soil layer was significantly higher than that in the 20–30 cm soil layers among different farming systems, except for the fields with wheat-maize rotations and permanent orchards ( $p < 0.05$ ). Overall, cotton fields showed the highest MaP and MiP abundance, followed by vegetable fields and orchards. Redundancy analysis revealed that tillage practices and plastic film management greatly influence the size distribution of MiPs. A strong negative correlation between large-sized plastic fractions (0.2–1 mm) and tillage frequency was tested while the years of application of plastic films and the abundance of plastic residues showed a strong positive correlation with small-sized plastic fractions (<0.2 mm). Our findings conclude that agricultural mulch films are an important source of MaPs and MiPs in agricultural soil and distributions are strongly influenced by agricultural management practices and farming systems. Further studies should take farming systems and farming practices into account, thereby exploring the potential mechanisms of plastic fragmentation and granularization in agricultural soil.

## 1. Introduction

Agricultural plastic mulch film has been used intensively worldwide for its many benefits such as improving crop yield, conserving soil moisture, increasing soil temperature and controlling weeds (Anikwe et al., 2007; Du et al., 2016; Li et al., 2013). As a country with one-fifth of the global population but only 8% of the arable land in the world, China has applied plastic film since 1978 (Zhou et al., 2009) and become

the world's largest user of agricultural plastic film (Daryanto et al., 2017). Unfortunately, large amounts of macro-plastics (>5 mm, MaPs) have been left to accumulated in soils due to improper management and recycling practices (Zhang et al., 2020a). In China, accumulated concentrations in soil range from 0.2 to 421.6 kg ha<sup>-1</sup> (Ren et al., 2023). MaPs undergo fragmentation as a result of biodegradation, ultraviolet radiation, and mechanical weathering processes (Lambert and Wagner, 2016), which continuously generate smaller plastic particles such as

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microplastics (MiPs, <5 mm) and nanoplastics (<1  $\mu\text{m}$ ) (Gigault et al., 2018; Moore, 2008). MiP abundance in Chinese farmlands has been reported to range from 1.6 to  $6.2 \times 10^5$  items  $\text{kg}^{-1}$  of dry soil, with an average abundance of  $4.5 \times 10^3$  items  $\text{kg}^{-1}$  (Ren et al., 2023). Recent research reported that the accumulation of MiPs and nanoplastics in soil affects water-stable soil aggregates, pH, soil organic carbon, nitrogen and microbial communities (de Souza Machado et al., 2019; Meng et al., 2022; Yang et al., 2021b). These accumulated compounds have also been associated with heavy metals and organic pollutants (Cao et al., 2021; Fu et al., 2021), resulting in complex pollution risks in the soil environment (Browne et al., 2013; Pérez-Reverón et al., 2023). MiPs could also affect the growth and reproduction of plants, soil mesofauna and invertebrates (Banerjee and Shelver, 2021; Zhou et al., 2020), or even be transferred from prey to predator along the food chain (Gao et al., 2020), eventually endangering human health (Yan et al., 2022). Therefore, MiP pollution has been ranked as one of the top ten environmental issues by the United Nations Environment Programme (UNEP, 2014).

Farming systems play a pivotal role in influencing the occurrence and characteristics of MiPs and MaPs (Liu et al., 2023a). Agricultural management practices used in different farming systems, such as irrigation, compost application and crop protection management, can affect the input of MiPs and MaPs, as well as the fragmentation processes of plastics in farmlands (Guo et al., 2023; Pérez-Reverón et al., 2022). Irrigation is a major source of soil MiPs, detected in different irrigation sources worldwide (Chia et al., 2021; Kumar et al., 2021) including wastewater, groundwater and surface water. Zhang et al. (2016) implied that some irrigation equipment, such as that used in the drip irrigation system, is used under mulch film. This might impede the collection of plastic residues, resulting in an excessive accumulation of soil MaPs. Fertilizer applications significantly increase the abundance of MiPs in farmlands (Zhang et al., 2020b). Additionally, they strengthen roots and stems, which in turn hinders the collection of plastic residues after harvest, leading to an increased accumulation of MaPs in agricultural fields (Li et al., 2022). The use of agricultural plastic products, such as mulch films and greenhouse films, result in a continuous generation of MiPs due to the fragmentation processes of plastic residues (Li et al., 2022). Yang et al. (2022) reported that MiPs could be released from mulch film under UV exposure, and the abundance of plastic film-derived MiPs increased with prolonged aging time. Huang et al. (2020) indicated that agricultural plastic mulching is a major source of MaPs, and the concentrations of MaPs and MiPs increase with increasing mulching years. Atmospheric deposition could also contribute to the MiP accumulation in farmlands (Yang et al., 2021a). Greenhouses, as a form of enclosed environment, could better resist the accumulation of MiPs from the air than open-field farmlands (Wang et al., 2022). When MaPs and MiPs enter farmlands, tilling can also contribute to fragmentation processes by physically creating smaller plastic particles which mix with soil, leading to a constant generation of MiPs (Song et al., 2017; Sun et al., 2024). Although numerous studies have explored the occurrence of MiPs and MaPs in agricultural fields (Xu et al., 2020a) and only few studies focusing on the MiP occurrence in different farming systems (Kim et al., 2021; Li et al., 2023b), the relationship between farming systems, agricultural management practices and characteristics of MiPs and MaPs have not been well investigated yet.

In this study, we selected Quzhou county, a typical agricultural county in the North China Plain (NCP) with various farming systems, as a case study to investigate the occurrence and characteristics of MiPs and MaPs in six farming systems (wheat-maize rotations, cotton, vegetables, permanent orchards, and greenhouses with and without mulching). The objectives of this study were to: a) compare the abundance of MiPs and MaPs among six farming systems; b) analyze the distribution characteristics of MiPs in terms of size, shape and polymer type; and c) explore the distribution pattern of MiPs across different farming systems and soil depths. The findings can extend our current understanding of plastic fragmentation in soil under various farming systems and

agricultural management practices.

## 2. Materials and methods

### 2.1. Study area

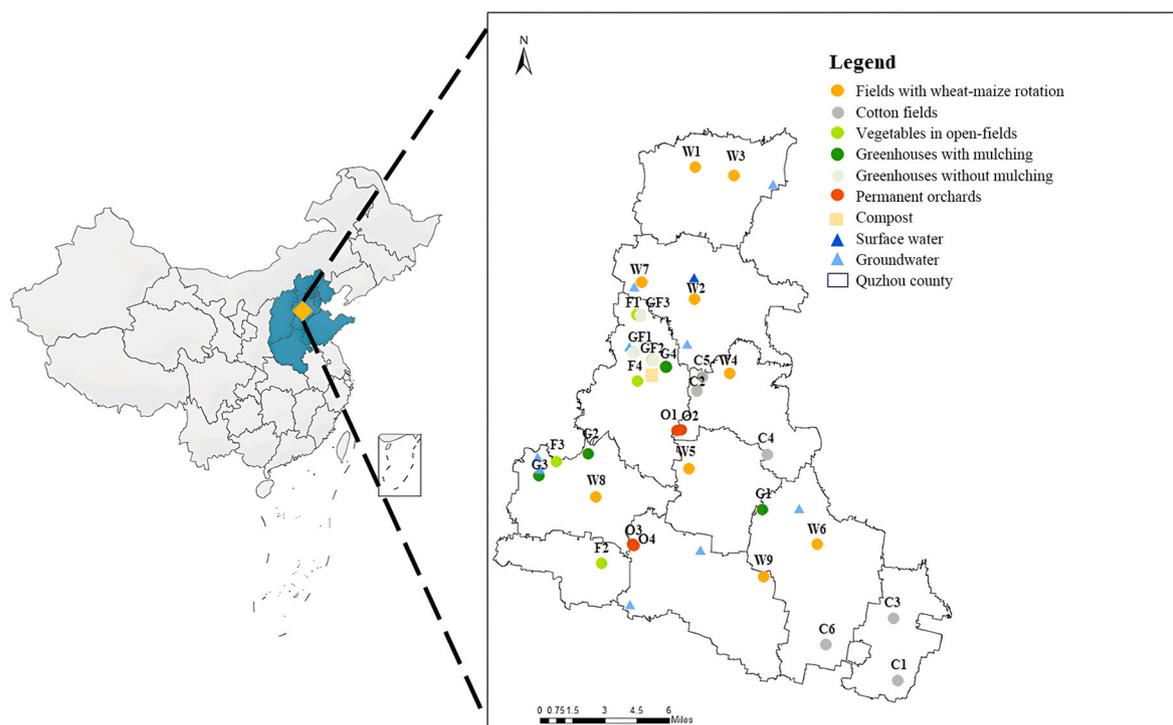
Quzhou County (114°50'22.3"–115°13'27.4"E, 36°35'43"–36°57'N) is situated in the northeast of Handan, Hebei, China (Fig. 1). The area experiences a humid subtropical monsoon climate, characterized by an annual mean temperature of 13.4 °C and an average annual precipitation of 556.2 mm (Mu et al., 2022). The dominant crops include wheat-maize rotations, cotton, and vegetables in open-fields and greenhouses, as well as apples and grapes. Surface water and groundwater serve as the primary sources of irrigation, and chicken manure is extensively utilized in fields for fertilization in vegetable production, greenhouses, and orchards. A total of 30 sampling sites were selected to represent local farming systems, including wheat-maize rotations (n = 9), cotton (n = 6), vegetables in open-fields (n = 4), greenhouses without plastic mulching (n = 4), greenhouses with plastic mulching (n = 3), and permanent orchards (n = 4) (Fig. 1). Mulch film was only applied once per year in cotton fields, vegetable fields, and greenhouses with plastic mulching. Transparent plastic mulch film was applied exclusively to cotton fields. In contrast, black plastic mulch film was widely used in open-fields and greenhouses using plastic mulching. Light blue greenhouse film was used in greenhouse farming systems. The diverse sources of plastic products used in the selected farming systems are illustrated in Fig. S1. Further detailed information regarding the utilization and disposal of agricultural plastics, irrigation sources, and compost usage is provided in Table S1.

### 2.2. Samplings

All samples were collected from April to May 2022. For soil samples, three sampling subplots (30 cm  $\times$  30 cm) were selected randomly at each sampling plot and soil samples were collected at three soil layers: 0–10, 10–20 and 20–30 cm. Approximately 0.5 kg of soil sample was collected from each subplot in each soil layer using a stainless steel shovel and stored in paper bags, and there was a total of 9 soil samples from each sampling plot. MaP samples were manually collected from each sampling subplot. We collected chicken manure with three replicates to represent compost samples. Meanwhile, as local sources of irrigation, surface and ground water samples were also collected from nine irrigation plots. A total of 3 L of bulk surface water (0–20 cm in depth) was collected in triplicate (1 L per collection) at each sampling site using a stainless steel bucket. Similarly, 3 L of groundwater was collected in triplicate (1 L per collection) from the irrigation water well (115° 0' 39.45" E, 36° 51' 55.66" N) using a pump. All water samples were stored in stainless steel bottles for further analysis. In total, 270 soil samples, 46 soil MaP samples, 3 compost samples, 8 surface water samples (totaling 24 L), and one groundwater sample (totaling 3 L) were collected. All these samples were transported to the laboratory for further analysis.

### 2.3. Laboratory analysis

Soil properties, including pH and soil organic carbon at each sampling site, were measured according to the method laid out by Zhuang et al. (2020) (Fig. S2a and Fig. S2b). As soil particles actively participate in the wear of MiPs and MaPs, consequently reducing the size of plastic particles (Ouyang et al., 2023), we hypothesized that the relationship between the size distribution of soil particles and the size distribution of MiPs may vary across various farming systems. Therefore, we selected one farmland from each farming system to compare its soil particle size distribution and MP size distribution in the 0–30 cm soil layer. Soil particle size distribution was measured following the instructions of ISO 13320.



**Fig. 1.** The location of Quzhou county (represented by the yellow square) in the North China Plain (shaded in blue) and the sampling sites of soil in six farming systems, irrigation, and compost. W1-W9: fields with wheat-maize rotations, C1-C6: cotton fields, F1-F4: vegetable fields, G1-G4: greenhouses without mulching practices, GF1-GF3: greenhouses with mulching practices, and O1-O4: permanent orchards. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

### 2.3.1. MaPs analysis

MaPs were picked up during sampling. We found that the majority of MaPs (>90%) were found in the 0–10 cm soil layer, and very few pieces were in the 10–30 cm soil layers. As a result of this observation, the collected MaPs from each subplot were mixed as a single MaP sample for further analysis. After collection, we cleaned these samples in the lab following a modified method based on a prior report (Meng et al., 2020). The clods and plant roots were manually removed from samples. Samples were then combined with tap water in a glass beaker. Samples were gently stirred with a glass rod and left overnight. The treated samples were subsequently transferred to another glass beaker filled with tap water. This beaker was then placed in an ultrasonic cleaner for 1 h and left overnight. This step was repeated twice until the solution in the beaker became clear, and no visible attachments were present on the surface of the MaPs. The cleaned samples were placed in plastic mesh bags, air-dried for 2 days at room temperature and weighed.

### 2.3.2. MiP analysis

The extraction and identification of MiPs in soil was carried out using a Nile Red solution modified by Qi (2021), achieving a recovery rate ranging from 82% to 90%. Briefly, after air-drying at room temperature, soil samples and compost samples were passed through a 2 mm metal mesh, and the MiP particles larger than 2 mm were collected as MaP samples. 5.0 g of each sample was transferred to a pre-cleaned beaker and thoroughly mixed with 230 mL of saturated NaCl solution on a magnetic stirrer at 260 rpm for 30 min. After 24 h, each supernatant containing MiPs was transferred to another beaker, and 4 M of NaOH solution was added to a volume of 500 mL to digest organic matter. The solution was mixed thoroughly on a magnetic stirrer at 260 rpm for 30 min. After standing for another 24 h, the supernatant was transferred to another pre-cleaned beaker. Extraction of MiPs from water samples was adapted from a previously reported method (Wang et al., 2018). Briefly, the water sample was initially filtered through a 50  $\mu\text{m}$  stainless-steel sieve. The filtrate was further filtered using 5  $\mu\text{m}$  mixed cellulose ester

(MCE) membranes (Shanghai Xingya Co., Ltd., China) under a vacuum pump. The particles retained on both the sieve and the membranes were thoroughly rinsed with distilled water into beakers. This was then treated with 50 mL of 30%  $\text{H}_2\text{O}_2$  at room temperature and kept in the dark for 48 h. Following this, the supernatant was carefully transferred to another beaker.

The supernatants from water samples, soil samples and compost samples were brought to a volume of 200 mL each by adding distilled water. Subsequently, Nile red solution (0.1  $\text{mg mL}^{-1}$  in methanol) was added to the supernatant to achieve a dye concentration of 0.5  $\mu\text{g mL}^{-1}$ . The mixture was thoroughly stirred with a glass rod. After 30 min, the supernatant was filtered through 0.2  $\mu\text{m}$  MCE membranes (Tianjin Jinteng Co., Ltd., China) using a vacuum pump and then the MCE membranes were stored in petri dishes. All MCE membranes were detected with a fluorescence microscope (Olympus BX53, Japan) within 24 h, which has been applied for MiP identification (Grause et al., 2022; Qiu et al., 2015; Zheng et al., 2023). The potential MiPs, stained with Nile Red solution, appeared green under blue fluorescence. The method for selecting the field of view was carried out following a previously reported method (Li et al., 2023a). The size range of the MiPs was defined as 3.23  $\mu\text{m}$  to 5 mm due to the instrument limitation of detection. After that, Image J software was used to count and measure potential MiPs in the photos captured by the fluorescence microscope. The abundance unit of MiPs in soil and compost was expressed as items  $\text{kg}^{-1}$ , and in water samples it was items  $\text{L}^{-1}$ .

In addition, a total of 97 suspected MiPs particles were randomly selected for chemical identification, comprising 84 particles from soil samples (10% of all suspected MiPs), 7 particles from compost samples (88% of all suspected MiPs), 5 particles from surface water (12% of all suspected MiPs), and 1 particle from groundwater sample (100% of all suspected MiPs). The identification was conducted using micro-Fourier transform infrared spectroscopy ( $\mu\text{-FTIR}$ , Bruker LUMOS, Germany). The obtained spectra were compared with the standard spectral libraries of polymers to determine the chemical composition of MiPs.

### 2.3.3. Quality control

To prevent contamination, a cotton lab coat and nitrile gloves were worn during the sample collection and treatment. All equipment and containers were rinsed three times with Milli-Q water before experiments were conducted. To assess background contamination, three blank samples were prepared for both solid and liquid sample treatments as the background contamination without soil, compost, and water in each batch of samples. Blank samples of solid sample treatment and liquid sample treatment revealed the presence of  $4.9 \pm 1.6$  and  $5.0 \pm 0.4$  MiP particles per field of view under fluorescence microscope, respectively. All the results were analyzed after correcting for the blank samples.

### 2.4. Statistical analysis

The results were analyzed using SPSS statistics software (25, IBM, USA) and were presented as average values  $\pm$  SD. In terms of the particle shape of MiPs, we used circularity to classify particles into fibers (0.0–0.3), fragments (0.3–0.6), and particles (0.6–1.0) (Prata et al., 2019). Before statistical analysis, normality of MiP abundance across all sites, size distribution of MiPs and soil particles were verified using the Kolmogorov-Smirnov test. One-way ANOVA was conducted considering farming systems and soil layers as fixed factors. In the case of the significance of ANOVA, least significant difference (LSD) tests were carried out for multiple mean comparisons. If data did not distribute normally, a nonparametric Kruskal Wallis test followed by a Mann Whitney test were applied to compare individual treatments. The significance level used for all the tests was 0.05. MiP abundance, MaP abundance, pH values, and soil organic carbon were all presented as box plots, with the solid black line indicating the average value. Scientific notation is applied to values of 1000 and above to ensure clarity and consistency in presenting large numbers. To explore the relationship between the shape distribution of MiPs in different sampling sites, principal component analysis (PCA) was performed with MiP shape distribution and sampling sites, and a redundancy analysis (RDA) was performed with size distribution of MiPs, soil particle size distribution, and data of agricultural activities from sampling sites, which were conducted using CANOCO software (version. 5.02).

## 3. Results

### 3.1. MaP properties among different farming systems

MaPs were found in all farming systems across Quzhou county, with the concentrations ranging from 0.2 to  $46.8 \text{ kg ha}^{-1}$  with an average

value of  $20.1 \text{ kg ha}^{-1}$  (Fig. 2). MaP abundance in vegetable fields ( $46.8 \pm 43.3 \text{ kg ha}^{-1}$ ) and cotton fields ( $37.1 \pm 18.4 \text{ kg ha}^{-1}$ ) was significantly higher than in fields with wheat-maize rotations ( $1.9 \pm 1.5 \text{ kg ha}^{-1}$ ) and greenhouses without mulching ( $0.2 \pm 0.1 \text{ kg ha}^{-1}$ ). Based on the MaP collection (Fig. S6), we noticed that MaPs from cotton fields and vegetable fields were mainly white and black plastics, corresponding to residues of white mulch films and black mulch films, respectively. However, the major colors of MaPs from greenhouses with mulching, greenhouses without mulching, and orchards were light blue and black, light blue, and transparent, respectively. A strong significant linear correlation was identified between MaPs and plastic mulch usage over the past five years in Quzhou county (Fig. S3), which implies that with every  $100 \text{ kg ha}^{-1}$  of plastic mulch usage over five years in farmlands, an additional  $8.9 \text{ kg ha}^{-1}$  of MaPs might be present in the fields.

### 3.2. Abundance and characteristics of MiPs among different farming systems

The average abundance of MiPs in different farming systems at depths of 0–30 cm ranged from  $8.5 \times 10^3$  to  $2.7 \times 10^4$  items  $\text{kg}^{-1}$  with an average value of  $1.2 \times 10^4$  items  $\text{kg}^{-1}$  (Fig. 3a). The highest abundance of MiPs ( $2.1 \times 10^4 \pm 1.1 \times 10^4$  items  $\text{kg}^{-1}$ ) was found in cotton fields, followed by permanent orchards ( $1.4 \times 10^4 \pm 3.7 \times 10^3$  items  $\text{kg}^{-1}$ ) and vegetable fields ( $1.3 \times 10^4 \pm 2.0 \times 10^3$  items  $\text{kg}^{-1}$ ). The average abundance of MiPs in fields with wheat-maize rotations was the lowest at  $6.8 \times 10^3 \pm 1.4 \times 10^3$  items  $\text{kg}^{-1}$ . The average MiP abundance in compost, groundwater and surface water were  $1.0 \times 10^3 \pm 908$  items  $\text{kg}^{-1}$ ,  $1.6$  items  $\text{L}^{-1}$  and  $0.3$  items  $\text{L}^{-1}$ , respectively (Fig. 3a and b). Among the six farming systems, cotton fields and permanent orchards had a significantly higher abundance of soil MiPs than fields with wheat-maize rotations.

The shape distribution of MiPs in the six farming systems is shown in Fig. 3c. The shape characteristics of MiPs in these systems were fairly similar. Fragment was the predominant shape, accounting for 45–62% of the particles found across the six farming systems. Particle-shaped MiPs constituted a relatively high proportion in farming systems, ranging from 32 to 45%. Additionally, a high proportion of fragmented MiPs was identified both in groundwater and compost samples, accounting for 67 and 81%, respectively. PCA was used to investigate the relationship between MiP shape distribution and different sampling sites (Fig. S4). Fig. S4 reflected the principal components (PCs) of 3 variables of MiP shape in Quzhou county, in which the first component showed strong contributions from fragment and particle shapes (59.1%), and the second component was dominated by fiber (36.3%). The size distribution of MiPs from six farming systems, compost and irrigation sources

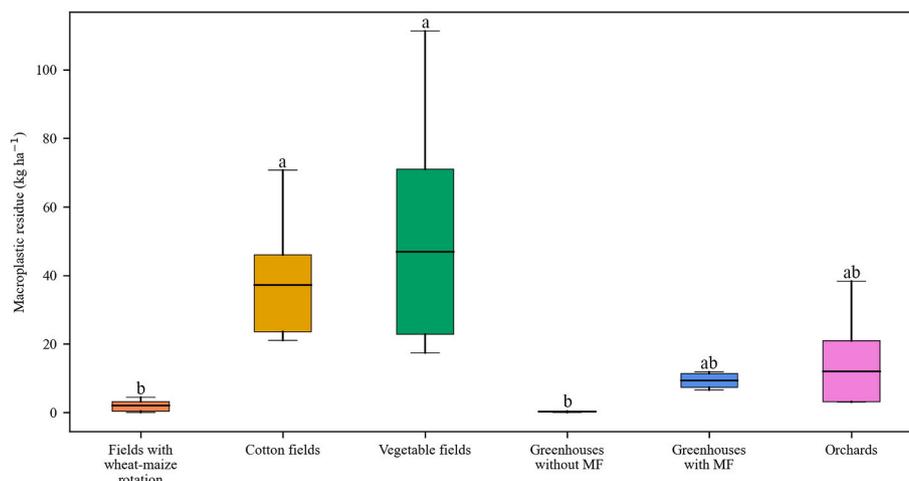
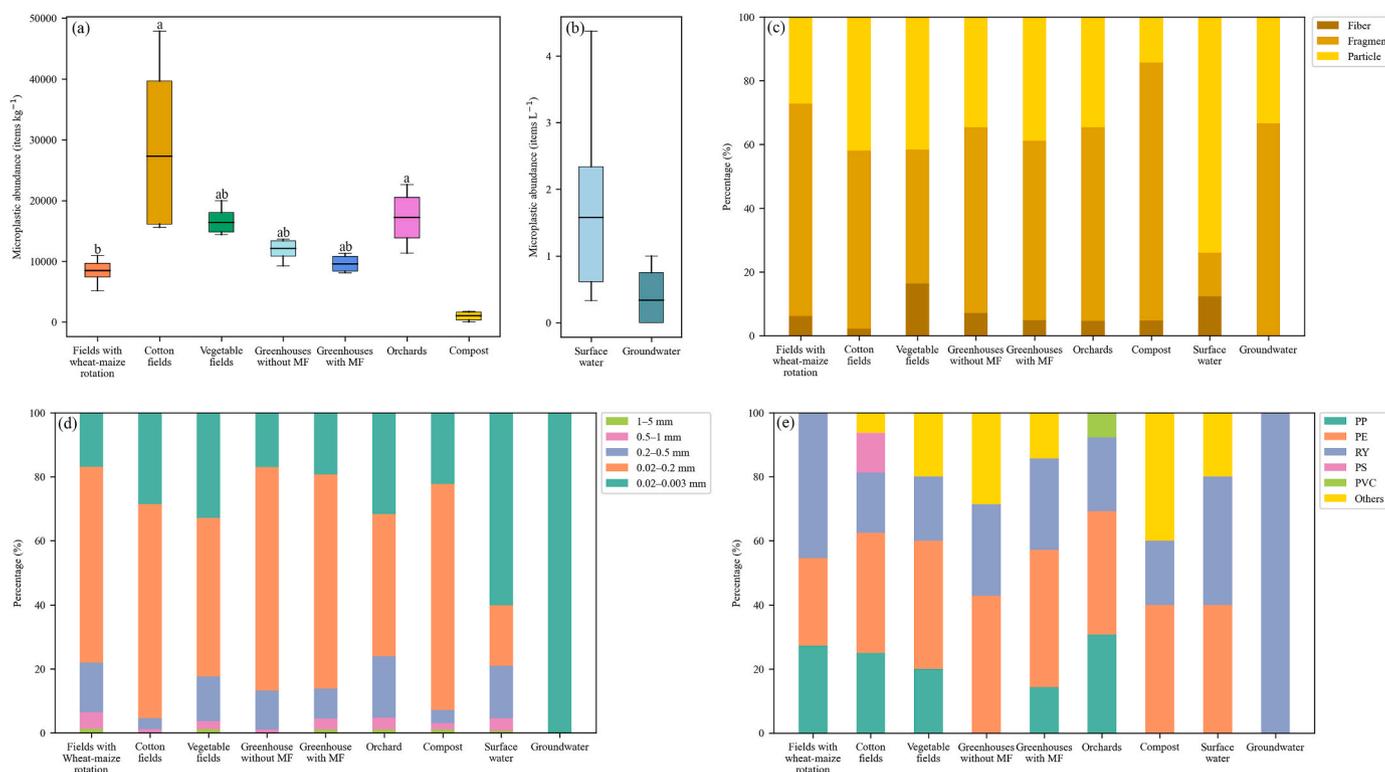


Fig. 2. MaP concentration ( $\text{kg ha}^{-1}$ ) in different farming systems. Greenhouse with MF refers to greenhouses with mulching, while greenhouse without MF refers to greenhouses without mulching. Different letters indicate significant differences between farming systems ( $p < 0.05$ ).



**Fig. 3.** MiP abundance (a, b), shape distribution (c), size distribution (d) and chemical composition (e) in samples from the six farming systems, compost, groundwater and surface water. Greenhouse with MF refers to greenhouses with mulching practices, while greenhouse without MF refers to greenhouses without mulching practices. Different letters indicate significant differences between farming systems ( $p < 0.05$ ).

are shown in Fig. 3d. This shows that MiPs < 1 mm were dominant, constituting 98–99% of all farming systems. MiPs between 0.2 and 0.02 mm accounted for the highest proportion, 45–70% in the six farming systems. A total of 97 particles were selected to identify chemical compositions using  $\mu$ -FTIR, and 78.72% of which were finally confirmed as MiPs. Fig. 3e shows the chemical composition of identified MiPs from different farming systems, compost and irrigation sources, including polypropylene (PP), polyethylene (PE), rayon (RY), polystyrene (PS), Polyvinyl chloride (PVC), and others, such as Nylon. Notably, the dominant chemical composition of MiPs in farmlands, except for the fields with wheat-maize rotations, was PE, with the percentage ranging from 37.5 to 42.86% across different farming systems. In contrast, the highest percentage of MiPs in farmlands with wheat-maize rotations was rayon (RY), accounting for 45.45%. Along with this, PE was common in compost samples (40%) and surface water samples (40%), and all the polymers found in groundwater sources were RY (100%).

### 3.3. Abundance and characteristics of MiPs in different soil layers of farming systems

The abundance of MiPs in all soil samples at different soil depths is shown in Fig. S5a, and Fig. S5b. Overall, the MiP abundance ranged from  $1.2 \times 10^3$ – $1.7 \times 10^4$  items kg<sup>-1</sup> in the 20–30 cm soil layer, which was significantly lower than MiP abundance in the 0–10 cm layer ( $7.2 \times 10^3$ – $3.5 \times 10^4$  items kg<sup>-1</sup>) and 10–20 cm soil layer ( $5.2 \times 10^3$ – $2.2 \times 10^4$  items kg<sup>-1</sup>). Cotton fields had the highest amount of MiPs in all three soil layers, ranging from  $1.7 \times 10^4$  to  $5.4 \times 10^4$  items kg<sup>-1</sup> in the 0–10 cm soil layer, from  $1.8 \times 10^4$  to  $4.3 \times 10^4$  items kg<sup>-1</sup> in the 10–20 cm soil layer, and from  $6.3 \times 10^3$  to  $4.7 \times 10^4$  items kg<sup>-1</sup> in the 20–30 cm soil layer. On the contrary, the MiP abundance in fields with wheat-maize rotations showed the lowest value for both top ( $7.3 \times 10^3$ – $1.7 \times 10^4$  items kg<sup>-1</sup>) and middle layers ( $6.6 \times 10^3$ – $1.2 \times 10^4$  items kg<sup>-1</sup>), whereas the amount of MiPs in greenhouses with mulching had the

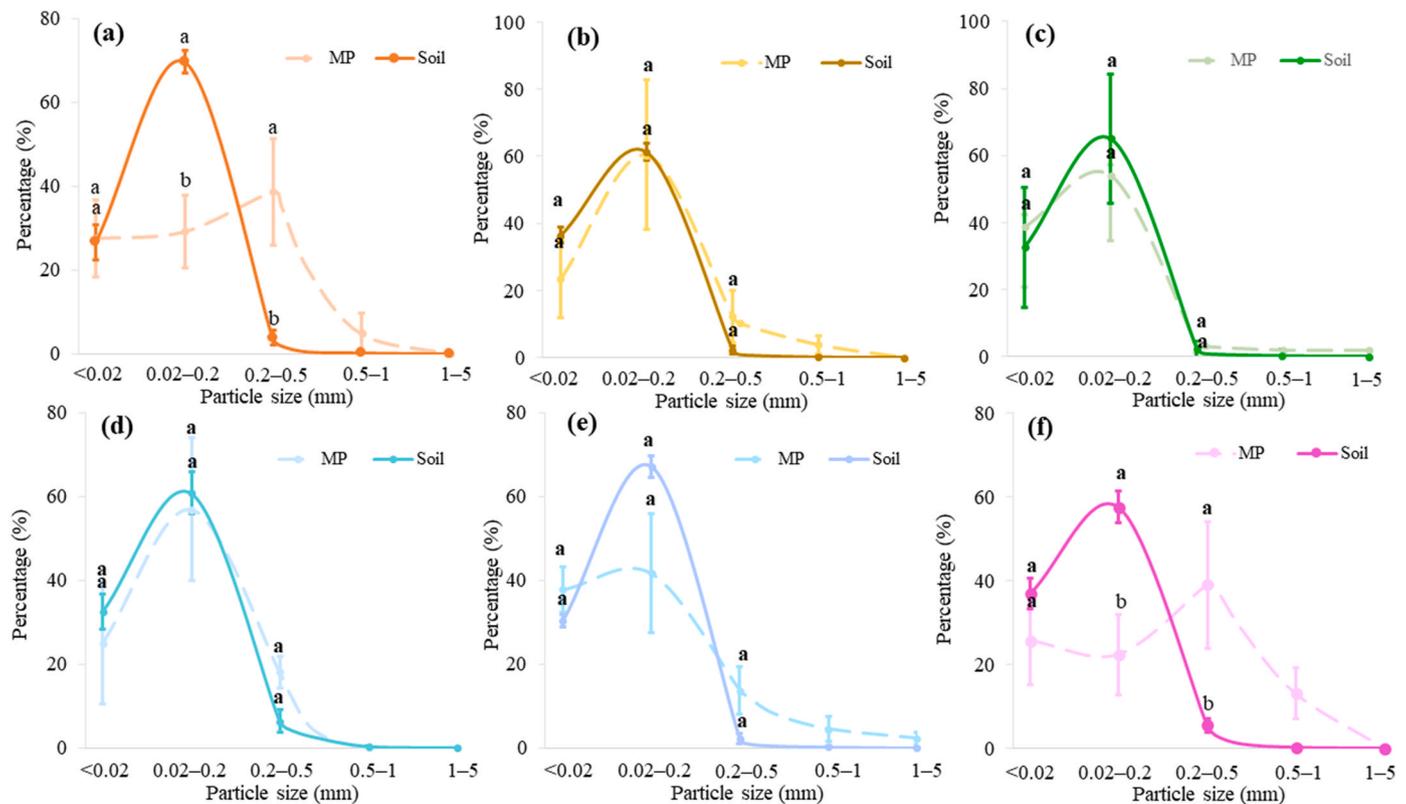
lowest value in the bottom layer ( $1.2 \times 10^3$ – $7.0 \times 10^3$  items kg<sup>-1</sup>). There was no significant difference in MiP abundance among the three soil layers in cotton fields and orchards.

Shape distribution and size distribution of MiPs across the three soil layers are presented in Fig. S5c and Fig. S5d. The dominant shape across the three layers was fragment, ranging from 56.1% to 57.1%. In different soil layers, more than 99% of MiPs were less than 1 mm. Size and shape distribution in different soil layers were similar.

### 3.4. Factor analysis of soil MiPs among different farming systems

The frequency distribution curve of both MiP size and soil particle size in the same sampling site are presented in Fig. 4. Across some farming systems, including cotton fields, vegetable fields, and greenhouses with or without mulching, there was no significant difference ( $p > 0.05$ ) between size distribution of MiPs and that of soil particles. Otherwise, the distribution trends of MiPs in wheat-maize rotations and orchards differed from those of soil particle size distribution. In these two farming systems, the dominant size range of soil particles was 0.02–0.2 mm, which was smaller than the dominant size range of MiPs (0.2–0.5 mm).

A redundancy analysis was conducted to investigate the correlation between agricultural management practices (ploughing frequency, application years of plastic films, and MaP concentration representing plastic management) and MiP size distribution. However, the soil particle size distribution varied across different farming systems, potentially influencing the MiP size distribution (Ouyang et al., 2023; Ren et al., 2020). To minimize the bias caused by the inhomogeneity in soil particle size distribution (Table S2), the ratio of MiP items to soil particle weight within a specific fraction was used to represent the MiP size distribution. RDA1 and RDA2 together explained 70.6% of the total variance. Ratios for large-sized fractions (0.2–0.5 and 0.5–1 mm) exhibited a strong negative association with ploughing frequency.



**Fig. 4.** Frequency distribution curve of soil particle size and MiP size in (a) fields with wheat-maize rotations, (b) cotton fields, (c) vegetable fields, (d) greenhouses without mulching practices, (e) greenhouses with mulching practices, and (f) permanent orchards. Different lowercase letters indicate significant differences between MiP size distribution and soil particle size distribution within same size fractions ( $p < 0.05$ ).

Conversely, ratios for smaller-sized fractions (0.02–0.2 and  $< 0.02$  mm) showed a low positive relationship with ploughing frequency, but a strong positive relationship with the years of plastic film application and plastic residues.

## 4. Discussion

### 4.1. MaP accumulation in different farming systems

This study provided firsthand evidence of the concentration of soil MaPs in different farming systems. Vegetable fields and cotton fields had the highest MaP abundance of all farming systems in Quzhou, which were much lower than same farming systems in other regions (Zhang et al., 2020a), especially in Gansu province ( $75 \text{ kg ha}^{-1}$ ) (Cao et al., 2023), Xinjiang province ( $230.9 \text{ kg ha}^{-1}$ ) and Inner Mongolia ( $77.6 \text{ kg ha}^{-1}$ ) (Huang et al., 2020). Compared to these regions, Quzhou County has a lower input of agricultural film and a shorter history of mulch film application (Ren et al., 2023), resulting in a relatively lower accumulation of MaPs in agricultural soil. The abundance of MaPs in wheat-maize rotations and greenhouses in Quzhou was similar to values from the same farming systems in other regions (Saadu et al., 2023; Zhang et al., 2020a), but the abundance of MaPs in orchards in Quzhou was lower than that in Argentina (Berenstein et al., 2024), which may be due to the limited sources (e.g. plastic fruit bags) of MaPs in orchards in Quzhou county (Fig. S1).

MaP accumulation can strongly weaken crop growth and soil physicochemical properties, including plant height, plant weight, soil water infiltration rate, and soil organic matter, thereby decreasing the crop yield (Zhang et al., 2020a). To mitigate the MaP accumulation in soil, it is necessary to investigate the potential sources of MaPs in different farming systems. In terms of wheat-maize rotations in Quzhou, although farmers did not apply any agricultural plastic products in farmlands,

MaPs can be transported through wind erosion and human activities (Ren et al., 2023). Conversely, greenhouses, being enclosed environments, can more effectively impede the migration of MaPs than open-field farmlands. In greenhouses, MaP accumulation is primarily attributed to the use of plastic strings and greenhouse films. In orchards, plastic fruit bags are the main source of MaPs.

Most MaPs in farmlands with mulching, including cotton fields, vegetable fields, and greenhouses with mulching, came from plastic mulch film. Among these farming systems, cotton fields and vegetable fields have a relatively long history of mulching and low recycling rates (Table S1), leading to a significant accumulation of MaPs in agricultural soil. Zhang et al. (2020a) found that cotton and vegetable fields are the two major sinks of MaPs, which is consistent with our findings. Overall, although MaP accumulation in Quzhou county is not as serious as in other regions, complex sources and remarkable contributions of mulch film still highlight the fact that agricultural plastic management is needed.

### 4.2. MiP accumulation in soils under different farming systems

MiP characteristics in different farming systems can infer the potential sources of MiPs in Quzhou county. Primarily, agricultural plastic applications mainly contribute to the accumulation of soil MiPs (Ren et al., 2023). Apart from fields with wheat-maize rotations, other farming systems employ a variety of agricultural plastic products, primarily made from PE and PP (Zhang et al., 2021), which are consistent with the dominant chemical types of soil MiPs found in these farming systems. Conversely, the lack of agricultural plastic sources in fields with wheat-maize rotations may explain the lower percentages of PP and PE detected. From the application to the disposal of plastic products in farmlands, plastics undergo continuous fragmentation, releasing smaller MiPs through biodegradation, ultraviolet radiation and mechanical

weathering processes (Lambert and Wagner, 2016). The predominance of smaller MiPs (<1 mm) among different farming systems implies that soil MiPs might suffer fragmentation processes in farmlands (Song et al., 2020; Yang et al., 2021a; Yang et al., 2022).

Except for agricultural plastic applications, other potential sources of MiPs also influence MiP occurrence. According to the results of survey (Table S1), it is evident that the applications of compost and irrigation continuously introduce MiPs to agricultural soil. Specifically, compost mainly introduces fragmented and PE-made MiPs to greenhouses, orchards, and vegetable fields. Groundwater primarily supplies fragmented and RY-made MiPs to fields with wheat-maize rotations and cotton fields, while particles from other farming systems might originate from irrigation by surface water. Furthermore, Li et al. (2023a) reported that the dominant type of MiPs from deposition in Quzhou county was RY, suggesting that atmospheric deposition might be one of the sources for RY-made MiPs in open fields. Along with these, human activities could also be potential sources, as PS and PVC are widely used in daily life (Dai et al., 2018; Zhang et al., 2022). Domestic washing processes for synthetic cloth and the use of personal care products might provide fibrous and spherical MiPs, respectively (Xu et al., 2020b).

When compared to MiP abundance in other agricultural fields worldwide, such as Urumqi in China ( $7.8 \times 10^3 \pm 2.8 \times 10^3$  items  $\text{kg}^{-1}$ ) (Li et al., 2023b), Wuhan in China ( $1.3 \times 10^4$  items  $\text{kg}^{-1}$ ) (Chen et al., 2020), Northern Germany ( $3.7 \pm 11.9$  items  $\text{kg}^{-1}$ ) (Harms et al., 2021), and Chile ( $306 \pm 360$  MiPs  $\text{kg}^{-1}$ ) (Corradini et al., 2021), the abundance of MiP in Quzhou county was relatively high, which could be attributed to the substantial contribution of MiPs from various sources. Although MiP concentrations in irrigation sources and compost in this study were quite moderate (Horton et al., 2017; Zhang et al., 2022), atmospheric deposition in Quzhou county contains remarkably higher amounts of MiPs ( $86\text{--}7.5 \times 10^4$  particles  $\text{m}^{-2} \text{day}^{-1}$ ) (Li et al., 2023a) compared to other regions in China, such as Guangzhou ( $51\text{--}178$  particles  $\text{m}^{-2} \text{day}^{-1}$ ) (Huang et al., 2021) as well as Paris, France ( $29\text{--}280$  particles  $\text{m}^{-2} \text{day}^{-1}$ ) (Dris et al., 2015). Additionally, annual tillage and UV exposure (Table S1) could accelerate the fragmentation of MaPs and MiPs, leading to the generation of ever-increasing MiPs (Ouyang et al., 2023; Yang et al., 2022). In summary, multiple MiP sources and fragmentation processes resulting from intensive farming have led to the high accumulation of MiPs in soil, indicating a severe MiP pollution issue in Quzhou county.

Once MiPs enter soil, they could be vertically transported through the soil profile via soil disturbance, biota activities, and infiltration (Ren et al., 2021; Rillig et al., 2017). Among these factors, tillage practices, serving as one form of soil disturbance, might explain the abundance distribution of MiPs along the soil profile. The abundance of MiPs in the bottom soil layer was significantly lower than its abundance in the middle and topsoil layers, but there was no significant difference of MiP abundance between 0–10 cm and 10–20 cm. Based on our survey, the tillage depth in Quzhou county was predominantly around 15–20 cm, implying that tillage practices could effectively move MiPs to the depth of the plow. Previous research (Hu et al., 2021; Huerta Lwanga et al., 2017) also indicated that there was no significant difference in the MiP abundance in the cultivated layer, but MiP abundance in the cultivated layer was significantly higher than in non-cultivated layer.

Apart from tillage, other factors also effect the distribution of MiPs along the soil profile. Among different farming systems, no significant differences of MiP abundance were observed among the three soil layers in cotton fields and orchards. In terms of cotton fields, sampling of certain fields was conducted after plowing, which might explain the distribution of MiP abundance in the cultivated layer. Non-significant differences in the distribution of MiP abundance in the cultivated layer and the non-cultivated layer might be explained by infiltration and biota activities (Rillig et al., 2017). As for orchards, our survey conducted in Quzhou county showed that compost was applied at a soil depth between 20 and 30 cm before sampling, revealing that soil disturbance by compost application may lead to the non-significant

distribution of MiP abundance along the soil profile.

The uniform distribution of MiP size and shape among the three soil layers in this study also reflects the vertical migration of MiPs involving multiple factors. Although previous studies (Liu et al., 2018; Yu et al., 2021) found that larger MiPs were more likely to accumulate in the top soil layer, our results showed that the size of soil MiPs was uniformly distributed among the three soil layers, which might be attributed to the frequent soil disturbances, infiltrations and biotic activities in Quzhou county (Rillig et al., 2017; Yu et al., 2019). Similarly, shape distribution in different soil layers were roughly the same, which is consistent with previous studies (Li et al., 2023b; Xu et al., 2022). Helm (2017) realized that shape characteristics might reveal the potential source of MiPs, implying that the major sources of MiPs entering agricultural soil have remained unchanged in recent decades.

In conclusion, agricultural management practices, such as irrigation, compost application, plastic management and tillage practices, could strongly effect MiP accumulation in different farming systems.

#### 4.3. Interactions between intensive agricultural management and MiP accumulation in soil

As mentioned above, farming systems have a significant effect on MaP and MiP accumulation. When we concentrate on the size distribution of MiPs, Fig. 4 shows that agricultural management practices within different farming systems, such as tillage practices and plastic management, may result in diverse size distributions of MiPs due to fragmentation processes. Mechanical wear and UV exposure can affect the fragmentation processes of MiPs and MaPs, resulting in an increasing abundance of MiPs as particle size decreases (Bai et al., 2022; Song et al., 2017; Yang et al., 2022). According to survey (Table S1), a great number of MaPs left in the soil after the harvest in cotton fields and vegetable fields may contribute to the accumulation of smaller MiPs (<0.2 mm) in agricultural soil due to continuous UV exposure. Tillage in vegetable fields, and in greenhouses with or without mulching practices were carried out twice a year, indicating that the fragmentation process might be accelerated by intensive tillage, leading to a substantial generation of smaller MiPs (<0.2 mm). Meanwhile, vertical mobility of MiPs in soil increased with decreasing size (Dong et al., 2018; Gao et al., 2021). Hence, although a considerable number of small MiPs may be produced, those smaller than 0.02 mm are more prone to entering deeper soil layers through biotic activities and infiltration compared to MiPs ranging between 0.02 and 0.2 mm, which leads to a decline in MiP abundance with decreasing size. Overall, in terms of farming systems except those with wheat-maize rotations and orchards, improper management of agricultural plastics, intensive tillage and vertical migration of MiPs together result in no significant difference between the size distribution of MiPs and soil particles. Conversely, in the case of fields with wheat-maize rotations and orchards, the lack of plastic applications and the low frequency of tillage could result in slower fragmentation processes creating MiPs, thereby resulting in larger MiPs.

The correlation between the size distribution of soil particles and that of MiPs under different farming systems might illustrate the influence of intensive farming on MiP size distribution, which can be further elucidated by considering the results of RDA. Previous research suggests that tillage practices contribute to the generation of MiPs by fragmenting larger MaPs or even larger MiPs (Maqbool et al., 2024). However, as shown in Fig. 5, ploughing frequency is negatively associated with ratios in large-sized fractions (0.2–0.5 and 0.5–1 mm) and weakly positively correlated with ratios in smaller fractions (<0.02 and 0.02–0.2 mm). The strong negative correlation between ploughing frequency and larger fractions might be related to farming history as mulch and greenhouse films are typically applied only once a year. With frequent tillage, plastic film residues undergo accelerated breakdown into smaller fragments. However, smaller MiPs have high mobility, allowing them to move into deeper soil layers (>30 cm) through infiltration and biotic activities (Rillig et al., 2017). This movement of smaller MiPs into deeper soil

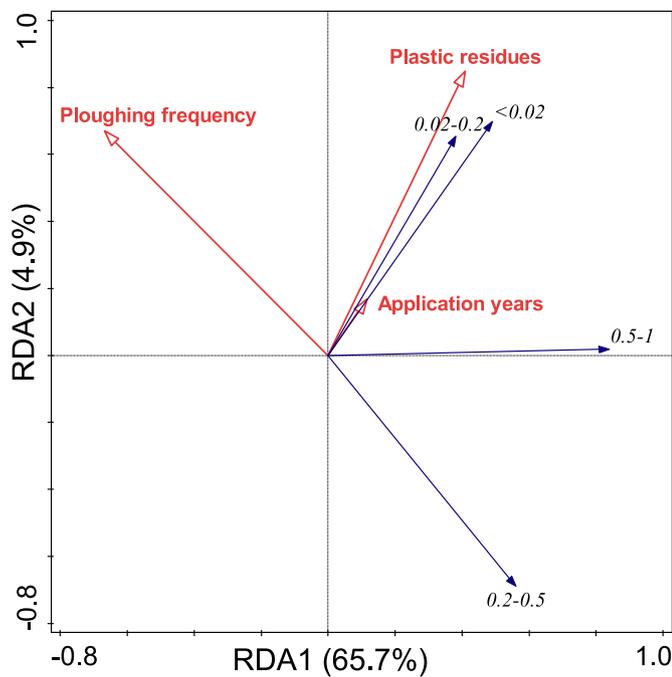


Fig. 5. Redundancy analysis of agricultural activities and ratios of MiPs/soil in different size fractions.

layer beyond 0–30 cm likely results in the weak correlation between tillage frequency and smaller MiP abundance in RDA analysis. Additionally, increasing years of plastic film application and increasing concentrations of MaPs can strongly enhance the accumulation of smaller-sized MiPs (<0.02 and 0.02–0.2 mm), potentially attributing to fragmentation processes induced by UV radiation. Notably, two vectors representing smaller-sized fractions indicate that MiPs smaller than 0.2 mm likely share similar sources and fates.

In summary, frequent tillage, long mulching histories and high abundance of MaPs play main roles in the size distribution of MiPs, leading to the continuous fragmentation processes of larger MiPs and the accumulation of smaller MiPs in agricultural soil (Song et al., 2017; Yang et al., 2022). These findings suggest that farmlands with intensive farming could face profuse accumulation of MaPs and MiPs, especially smaller MiPs. In China, for example, Xinjiang and Shandong are the typical film-mulching regions, with  $2.4 \times 10^5$  tons and  $1.1 \times 10^5$  tons used in 2018, respectively (NBSC, 2020). The application of mulch film in these regions has a long history of over 30 years. Xinjiang typically conducts single-crop cultivation annually, whereas Shandong cultivates at least two crops per year. The intensive use of mulch films and tillage practices in both regions likely contributes to significant accumulation of MiPs in the soil, with concentrations ranging from  $1.1 \times 10^4$  to  $7.8 \times 10^4$  items  $\text{kg}^{-1}$  in Xinjiang and 310 to  $5.7 \times 10^3$  items  $\text{kg}^{-1}$  in Shandong (Liu et al., 2022; Yu et al., 2021). Several studies (Qi et al., 2023; Yu et al., 2021) revealed that the dominant size and chemical composition of soil MiPs in Xinjiang and Shandong were <0.5 mm and PE, implying that serious fragmentation processes exist possibly due to an enrichment of MaPs, a long history of plastic film application and frequent tillage practices (Ren et al., 2023; Zhang et al., 2015). As countries that have been using mulch films on a large scale for over 30 years, South Korea, Germany, and Spain have not observed large amount of MaPs and MiPs in their fields with intensive farming. However, the remarkable accumulation of smaller MiPs in farmlands still reveals that these regions may face the risk of pollution of smaller MiPs due to intensive agriculture (Beriot et al., 2023; Kim et al., 2021; Piehl et al., 2018). Smaller MiPs pose potential risks to the health and function of terrestrial ecosystems, including soil respiration (Liu et al., 2023b), health of earthworms and plant functional traits (Huerta Lwanga et al., 2016; Pehlivan

and Gedik, 2021), and even groundwater contamination through leaching or penetration into soil pores (Viaroli et al., 2022). Thus, assessing pollution by smaller-MiPs is urgently needed, especially in farmlands with intensive farming. The fragmentation processes of plastics in farmlands are driven by multiple factors, such as mechanical wear, UV radiation and biodegradation (Lambert and Wagner, 2016), which might have different effects on the occurrence and characteristics of MiPs (Meng et al., 2023; Song et al., 2017). Therefore, further studies could also focus on figuring out the mechanism of fragmentation processes derived from different factors, which could help us assess and predict MiP pollution in agriculture with various agricultural management practices.

## 5. Conclusion

This study investigated the characteristics of MiPs and MaPs among six farming systems in Quzhou county where orchards, vegetable fields, and cotton fields exhibited higher concentrations of MaPs and larger abundances of MiPs than greenhouses and wheat-maize rotations. Finding the dominant size of MiPs (<1 mm), shape (fragment) and polymer (PE), indicates that agricultural plastic products, such as mulch films, are the important sources of MaPs and MiPs. These particles may further undergo severe fragmentation processes. Agricultural management practices, such as tillage and tillage frequency, significantly impact the distribution of MiPs in cultivated soil layers. In conclusion, intensive farming, such as frequent tillage practices and agricultural plastic management practices in cotton fields, vegetable fields and greenhouses, seems to accelerate the fragmentation of MaPs and MiPs, leading to the accumulation of smaller MiPs in soil. Although there are some reports working on the screening of macro-, micro- and/or nano-plastics, understanding the process of plastic granulation and interactions with soil particles are urgently needed to assess their risks in the soil ecosystem.

## CRedit authorship contribution statement

**Hanyue Zhang:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation. **Xiaomei Yang:** Writing – review & editing, Supervision, Conceptualization. **Kai Wang:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Jixiao Cui:** Writing – review & editing, Resources. **Coen J. Ritsema:** Supervision. **Changrong Yan:** Resources. **Xuejun Liu:** Supervision, Funding acquisition. **Violette Geissen:** Writing – review & editing, Supervision.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

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## Data availability

Data will be made available on request.

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