

RESEARCH ARTICLE

# Distribution characteristics of plastic film residue in long-term mulched farmland soil

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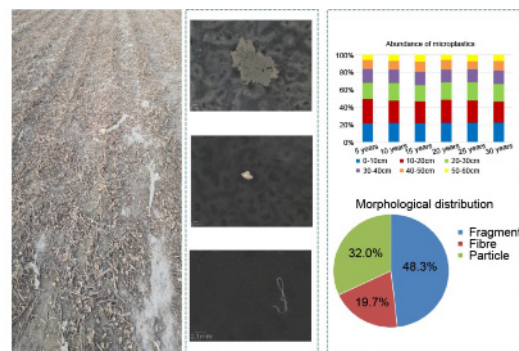
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## HIGHLIGHTS

- Occurrence of microplastics was widespread in long-term mulched farmland soils.
- Abundance exhibited obvious differences in different film mulching durations.
- Plastic film residue was the important source of farmland soil microplastics.
- This study offers useful data on microplastic pollution in long-term mulched areas.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Soil contamination from film debris following the prolonged application of mulching film has emerged as a worldwide concern. However, the extent that mulching films contribute to soil microplastics, and the spatial distribution of soil contamination from film debris remain unclear. In this study, the cotton field in Xinjiang (China), which underwent film mulching for a prolonged period of 5–30 years, was selected as the research location. A total of 360 soil samples were collected, aiming to study the spatial distribution characteristics of mulching film debris pollution. The samples were extracted using the density flotation method combined with stereomicroscopic; the source, composition, abundance, and distribution characteristics of soil MPs were identified by the scanning electron microscopic, and Fourier transform infrared spectroscopic analyses. In soil mulched for a 30 year period, the abundance of microplastics across the studied soil depth (0–60 cm) was  $78.51 \pm 2.57$  n/(100 g). The  $\mu$ -FTIR analyses revealed that the composition of the microplastics matched that of polyethylene materials. Therefore, plastic mulching could be inferred as a major contributor to microplastic pollution in agricultural lands. Overall, it is necessary to study the distribution characteristics of plastic film remaining for further study of plastic pollution in farmland soils.

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## 1 Introduction

The extensive use of plastic products has increased the annual production of plastics by more than 300 million tons worldwide since 2014 (Kaur et al., 2022). However, the

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overuse of plastic products and their lower recycling levels, ranging from 6%–26%, enable many waste plastics to enter the natural ecological environment (Alimi et al., 2018). The agricultural mulching film, a plastic film used for agricultural crop production during the sowing and growth periods, has become a major tool for maintaining increased temperatures, moisture conservation, and pest control (Yan et al., 2014; Sun et al., 2020), contributing greatly to food safety in the arid areas worldwide. Recently, residual plastic film debris-induced soil contamination has been observed in these areas due to the lack of prolonged, continuous, and widespread film mulching application in some farmland regions and sufficiently effective plastic mulching film recycling methods. The mulching film used in this study was the polyethylene (PE) material with a stable molecular structure, with non-degradable properties in soil environments (Wang, 1998), contributing to plastic pollution. The PE films might gradually decompose, with the influence of light, ultraviolet radiation, or mechanical tillage, which might be the source of a large number of microplastic particles and plastic component compounds in the soil.

Microplastics (MPs) are an emerging pollutant type of high global concern and defined as plastic debris and particles with a particle size smaller than 5 mm (Thompson et al., 2004; Andrady et al., 2011; Blettler et al., 2017). In recent years, the extent of environmental pollution and ecological damage by MP pollution has increased worldwide concern. Studies have identified varying levels of MP pollution in all aquatic environments worldwide, including the oceans (Zhang et al., 2019b), lakes (Anderson et al., 2017; Dusaucy et al., 2021), rivers (Xiong et al., 2019), and sediments (Ding et al., 2019). Rillig (2012) first raised the MP pollution related concerns in the soil ecosystems in 2012. Recently, several studies have assessed the potential of MP pollution in the soils. For instance, the plastic debris in the farmland surface soils could be slowly degraded due to aging by ultraviolet radiation and mechanical abrasion, leading to the transfer and accumulation of fine microplastic particles in deeper soil layers (Song et al., 2017). Liu et al. (2018) reported that the concentration and particle size of MPs in the vegetable field surface soils in suburban Shanghai were higher than those in deeper soil layers. However, the relative contribution of different sources of MPs present in the farmland soils remains unclear. Nizzetto et al. (2016) and Zhang et al. (2019c) found that film mulching and the application of sludge and compost were the primary factors responsible for the accumulation of MPs by the agricultural soil ecosystem. Zhou et al. (2020) investigated the non-mulched and mulched farmlands in the coastal plain of Hangzhou (Eastern China) and found that the average abundance of soil MP particles in the mulched farmland was about two-fold higher than in the non-mulched farmland.

The mulching film residues are a primary source of environmental pollution in farmland soils subjected to a prolonged and continuous film mulching, a practice commonly applied in arid crop-producing fields, such as

northwest China (Huang et al., 2020). Studies have reported that the presence of a large volume of plastic debris in the soil might affect the gas exchange capacity and drip irrigation transport of water throughout the soil (Hodson et al., 2017), reducing the soil quality, affecting crop absorption of organic fertilizers and water from the soil (Wang et al., 2021), germination rate and seed growth process, and reducing crop yields (Xiao et al., 2022). Some scholars pointed out that MPs impact soil organic matter and agricultural ecosystem function (Xiao et al., 2021; Tong et al., 2022). Accumulating studies have proposed that film mulching is a potential source of MPs in farmland soils (Ramos et al., 2015; Qi et al., 2018). However, no comprehensive evidence on MP formation from mulching films is reported, except for the theoretical information, confirming MP formation by film fragmentation. Additionally, there is limited information about the distribution characteristics of plastics in the farmland soil of long-term coverage areas.

The typical cotton planting area in Xinjiang (northwest China) was selected as the study location to assess the extent of soil pollution by the plastic film debris as a result of long-term mulching film application. Xinjiang is an important cotton production region in China, accounting for 2.502 million ha of the total cotton planting area in China of 3.169 million ha (China Statistical Yearbook, 2021; Xinjiang Statistical Yearbook, 2021). The objectives of this study were (i) to compare the abundance of plastic in the farmland soil at different film mulching periods; (ii) to detect the appearance characteristics and polymer types of MPs through a microscope and Fourier transform infrared (FTIR) spectroscopy; (iii) to explore the distribution law of plastic waste in the farmland with different film mulching years, and identify the potential sources of plastic waste. This research might increase the awareness of plastic damage to farmland soil. Additionally, the influence of prolonged and continuous film mulching on plastic residue distribution aspects will provide a novel interpretation of the risks associated with the application of mulching films and establish effective microplastic pollution control systems and technologies.

## 2 Materials and methods

### 2.1 Sample collection

The sampling sites were located in Xinjiang (northwest China), an arid region with low levels of annual rainfall. Crop planting in this region is now heavily dependent on film mulching. As for cotton, it is typically planted under continuous film mulching conditions and water-saving drip irrigation systems (i.e., long-term continuous film mulching). The average mulching film coverage in the sampling area is 82.50 kg ha<sup>-1</sup>. The soil samples were collected from 60 sampling points in the cotton fields subjected to 5, 10, 15, 20, 25, and 30 years of continuous mulching (10 sites in

each duration) (Alar and Huyanghe, Xinjiang), with a maximum area of 40 ha and a minimum area of 2 ha. Before the spring sowing of cotton seeds, the geographical coordinates of 50 sampling points were obtained in typical film-mulched cotton fields around Alar using the Global Positioning System (GPS, USA) tool, while the coordinates of 10 sampling points were obtained in Huyanghe. The distribution of sampling points is depicted in Table 1. A 100 cm × 100 cm square was randomly selected using a square frame at each sampling site, allowing the soil layers to be removed at 10 cm depth intervals. The vertical profile of each sampling point was divided into 6 soil layers (each soil layer 10 cm deep) (Figure S1). The mulching film debris of each layer was carefully screened and passed through a 5 mm standard test sieve (GB/T6003.1-2012, 4 mesh, Shaoxing Shangyu Huafeng Hardware Instrument Co. Ltd.,

China). The organic debris and impurities, such as clods, cotton stalks, and roots, were removed before transferring the soil layers into the sample bags. Additionally, 200 g of screened soil was collected in cotton bags from each sample to prevent plastic contamination. A total of 6 samples were collected from each sampling site, with a total of 360 soil samples collected from the 60 sampling points in the cotton fields subjected to film mulching for different durations (10 sites selected for each duration of 5, 10, 15, 20, 25, and 30 years).

## 2.2 Extraction methods

The plastic content of soil samples was extracted and classified as either film residue debris (particle size >5 mm) or microplastic particles (particle size <5 mm). The extraction methods used for the two plastic component types were as follows:

**Table 1** Geographic locations and sampling sites in the study area.

Sampling point	Longitude, latitude	Sampling point	Longitude, latitude
1	40.587528, 81.129919	31	40.566598, 81.123676
2	40.599506, 81.086881	32	40.566641, 81.123315
3	40.653235, 81.106923	33	40.566397, 81.123029
4	40.608733, 81.265253	34	40.545150, 81.019390
5	40.615720, 81.292167	35	40.545912, 81.019458
6	40.623539, 81.348234	36	40.545647, 81.019583
7	40.623436, 81.348144	37	40.550196, 80.983528
8	40.623275, 81.348327	38	40.550201, 80.983316
9	44.789023, 84.666481	39	44.846681, 84.689827
10	44.785930, 84.759520	40	44.631528, 84.884491
11	40.638902, 80.787108	41	41.328421, 80.837965
12	40.638808, 80.787129	42	41.328396, 80.837640
13	40.638946, 80.787323	43	41.328773, 80.837425
14	40.586986, 80.980275	44	40.562659, 81.046263
15	40.544595, 81.163036	45	40.481766, 81.224578
16	40.544435, 81.163126	46	40.483904, 81.333190
17	40.544610, 81.163179	47	40.652941, 81.494298
18	40.544027, 81.163324	48	40.667989, 81.522288
19	40.544348, 81.163293	49	44.812958, 84.702444
20	40.550709, 81.085708	50	44.762123, 84.803510
21	41.351220, 80.805746	51	40.581670, 80.850541
22	41.351187, 80.805621	52	40.485572, 81.269461
23	41.351606, 80.805352	53	40.482794, 81.244223
24	40.598399, 80.972263	54	40.507432, 81.328216
25	40.595738, 81.332725	55	40.701725, 81.563310
26	40.598624, 81.229216	56	40.554035, 81.480198
27	40.598733, 81.229864	57	40.625729, 81.449939
28	40.641487, 81.475719	58	40.605631, 81.469555
29	44.654734, 84.950409	59	44.706730, 84.766130
30	44.798403, 84.857712	60	44.678556, 84.945195

### 2.2.1 Extraction method for film debris samples

The film debris samples were transferred to the laboratory, and the soil and other impurities attached to the films were manually cleaned with water. Then, the samples were soaked in 200 mL of 30% H<sub>2</sub>O<sub>2</sub> for 24 h at 50°C to digest the organic matter on the surface. Afterward, the finer impurities attached to the surface of film debris were removed with distilled water for 30 min using an ultrasonicator (WN-030S, Weineng Ultrasonic Equipment Co. Ltd., China). The film debris was then dried naturally under ambient conditions. The weight of each piece of film debris was weighed using an electronic balance (precision 0.01 mg, Quintix35-1CN, Beijing saidoris Instrument Co. Ltd., China), and the statistical classification analysis was performed using Excel 2019. According to the weight of each piece, the film debris was classified into group 1 (0, 10] mg piece<sup>-1</sup>, group 2 (10, 25] mg piece<sup>-1</sup>, group 3 (25, 50] mg piece<sup>-1</sup>, group 4 (50, 100] mg piece<sup>-1</sup>, and group 5 (>100) mg piece<sup>-1</sup>. The number of pieces of plastic film debris in each group was counted per sample.

### 2.2.2 Extraction method for MP samples

The separation of microplastic particles was performed in a closed laboratory. Before separation, the collected soil samples were dried naturally at room temperature of 25°C. The MPs from the soil samples were extracted, following a previously reported density separation method with a saturated NaCl solution (Kumar et al., 2020; Chen et al., 2020; Wang et al., 2021). The extraction steps were as follows: (i) 10 g of soil from each sample was added to a 500 mL beaker with 200 mL of saturated NaCl solution, and then stirred continuously at 100–200 r min<sup>-1</sup> for 30 min using a magnetic stirrer under ambient temperature

conditions. Later, 50 mL of the saturated NaCl solution with a molarity of  $5 \text{ mol L}^{-1}$  was used to rinse the glass rod into the beaker, which was then covered with frosted glass to prevent the deposition of plastic particles in the air in the beaker. The solution was left to stand until the soil particles precipitated. (ii) The suspension was left to stand for 12 h, and then 100 mL supernatant was transferred into a 250 mL beaker with a glass straw after solid-liquid stratification. The inner wall of the glass straw was rinsed several times with a small amount of distilled water. The rinsed liquid was transferred to the beaker. The density flotation extraction method was repeated three times, reusing the same saturated NaCl solution. After density separation, 100 mL of 30% hydrogen peroxide (Shanghai Sinopharm group, China) was soaked for 12 h at  $50 \text{ }^\circ\text{C}$  to digest the organic matter. The final solutions were filtered through a  $0.45 \text{ }\mu\text{m}$  mixed cellulose filter membrane (HAWP04700, Shanghai Xuanyi Instrument Equipment Co. Ltd., China). The filter membrane containing the microplastic particles was transferred to a glass petri dish using stainless steel tweezers. (iii) The microplastic particles were collected using stainless steel fine-tip tweezers by under the observation of a stereomicroscope (TD-2KHU, Shenzhen Sanqiangtaida Optical Instrument Co. Ltd., China). The selected MPs were transferred to a slide, covered using a cover glass, and the microplastic particles were then observed at different magnifications. The number of microplastic particles collected from each soil layer was screened and counted visually. The appearance and morphology of MPs were photographed and recorded by a microscope connected to a high-definition display system. The microplastic particles were classified according to their morphological characteristics, and the quantity of each class was counted. We could only detect the MPs with a minimum particle size of  $1 \text{ }\mu\text{m}$  due to the limitation of more precise instruments, but there might be nano-sized MPs with smaller particle sizes that could not be accurately detected.

### 2.3 Method of identification of microplastic particles composition

The composition of the extracted MP samples was identified by FTIR spectroscopy (Thermo Scientific, Nicolet iN10, US) in  $\mu$ -FTIR transmission mode. The spectral data were collected at the wavelengths from  $650\text{--}4000 \text{ cm}^{-1}$ . Each sample was scanned 32 times with a resolution of  $4 \text{ cm}^{-1}$  and a duration of 3 s. The spectra in samples, which were screened against the polymer standard spectral library to determine the MP compound types, were identified using OMNIC software (Thermo Fisher Scientific, US). The obtained spectral characteristic peaks were compared with the characteristic peaks in the reference polymer library. When the recognition matching degree of the spectrogram exceeds 70%, the sample composition could be determined as the corresponding component (Liu et al., 2020; Wang et al., 2021).

### 2.4 Scanning electron microscope analysis

The morphology, surface pores, and cracks of the MP samples extracted from the soil were studied using scanning electron microscopy (SEM). Firstly, the MP sample was placed on the sample table conductive tape, and the taped sample table was placed on a carbon coater (JFC-1600 JEOL, Japan Electronics Co. Ltd.) at a current of 20 mA and a coating duration of 80 s. The sample table was placed on the electron microscope (JSM-6700F JEOL, Japan Electronics Co. Ltd.), initially in the sample exchange cell and then into the sample cell using a vacuum (up to  $5 \times 10^{-5} \text{ Pa}$ ) to start the test. The test voltage was adjusted to 5 kV, and the definition was adjusted according to the target area. Later, a slow scan was performed. The contrast and brightness of images were adjusted before saving. Meanwhile, the same acceleration voltage and amplification were provided to the energy dispersive X-ray spectrometry (EDS). The amp time constant was set to maintain a "Dead Time" at 20%–40%. After setting these two parameters, the spectrogram was collected, and the energy of different photons was detected to determine the element according to the characteristic response of X-rays with different frequencies emitted by different elements.

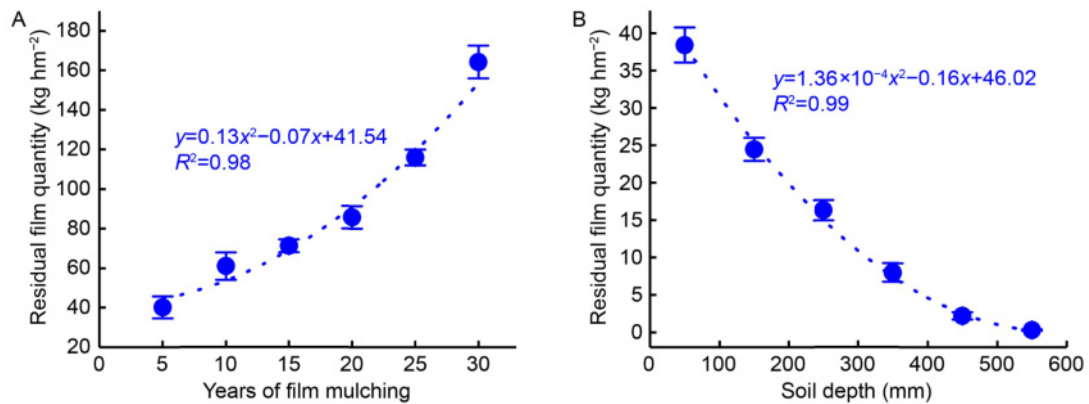
### 2.5 Statistical analysis

All the experimental results were expressed as mean values  $\pm$  standard error ( $X \pm \text{SE}$ ). The abundance of MPs in soils was expressed as  $(n \pm \text{SE}) \cdot (100 \text{ g})^{-1}$ . Statistical analysis was performed using the software IBM SPSS v.26, while Excel 2019 was used for data processing. The number and quality of film debris and the abundance of MPs in the soil were analyzed by one-way analysis of variance (ANOVA), and  $p$  values  $< 0.05$  were considered significant. ArcGIS v.10.6 and Origin 2017 software were used to plot the data.

## 3 Results and analysis

### 3.1 Abundance of plastic mulching film debris ( $> 5 \text{ mm}$ )

Different qualities and densities of mulching film debris were observed in all samples. The residual film quality in the agricultural fields with different mulching times is depicted in Figure 1A. The concentration of the mulching film debris in the soils that were continuously covered in mulching film for 5 years was  $40.22 \pm 5.53 \text{ kg ha}^{-1}$ . When the duration of continuous mulching time reached 10 and 15 years, the film debris content in soil reached  $60.84 \pm 6.91 \text{ kg ha}^{-1}$  and  $71.08 \pm 3.23 \text{ kg ha}^{-1}$ , respectively, which was close to the Chinese national standard limit of  $75.00 \text{ kg ha}^{-1}$ . The film debris content increased further to  $85.56 \pm 5.66 \text{ kg ha}^{-1}$ ,  $115.83 \pm 3.96 \text{ kg ha}^{-1}$ , and  $164.08 \pm 8.31 \text{ kg ha}^{-1}$  in the soil subjected to 20, 25, and 30 years of continuous film



**Fig. 1** (A) Residual film quantity in agricultural fields with different mulching times; (B) Residual film quantity in different layers of soils in agricultural fields.

mulching, respectively, exceeding the national standard limit for mulching film debris in soils. These findings indicated that the concentration of mulching film debris increased with increased mulching time durations. Additionally, the total weight of mulching film debris per sample was negatively correlated with the soil depth (Figure 1B). The average mass of debris in all 60 samples was  $(3.48 \pm 0.59) \times 10^4$  kg ha<sup>-1</sup> at depths of 30–60 cm. Figure 1 depicts the accumulation of mulching film debris on the surface and tillage layers (0–30 cm), while smaller film debris pieces in the soil migrated downward, allowing the film debris to be accumulated by deeper soil layers.

The distribution of large mulching film debris (>5 mm) in the cotton field soils subjected to different mulching times exhibited significant differences. The weight distribution of film debris in soils subjected to different mulching times is depicted in Figure S2. The quantity of mulching film debris in soil significantly increased with the mulching time. Additionally, the frequency of mulching film debris (i.e., the number of film debris pieces) was highest at a depth of 0–10 cm, reaching  $(66.40 \pm 5.15) \times 10^4$  pieces ha<sup>-1</sup>, and thereafter showed a decreasing trend with deeper soil depths, with an average of  $(7.97 \pm 1.13) \times 10^4$  pieces ha<sup>-1</sup> in the soil at 40–60 cm depths. The abundance of smaller film debris gradually increased with longer mulching durations (Figure S2). After 5 to 30 years of film mulching, the quantities of the film debris <50 mg increased significantly. However, film debris  $\geq 100$  mg also increased significantly with longer mulching durations. This might be because, after soil cultivation, the film debris migrates due to wind, run off or agricultural activities.

The weight distribution of the film debris in soils subjected to different mulching times is depicted in Figure S3. The results showed that the distribution of mulching film residues decreased in lower depths with increased mulching times. The weight of the film debris in 0–30 cm soil layers was significantly higher than in the 30–60 cm soil layers ( $p < 0.05$ ). Data on the average weight of film debris at different layers were assessed using regression analysis. The results indicated that the total amount of film debris decreased with the increase of soil depth and decrease in

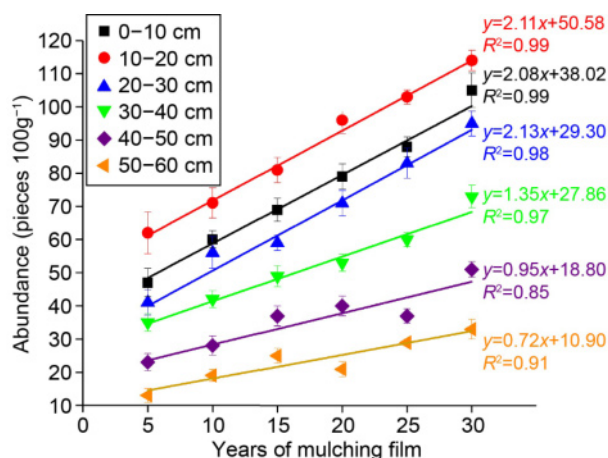
the weight of each piece of film debris. When the soil depth was greater than 40 cm, there were less visible film debris pieces, which were mainly finer and smaller than 0.03 g. The total mass of mulching film debris also decreased with increased soil depth, with the mass being the highest at the 10–20 cm soil depth. When the duration of continuous film mulching was 5 years, the number of film debris pieces in soil was the lowest (109 pieces of film debris) (Figure S3a). The film debris was mainly distributed in the upper 0–30 cm layer, and the average number of film debris pieces was 90 pieces. The average weight of the film debris in the 0–10 cm, 10–20 cm, and 20–30 cm soil layers was 56.95 mg, 75.75 mg, and 55.01 mg, respectively. However, the frequency of film debris detection in the 30–60 cm soil layer was 19 pieces only, while the film debris weight in the 30–60 cm soil layer decreased significantly to an average of 15.90 mg ( $p < 0.05$ ). When the mulching duration was 10–25 years, the number of film debris pieces increased significantly, reaching 145, 172, 228, and 258 pieces, respectively, indicating the gradual fragmentation of film debris. A higher abundance of small debris pieces weighing <0.05 g were observed in the 0–10 cm soil layer (Figures S3 b, c, d, and e). When the mulching duration was 30 years (Figure S3f), the number of film debris pieces increased further, reaching a total of 300 pieces of film debris. Among these, the number of film debris pieces in the 30–60 cm depth was 87 pieces. A large number of tiny mulch film weighing less than 0.05 g appeared in the 0–30 cm surface soil, indicating that the film residue was more fragmented and distributed in the whole surface soil. Moreover, the mulching film debris continually migrated to the soils at a depth of 30–60 cm. Figure S3 depicts that the residual mulching film debris in the soil becomes more fragmented with increasing time. Further analysis showed that the occurrence of film debris increased gradually with increased mulching durations, with smaller film debris particles infiltrate deeper soil layers. During the process of infiltration, the mulching film debris was continuously oxidized and broken down into smaller debris, gradually forming MPs.

### 3.2 Residual amount of microplastics

#### 3.2.1 Microplastic abundance

The abundance of MPs was used to represent the number of MPs present in the soil, and the abundance of MPs was assessed in the air-dried soil per 100 g ( $n(100g)^{-1}$ ). MPs were detected in all the farmland sampling sites, with 100% occurrence of MPs throughout the whole 0–60 cm soil layer. In all sampling sites, the average abundance of MPs in the 0–60 cm soil layers ranged from  $20.00 \pm 5.77$  to  $93.33 \pm 15.42$   $n(100g)^{-1}$ . MP abundance consistently decreased with the deepening of soil layers, indicating that MPs could gradually spread and migrate from shallow to deep soils.

The results showed that in the prolonged continuous mulching of cotton fields, the mulching film debris remained in the soils due to incomplete decomposition, contributing to a potential source of MP pollution. The abundance and distribution characteristics of MPs at different depths in soils subjected to different mulching times are depicted in Figure 2. The results showed that the abundance of MPs in the soil decreased with increased soil layer depth. The abundance of MPs was the highest in the 10–20 cm soil layer, which might be due to surface weathering combined with agricultural activities, such as tillage and drip irrigation, causing MPs to infiltration deeper into the soil. For instance, in the cotton fields subjected to 10 years of film mulching, the abundance of MPs in the (0, 100] mm, (100, 200] mm, (200, 300] mm, (300, 400] mm, (400, 500] mm, and (500–600] mm soil layers was  $60.00 \pm 2.58$ ,  $71.00 \pm 4.58$ ,  $56.00 \pm 4.76$ ,  $42.00 \pm 2.49$ ,  $28.00 \pm 2.91$ , and  $19.00 \pm 1.80$   $n(100g)^{-1}$  soil, respectively. The average abundance of MPs in the 0–60 cm soil layer of the cotton field subjected to 30 years of continuous mulching was  $78.51 \pm 2.57$   $n(100g)^{-1}$ . The comparative study between the abundance of MPs in soil and different film mulching durations indicated that the total amount of MPs in the cotton field soils increased significantly with increased mulching duration.



**Fig. 2** The abundance and distribution characteristics of MPs (pieces  $(100g)^{-1}$  soil) in agricultural fields.

#### 3.2.2 Microplastic composition

The  $\mu$ -FTIR spectrum was analyzed to establish the composition of MPs in the cotton field soils, and the spectrum is depicted in Figure 3. According to the spectral data, the absorption peaks at 719–722, 1004–1018, and 1536–1538  $cm^{-1}$  reflected the conformational structure of PE, while the saturated absorption peak at 1463–1466  $cm^{-1}$  corresponded to the torsional vibration of  $-CH_2$ . The saturated absorption peaks at 2850–2854 and 2918–2924  $cm^{-1}$  corresponded to the symmetric and antisymmetric stretching vibration of saturated- $CH_2$ , respectively. The spectral characteristic peaks of the obtained samples were compared with the standard spectral library to obtain the approximate value, the results showed that the composition of the microplastics matched that of the PE materials, indicating that these particles originated from the PE films in the field soils in the study area.

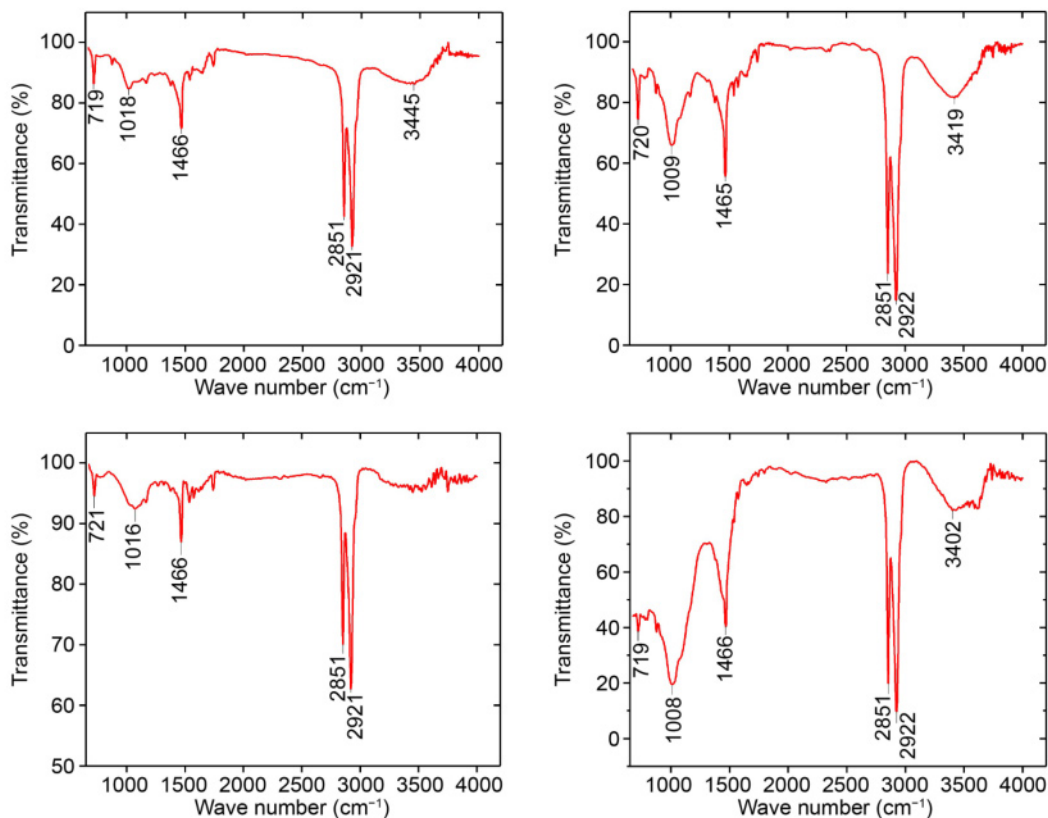
#### 3.2.3 Morphological characteristics of microplastics

The differences in the morphological composition of MPs were observed under different mulching durations. The visual microscopic observation showed the presence of a large number of microplastic particles with a size of  $<5$  mm in the soil, mainly exhibiting fragments, particles, and fibers. The microplastic fragments (Figure 4A, B) referring to MPs with particle sizes of  $>0.1$  mm had the most irregular and diverse range of shapes. The edges of the microplastic particles (Figure 4C, D) were relatively broken, referring to the MPs with a particle size of  $<0.10$  mm, and their aspect ratios were not less than 60%. The microplastic fibers (Figure 4E, F) referred to fine filamentous MPs with a soft texture, a width of  $<0.05$  mm, and a length of up to 2 mm. The fibers were mainly bent or twisted together, forming the fine filaments or ultrafine strips. The distribution of different shapes of MPs with varying mulching times is depicted in Figure S4. The number of microplastic fragments in the cotton field soil with prolonged film mulching was larger than other morphological types, followed by microplastic particles, while the microplastic fibers accounted for the smallest proportion.

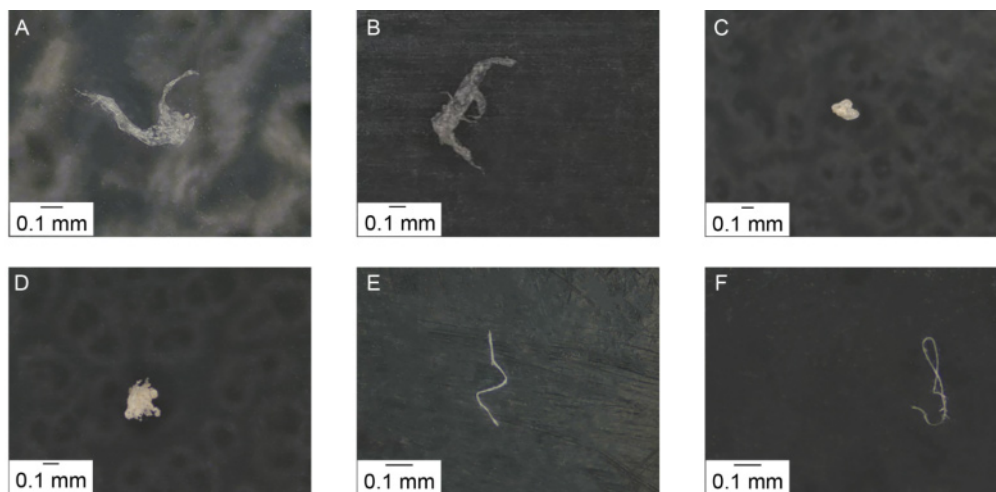
#### 3.2.4 Microplastic surface characteristics

In the soil environment, the mechanical, chemical, and biological processes affect the surface morphology of MPs, showing typical degradation due to gradual cracking of the surface.

Figure 5 indicates the primary microplastic characteristics. The shapes of the microplastic fragments (Figure 5A) were extremely irregular, and their particle sizes were  $>0.1$  mm. The surface of the MP fragments was relatively uniform, containing numerous pores (Figure 5B), which might have resulted from the weathering of film debris in the soils. The microplastic particles (Figure 5C) were ellipsoidal, with very



**Fig. 3** μ-FTIR spectrum analysis of microplastics in cotton field soil.

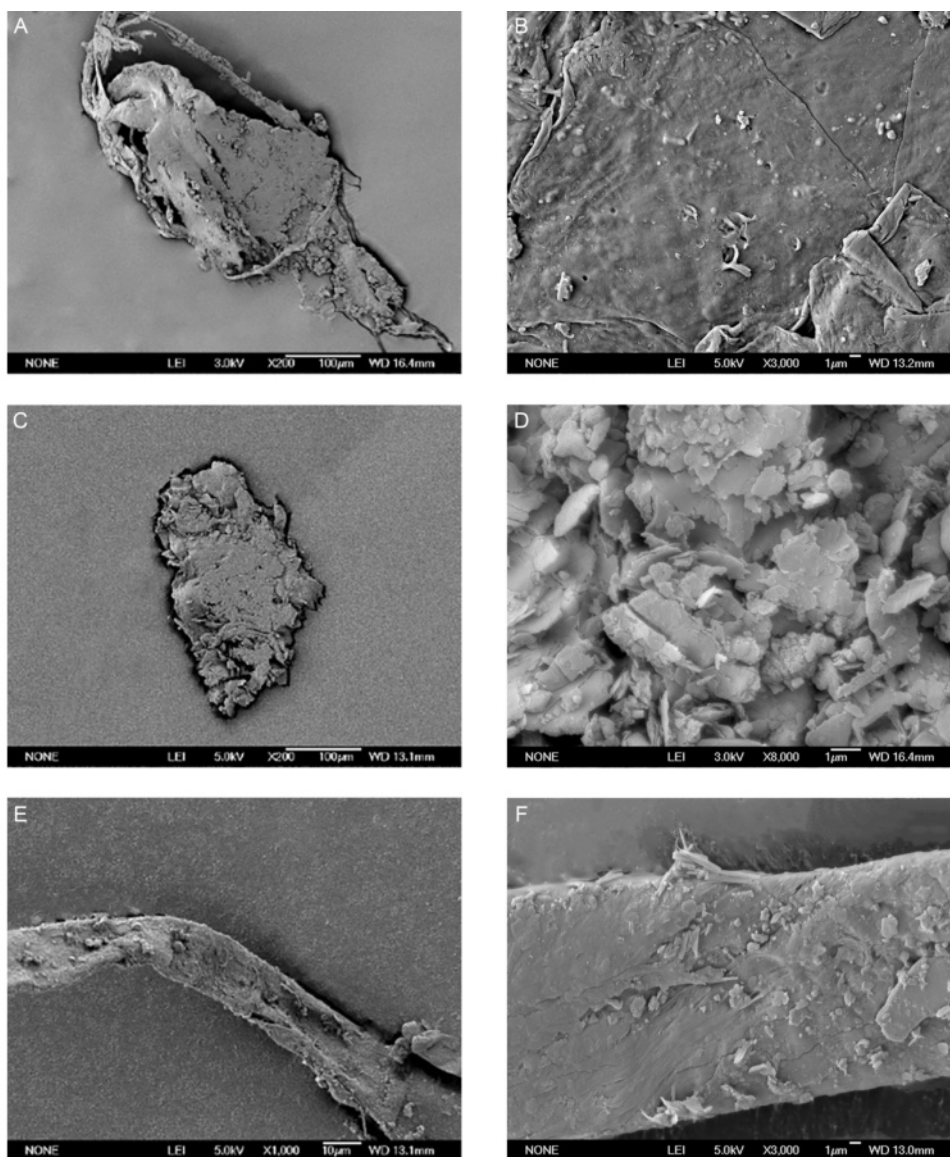


**Fig. 4** Stereomicroscopic image of soil MPs samples of different forms. (A, B) Fragment microplastics; (C, D) Particle microplastics; (E, F) Fiber microplastics.

small particle sizes and a higher level of damage to the particle edges. The surface of the MP particles was uneven, with some amorphous substances and debris attached (Figure 5D). These amorphous substances exhibited a wide range of structures and morphological characteristics due to continuous oxidation. The microplastic fibers (Figure 5E) were usually in the form of fine filaments or a bundled mass. Numerous fine filaments and cracks were observed on the

surface of MP fibers (Figure 5F), which might be related to the degradation of fibers, potentially increasing the absorption of various organic and inorganic pollutants from the soil. Additionally, their surface exhibited irregular fracture and torsion patterns, which might have happened due to the splitting of the mulching film by an external force during agricultural production.

The EDS data (see Supplementary information) detected



**Fig. 5** SEM image of MPs with different characteristics in soil. (A, B) Fragment microplastics, (C, D) Particle microplastics, (E, F) Fiber microplastics.

some silicon dioxide ( $\text{SiO}_2$ ) on the surface of MPs, which could affect the surface morphology and other properties. Additionally, some metal elements (Na, Al, and Pt) were adsorbed to the surface of MPs, changing the soil characteristics and further polluting the soil environment.

## 4 Discussion

### 4.1 Distribution characteristics of microplastic abundance

In Southeast Mexico, around  $2770 \text{ pieces kg}^{-1}$  MPs were detected in the traditional home garden soil layers (Huerta et al., 2017). Similarly, in Sydney, around  $300\text{--}67500 \text{ mg kg}^{-1}$  were detected in the industrial zone soil (Fuller et al., 2016). In this study, the average abundance of MPs at the soil

depths of 0–60 cm was investigated in the cotton fields subjected to 5–30 years of film mulching. The observed abundances ranged from  $(36.83\pm 3.24)\text{--}(78.51\pm 2.57) \text{ n (100g)}^{-1}$ , which were consistent with the soil MP abundance reported by Scheurer and Bigalke (2018) in a flood plain area of Switzerland ( $593 \text{ pieces kg}^{-1}$ ) and in the farmlands ( $306\pm 360 \text{ particle kg}^{-1}$ ) and grasslands ( $184\pm 266 \text{ particle kg}^{-1}$ ) of the Central Valley of Chile reported by Corradini et al. (2021). Since the NaCl solution with low density was used in this study, some high-density MPs might have lost during density separation, resulting in the miscalculation of MP abundance. However, the abundances observed in the present study were still higher than the 0–3 cm and 3–6 cm soil layers in the vegetable fields in suburban Shanghai (China), which were  $78.00\pm 12.91$  and  $62.50\pm 12.97 \text{ pieces kg}^{-1}$ , respectively (Liu et al., 2018). Therefore, it could be



inferred that the MP pollution of the farmlands in the Xinjiang region might be more severe than measured. As a global application to increase agricultural production, the greenhouse residues and plastic film could increase the abundance of MPs in the soil. Another report showed 0–5 cm, 5–10 cm, and 10–25 cm soil layers ( $1443 \pm 977$ ,  $1362 \pm 829$ , and  $1860 \pm 1212$  pieces  $\text{kg}^{-1}$ , respectively) in an agricultural facility in Shouguang (Shandong Province, China), which is the largest greenhouse vegetable production base in north China (Yu et al., 2021). Therefore, the plastic film might further decompose and degrade after entering the soil, thus increasing the MP pollution. In contrast, continuous mulching for 5, 10, 15, 20, 25, and 30 years resulted in the average abundances of  $36.83 \pm 3.24$ ,  $46 \pm 2.55$ ,  $53.34 \pm 1.97$ ,  $59.99 \pm 1.84$ ,  $66.66 \pm 0.61$ , and  $78.51 \pm 2.57$  n ( $100\text{g}^{-1}$  soil, respectively). The change of MP abundance at different film covering years was mainly affected by the physical, chemical, biological, and agricultural production activities, resulting in continuous fragmentation and decomposition of the film debris over time. However, the number was lower than the previous investigations of plastic film mulched land. For instance, the abundance of microplastics observed by PFM in the topsoil (0–10 cm) of plots in the Northeast long-term film mulching area is as high as 10586 and 7183 particles  $\text{kg}^{-1}$  (Li et al., 2022). Since the study area in the present study was open farmland located far away from factories, cities, or other obvious pollution sources, lower MP abundances were observed than those reported in the suburban farmland and facility farmland study areas. Additionally, different soil sampling depths, soil types, planting crops, and MP extraction methods might also attribute to the differences in the reported soil MP abundances.

#### 4.2 Downward migration of microplastics into subsoil layers

The present study shows that the morphological profile of MPs in soils varies in different regions. For instance, the reported distribution of MPs in Chile found the proportion of fibers to be 90% (Corradini et al., 2021). Similarly, in the riparian forest buffer zone of Dianchi Lake (Yunnan, China), the MP fibers were found to be dominant in soils, accounting for 92% of MPs (Zhang et al., 2018). This could be attributed to the long-term application of sludge and sewage irrigation in these areas. However, in the agricultural soils from the lower reaches of the Yangtze River and its estuary, the fragments were the main form of MPs, accounting for 49%, while the fibers accounted for 44% and the thin film fragments accounted for only 7% (Cao et al., 2021). In the Daliao River Basin soil, the total proportion of MP debris and thin film fragments exceeded 89%, while the proportions of foam, fibers, and particles were 6.75%, 2.15%, and 1.53%, respectively (Han et al., 2020). Among the soil MPs in suburban Wuhan, the debris, fibers, and other types accounted for 53%, 15.2%, and 31.8%, respectively (Zhou

et al., 2019). In the present study, the fragments were abundant in the farmlands subjected to continuous mulching for 30 years, accounting for 42.8% of soil MPs, followed by particles and fibers, accounting for 35.3% and 21.9%, respectively. Furthermore, the MP debris, thin film fragments, and fibers have been reported in the farmland soils in Southeastern Germany (Piehl et al., 2018). The major fragment MPs might be due to the widespread application of plastic mulching films during agricultural production. Additionally, the presence of MPs was detected in deeper soils of (300–400 mm (excluding 300), 400–500 mm (excluding 400), and 500–600 mm (excluding 500)) within the tillage layer. The morphological classification of MPs in different depth soil layers is summarized in Supplement Table 1. MPs at 0–400 mm depths existed in the form of fragments, fibers, and particles, while MPs at 400–600 mm depth were mainly in the form of fibers and particles, indicating that MPs in the cotton field soils are continually fragmented, splitting into smaller particle sizes. Cao et al. (2021) also determined MPs at 40–80 cm soil depths in the mulching farmlands in the lower reaches of the Yangtze River (China). The results suggest that the MPs are migrating from shallow soils to deeper soil layers. Therefore, investigating the distribution of MPs at higher abundances and finer particle sizes is highly necessitated to strengthen the current understanding of MP migration mechanisms in the farmland soils and the possible ecological hazard.

#### 4.3 Microplastic source analysis

Composition identification plays an important role in the determination of sources and MP-induced hazards. The  $\mu$ -FTIR spectrum showed that 95.72% of MPs in the cotton field soils from Xinjiang were composed of PE. These results were consistent with the characteristic peaks of PE reported in the previous studies (Liu et al., 2019; Zhang et al., 2019a). Moreover, PE is the main component of agricultural mulching films applied in the cotton fields in Xinjiang. PE has also been reported to be the main type of soil MPs in the vegetable fields of Wuhan (Liu et al., 2019) and Harbin suburbs (Zhang et al., 2020). Therefore, it can be speculated that a large number of MP particles in the farmland soils originate from the fragmentation and cracking of residual mulching film after prolonged film mulching application.

## 5 Conclusions

This study surveyed the distribution characteristics of plastics among the continuously mulching farmlands for 5–30 years in Xinjiang. The number and proportion of large plastic and microplastic fragments in the farmland soil increased significantly under continuous mulching treatment. The primary component of MPs in the farmland soils was

PE, while agricultural film mulching was the primary source of MPs in the absence of other pollution sources. The major morphological types of MPs were fragments, fibers, and particles. The MPs were mainly distributed in the 0–30 cm surface soil layers, although they were also found in deeper soil layers, indicating that MPs with smaller particle sizes might continuously migrate, spreading to deeper soil layers. Overall, this study demonstrated important data of the farmland plastics distributing characteristics and explored some important MP pollution sources with different coverage years in typical farmland covered for a long time in Xinjiang, China. This study will provide a theoretical basis for global farmland film mulching pollution and MP prevention and control. However, it is necessary to improve pollution control strategies for farmland mulching films and develop new residual film recovery machinery and degradable plastic film technologies to reduce the possibility of soil MP formation from the source.

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## Electronic supplementary material

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