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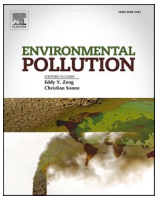

Macro- and microplastic accumulation in soil after 32 years of plastic film mulching☆

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ABSTRACT

Plastic film mulch (PFM) is a double-edged-sword agricultural technology, which greatly improves global agricultural production but can also cause severe plastic pollution of the environment. Here, we characterized and quantified the amount of macro- and micro-plastics accumulated after 32 years of continuous plastic mulch film use in an agricultural field. An interactive field trial was established in 1987, where the effect of plastic mulching and N fertilization on maize yield was investigated. We assessed the abundance and type of macroplastics (>5 mm) at 0–20 cm soil depth and microplastic (<5 mm) at 0–100 cm depth. In the PFM plot, we found about 10 times more macroplastic particles in the fertilized plots than in the non-fertilized plots (6796 vs 653 pieces/m^2), and the amount of film microplastics was about twice as abundant in the fertilized plots than in the non-fertilized plots (3.7×10^6 vs 2.2×10^6 particles/kg soil). These differences can be explained by entanglement of plastics with plant roots and stems, which made it more difficult to remove plastic film after harvest. Macroplastics consisted mainly of films, while microplastics consisted of films, fibers, and granules, with the films being identified as polyethylene originating from the plastic mulch films. Plastic mulch films contributed 33%–56% to the total microplastics in 0–100 cm depth. The total number of microplastics in the topsoil (0–10 cm) ranged as 7183–10,586 particles/kg, with an average of 8885 particles/kg. In the deep subsoil (80–100 cm) the plastic concentration ranged as 2268–3529 particles/kg, with an average of 2899 particles/kg. Long-term use of plastic mulch films caused considerable pollution of not only surface, but also subsurface soil. Migration of plastic to deeper soil layers makes removal and remediation more difficult, implying that the plastic pollution legacy will remain in soil for centuries.

1. Introduction

Plastic film mulching had been called the “white revolution”, due to its positive effects on yield and the white visual appearance of transparent plastic mulch films. However, the perception of plastic mulch film is now changing to one of “white pollution” due to the progressive build-up on plastic waste in many soils (Liu et al., 2014). Plastic mulch films (PFM), generally made of polyethylene, have been used for agricultural production since 1956, and their global market has grown continually, reaching about 2×10^6 tons in 2017 (Moine and Ferry, 2019).

Plastic mulch allows crops to make full use of limited light, heat, water, or nutrients, resulting in early maturity, increased yields, and improved crop quality (Gao et al., 2019; Li et al., 2000; Wang et al., 2021). The invention of plastic film mulching has increased global grain crop and cash crop yields by 20%–35% and 20%–60%, respectively (Liu et al., 2014; Qin et al., 2014; Sun et al., 2020). However, the increasing use of agricultural plastic films has also led to pollution of farmland with plastic residues (Zhang et al., 2016). Accumulation of plastic can negatively impact soil health, crop growth, food safety, as well as

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impacting on the wider environment when transferred to watercourses (Zeng et al., 2013).

Although agricultural plastic mulch films are generally removed from the fields after crop harvest, complete removal is often impossible. Plastic films tend to become brittle and tear apart when they are removed from the fields, leaving behind plastic residues. Repeated use of plastic mulch films has led to substantial accumulation of macro- and microplastic debris in soils, particularly in China (Huang et al., 2020; Liu et al., 2018; Zhang et al., 2016), where plastic mulch films are thinner (8 μm) than those used in the US (50 μm) and therefore more readily disintegrate during removal from the fields.

Plastic mulch films buried in soil at the edges of planting rows can become entangled with roots (Zhao et al., 2017), and it can thus be expected that enhanced root growth will make it more difficult to completely remove plastic films after harvest. Root growth can be promoted through fertilization (Durieux et al., 1994), and therefore, fertilization can potentially affect the abundance of plastic residues in soil under plastic film mulching. Further, fertilization can change the activity and structure of the soil microbial community (Zhong et al., 2015), which may impact the degradation of plastic films and thereby influence the abundance of plastic residues. However, to our knowledge, no studies have investigated whether N fertilization impacts the accumulation of plastic residues in the field.

Plastic residues in soils can be categorized into macroplastics (>5 mm) and microplastics (<5 mm) (Frias and Nash, 2019; Galloway et al., 2017). While macroplastics frequently remain on or near the soil surface, microplastics can be translocated vertically in soil through leaching and bioturbation (Bläsing and Amelung, 2018; Rillig, 2012; Yu and Flury, 2021a). There have been numerous studies on the transport of microplastics in porous media, but field studies to assess the long-term migration of microplastics in terrestrial environments are lacking (Yu and Flury, 2021a). Microplastic particles have been observed up to 80-cm depth in an agricultural soil at a site with 50 mm annual precipitation in Xinjiang Province, China (Hu et al., 2021). It is thus likely that plastics can also migrate deep into soil in regions where the annual precipitation is larger than in Xinjiang.

Our goal in this paper was to quantify the amount of plastic accumulated in soil after long-term, continuous use of plastic film mulching. A long-term experiment was established in 1987 to evaluate the effects of plastic film mulching and N fertilization on maize yield and quality in Shenyang, China. In spring 2019, we evaluated the abundance of macro- and microplastics (>5 mm and <5 mm, respectively) in the soil, and macroplastic was once more investigated in 2020. We hypothesized that (1) the abundance of macro- and micro-plastics derived from long-term used plastic film mulch is larger in plots that have received more N fertilization, because root entanglement will result in lower removal rates of plastic films after harvest, and (2) plastic film-derived microplastics can migrate down to the depth of 100 cm.

2. Materials and methods

2.1. Site description and experimental design

The site was a long-term plastic film mulching and fertilization experiment (41°49′ N, 123°34′ E), which was established in 1987 at Shenyang Agricultural University, China. The site has a temperate continental monsoon climate, and is hot and rainy in summer but cold and dry in winter, with an average annual temperature of 7.2 °C and average annual precipitation of 730 mm. The soil is classified as brown earth according to the Chinese Soil Taxonomy (Haplic-Udic Alfisol according to US Soil Taxonomy), developed from loess-like parent material. Conventional tillage was used to a 20 cm depth and maize (*Zea mays* L.) was continuously planted as a monoculture for the past 50 years.

The experiment was a split-plot design with or without plastic film mulching as main plots and chemical N fertilization treatments as subplots. The experimental design had three replicates for the with and without PFM treatments. In each main plot, subplots included no fertilization (N_0) and 135 kg N hm^{-2} $year^{-1}$ (N_{135}), with an area of 69 m^2 for each plot. After sowing and fertilization (urea), the whole surface (all ridges and furrows) of the plot was mulched by polyethylene film (colorless transparent, 8 μm thick, 1.2 m wide rolls). The edges of the mulch (ca. 10 cm) were covered by soil to fix the mulch in place. After the maize germinated, a handheld device was used to drill holes about 1 cm in diameter into the plastic to allow plants to emerge. The maize was harvested at the end of September by cutting the stalks above ground. Plastic films were manually retrieved by turning up the edge of the plastic mulch after loosening the soil with small rakes and removing the film from the field. A more detailed description of the split-plot experiment is given in Ding et al. (2019).

2.2. Macroplastic collection and quantification

In spring 2019 (before seeding), the amount of macroplastic debris was determined by a quadrat survey in the individual PFM plots. In each replicated plot, a block of 0.5 m × 0.5 m was randomly selected and the soil from the 0–20 cm horizon in the block was dug out and sieved through a 5-mm metal mesh. Such block sampling has been shown to be the most effective for plastic sampling in terrestrial systems (Yu and Flury, 2021b). All the visible plastic residues were picked up and collected. Soil and roots attached to plastic residues were carefully removed to avoid breaking up the plastic residues. The surface area of individual plastic residues was measured with an area meter with grids (1 cm × 1 cm). The areas were classified into three categories of <4 cm^2, 4–25 cm^2 and >25 cm^2 and the number of plastic particles in each category were counted. Then, the residues were cleaned by ultrasonication and air-dried. The mass of macroplastics was then determined with an electronic balance with an accuracy of 0.0001 g. To verify the accuracy of the results, the amount of macroplastics was once more determined with the same procedure in autumn 2020 (after harvest).

2.3. Microplastic extraction and counting

In the spring of 2019 (before seeding), one soil core was taken from a random location in each plot with a soil coring drill (diameter: 8.5 cm) in depth increments of 0–10 cm, 10–20 cm, 20–40 cm, 40–60 cm, 60–80 cm, and 80–100 cm. Soil samples were passed through a 5-mm metal mesh to remove macroplastic residues and roots, and then air-dried. The bulk density of the soil in each depth increment was determined from the measured dry weight and the volume of soil core increment. Microplastic extraction in soil followed the method of Liu et al. (2018) with slight modification. Specifically, 20 g of air-dried soil was transferred into a 250-mL glass beaker, and 200 mL of 6.24 mol/L $ZnCl_2$ with a density of 1.6 g/cm^3 added. The beaker was then sonicated using an ultrasonic cleaning machine (SB-800 DTD, SCIENTZ; 60 Hz, 20 min) to break up soil aggregates, and then automatically stirred for 10 min. The beaker was then left to stand for 24 h to allow particles to separate by density (low-density polyethylene has a density of 0.91–0.94 g/cm^3 and float to the surface). The supernatant was passed through a membrane filter (blended cellulose, 0.45 μm, Solarbio), and the materials on the filter were transferred to 200 mL of 30% H_2O_2 solution, and heated at 60 °C to remove soil organic matter (Wang et al., 2018). Afterwards, the solution was again filtered through a membrane filter (pore size: 0.45 μm), and the membrane filter was transferred to a 60 mm diameter Petri dish and dried at 60 °C (Weithmann et al., 2018; Wu et al., 2020). The extraction method was reported to have a recovery rate of 90% for plastic particles (1–5 mm) (Liu et al., 2018). A blank control without soil was set up to check whether any plastic contamination occurred during the extraction process.

The microplastics on the filter membrane were observed under a stereo microscope (ZEISS, Stemi, 2000-C) with 50× magnification. Ten fields of view were randomly selected for each membrane under the microscope. For each field of view, the shape, size and color of each

microplastic and the total amounts were recorded. We classified the shape into three categories: films, fibers, and granules. Film-shaped microplastics generally appeared shiny under light, with rounded edges. Fiber-shaped microplastics were defined as curled glossy filaments. Granular microplastics were defined as glossy particles that appeared roughly spherical. The size of microplastics was recorded as its longest side.

2.4. Testing plastic film removal and recovery in the field

Plastic mulch films were always removed from the fields after each growing season; however, complete removal was not possible. Plastic mulch films were removed by manually collecting all exposed, visible plastics on the soil surface and by pulling out the buried plastic films at the sides of the ridges after loosening up the soil with a rake. To test whether there is a difference in removal and recovery of plastic mulch films between the plots treated with different N fertilization, we measured the recovery rate of plastic mulch film removal in autumn 2020. We recovered the plastic films from a 2 m long strip in the field by the same method that was used in the long-term field experiment as described above. The collected plastic films were washed, air-dried, and weighed as described above. The recovery rate was calculated by dividing the weight of collected plastic film by the weight of the original, new plastic film.

2.5. μ-FTIR and SEM measurements

We selected typical specimens of the three types of microplastics under the microscope and stored them on a clean, smooth sheet of tin foil. Seven films, seventeen fibers, three granules were randomly selected for μ-FTIR. The plastic particles were then analyzed by μ-FTIR (PerkinElmer Spotlight 400) under transmittance mode, with a MCT detector. The spectral range was set to 4000–750 cm^{-1} with a resolution of 4.00 cm^{-1} and scanning was performed 16 times. The grating value was 600, while the grating size and number of accumulations varied with the size of the sample. The measured spectra were compared with a spectral library (Sadtler Infrared spectrum database) to identify the type of plastic polymers (with a confidence match of >60%). We also analyzed the spectra of the initial plastic film by FTIR.

The collected macroplastics, three of each microplastic shape (film, fiber, and granule) microplastics, and the recovered plastic mulch in recovery measurement were examined for surface morphology with a field emission scanning electron microscopy (SEM, Hitachi, Regulus 8100, Japan), and further analyzed for elemental composition on the sample surface by coupled energy dispersive spectrometry (EDAX, 15 keV, working distance 13.8 mm; Ametek Inc, Berwyn, PA, USA). Before SEM analysis, the plastic mulch was cleaned by ultrasonication to remove impurities on the surface, and then sputter-coated with gold.

2.6. Quality assurance and quality control

When collecting samples in the field, we chose cotton bags to store soil samples to avoid the risk of plastic contamination. To avoid microplastic contamination in the laboratory, we took several precautions. All materials used in the extraction experiments were made of glass or metal and were washed with pure water three times before use. Glassware was always covered with aluminum foil. Solutions were filtered through a membrane filter (blended cellulose, 0.45 μm, Solarbio) before use. Cotton labcoats were always worn during the laboratory experiments. Blank control samples (three replicates) without soil were run concurrently with the soil samples to check for potential plastic contamination during the extraction process. No plastics were detected in these blank control samples.

To test the efficiency of microplastic extraction method, we conducted a recovery test. We added previously extracted microplastics (30 films and 30 fibers) to 20 g microplastic-free soil samples with three replicates, and used the extraction protocol described above to extract microplastics. The recovery rates were 87% ± 3% for film microplastic and 90% ± 7% for fiber microplastic.

2.7. Calculations and statistical analysis

The average macroplastic weight per piece was calculated as follows:

$$M = \left(\sum_{i=1}^{n} W_i\right) \Big/ n$$

Where M is the average mass of a plastic piece of residual film (mg/piece); n is the total number of residual film pieces in a given plot, W_i (mg) is the weight of the ith piece of residual film. A smaller M indicates a higher degree of fragmentation (Yan, 2015).

The macro- and microplastic data and the mulch recovery rates were statistically analyzed using a one-way ANOVA (SPSS 20.0 software, Chicago, IL). The significance level was set as $\alpha = 0.05$. The infrared spectra of the μ-FTIR analysis were processed using the Omnic® software package (Thermo Fisher Scientific Inc, Waltham, MA).

3. Results

3.1. Abundance of macroplastics in mulched plots

In the plastic mulched plastic plots, the plots with N fertilization (N_{135}-with PFM) had a higher abundance of macroplastics compared to the non-fertilized plots (N_0-with PFM) ($P < 0.001$; Fig. 1A, and $P < 0.001$; Fig. 1B). The total amount of macroplastic pieces in the topsoil (0–20 cm) of the N_{135}-with PFM (6796 ± 1070 pieces/m^2) was about 10 times higher than in the N_0-with PFM (653 ± 245 pieces/m^2). The weight of macroplastics in the N_{135}-with PFM (36.0 ± 3.7 g/m^2) was about 6 times higher than in the N_0-with PFM plot (6.4 ± 3.3 g/m^2). Accordingly, the weight per piece of macroplastic was smaller in the N_{135}-with PFM plot ($P = 0.024$; Fig. 1C), suggesting that more fragmentation had occurred. The majority of the macroplastic pieces (75%–83%) were <4 cm^2 in area (Fig. 1D). Corresponding with the extent of fragmentation, the fertilized plots (N_{135}-with PFM) had a larger proportion of plastics <4 cm^2 in comparison to the non-fertilized plots (N_0-with PFM) (Fig. 1D).

3.2. Abundance, type, and vertical distribution of microplastics

The abundance of total microplastics decreased with soil depth (Fig. 2A). Generally, the PFM plots generally had greater amounts of microplastic than the plots without PFM in all the soil layers, for total amount (Fig. 2A) and film plastics (Fig. 2B). The total microplastic concentration in the 0–10 cm depth was 10,586 ± 3560 particles/kg (mean and standard deviation) for the N_{135}-with PFM plot and 7183 ± 1633 particles/kg for the N_0-with PFM treatment.

The amount of film-derived microplastics was larger in the N_{135}-with PFM (8318 ± 2024 particles/kg) than in the N_0-with PFM (4033 ± 472 particles/kg) in the 0–10 cm layer ($P < 0.05$; Fig. 2B). In contrast, the amounts of fiber microplastics in all soil layers were smaller under N_{135}-with PFM than under N_0-with PFM (Fig. 2C), but the difference was not significant ($P = 0.29$). Compared to film and fiber plastics, the amount of granular microplastics was much smaller, and no granular microplastics were found below 60-cm depth (Fig. 2D). Generally, fiber plastic dominated in the three types of microplastics in the 100 cm soil profile, except for N_{135}-with PFM (Fig. 2E). The total amounts of microplastic in the entire 100 cm soil profile were 7.4×10^6, 6.6×10^6, 2.6×10^6, and 2.5×10^6 particles/m^2, for N_0-with PFM, N_{135}-with PFM, N_0-without PFM, and N_{135}-without PFM plots, respectively. The amount of total microplastics in the PFM plots was about 2.8 times of that in the without PFM plots (Fig. 2E). In PFM plot, the proportion of film microplastics in total microplastics was 33% for N_0-with PFM and 56% for N_{135}-with

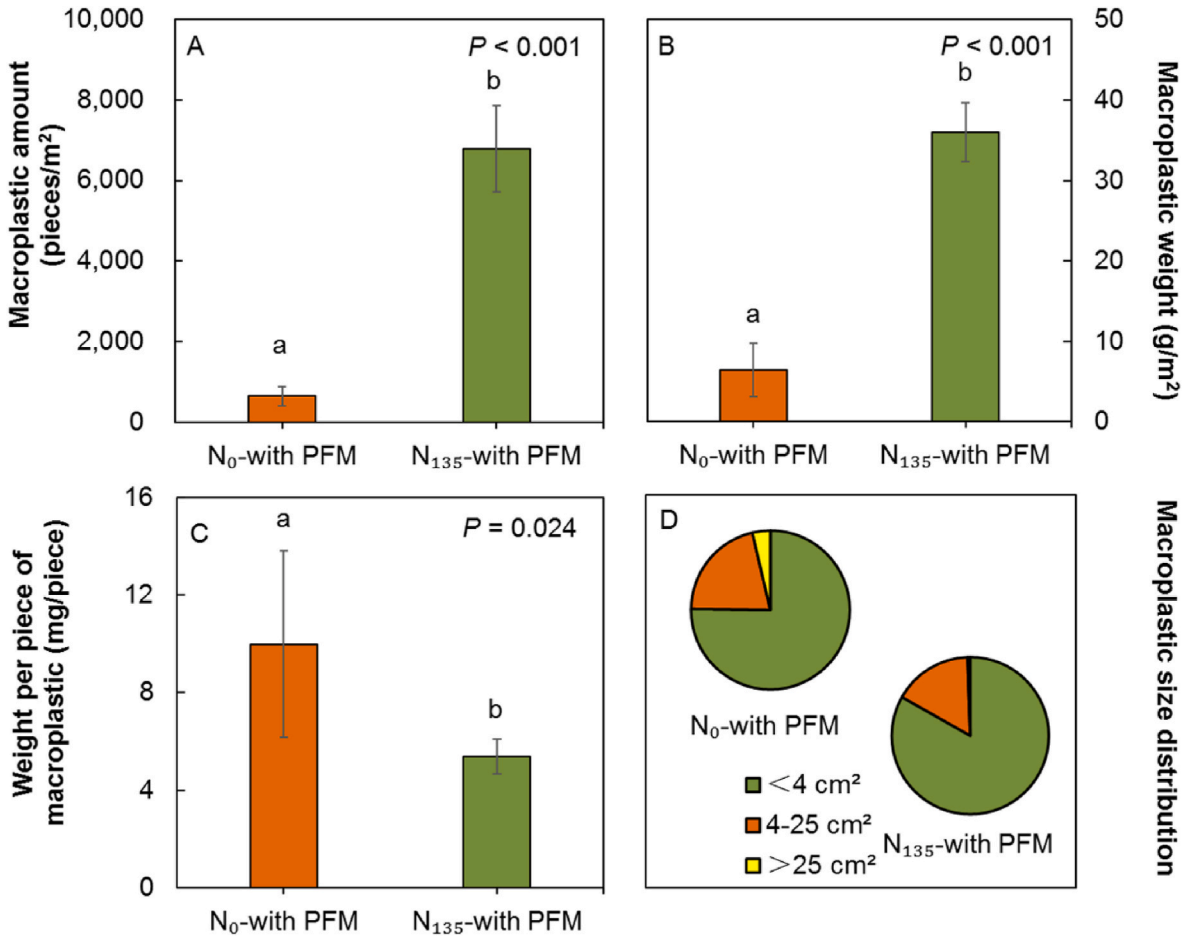

Fig. 1. Influence of N fertilization on the amount and size distribution of macroplastics in the topsoil (0–20 cm) of a long-term plastic film mulching (PFM) experiment under maize cultivation. A: macroplastic amount; B: macroplastic weight; C: weight per piece of macroplastic; D: relative proportion of three sized macroplastics (<4 cm^2, 4–25 cm^2, >25 cm^2). N_0-with PFM: no N fertilizer under plastic film mulching; N_{135} -with PFM: N fertilizer applied with 135 kg N ha^{-1} yr^{-1} under plastic film mulching. N fertilizer applied with 135 kg N ha^{-1} yr^{-1}. The macroplastic data include samples collected in 2019 and 2020. Error bars represent standard deviations ($n = 6$). Different letters indicate statistical significance at $P < 0.05$.

PFM (Fig. 2E).

3.3. Types of microplastics identified with μ-FTIR

Different kinds of colored microplastics were detected, including black, brown, yellow, blue, and transparent. Film and granular plastics were all transparent, and fiber plastic included colored and transparent ones. Film plastics were identified as polyethylene and its copolymers, including low-density and linear polyethylene, which had the same characteristic peaks as the original polyethylene plastic mulch film (Fig. 3), confirming that the film microplastics were likely derived from the plastic mulch film. The fibrous microplastics were identified as rayon, polyester terephthalic acid (Kershaw et al., 2011), polypropylene, and poly (ethylene terephthalate), and polyester terephthalic acid. The granular microplastics mainly consisted of poly (N-methyl acrylamide), although the match of the spectrum was not overly good (66% match, Fig. 3I).

3.4. Surface morphology of microplastics

Film plastics showed pronounced surface erosion characteristics, and the surface morphology was characterized by grooves, pits, and holes (Fig. 4A). The surface of the holes and outer edges show clear evidence of weathering. The surface of fiber microplastics, on the other hand, was relatively smooth, and the degree of weathering was weak, with slight scaling-type abrasion (Fig. 4B). The surface of granular microplastics showed uneven protrusion and pits of different degrees. More types of elements were observed on the granular surfaces (C, N, O, Na, Mg, Al, Si, S, Cl, Ca) than on films (C, N, O, Al, Si) and fibers (C, N, O, Al, Si).

4. Discussion

4.1. Relative abundance of macro- and microplastics in soil

The N_{135}-with PFM and N_0-with PFM contained 36.0 g/m^2 and 6.4 g/m^2 macroplastic residues (Fig. 1B), respectively. These amounts belong to the categories of heavy (>7.5 g/m^2) and high pollution (5–7.5 g/m^2), respectively, according to China's national standard of "Limits and Determination of Residue of Mulching Film in Farmland" (GB/T 25,413–2010).

The abundance of macroplastics in other studies with shorter durations of plastic mulch film applications were generally lower than that of our fertilized plot. The abundance of film-derived macroplastic residues was 0.24–0.82 g/m^2 after 5 years of plastic film mulch use in Harbin, Baoding, and Handan, China (Xu et al., 2006). In Xinjiang Province, China, residual plastic film were reported to be 3.8 g/m^2 (≤5 years of plastic film mulching), 10.1 g/m^2 (5–10 years), 12.4 g/m^2 (10–20 years), and 19.0 g/m^2 (>20 years) in the top 0–30 cm of the soil (Zhang et al., 2016). Another study conducted in Xinjiang province reported that the macroplastic amount ranged from 12.2 to 35.2 g/m^2 over 5–19 years of plastic film mulching (He et al., 2018). Thus, the macroplastic concentrations in our field site are similar to those of other field sites in

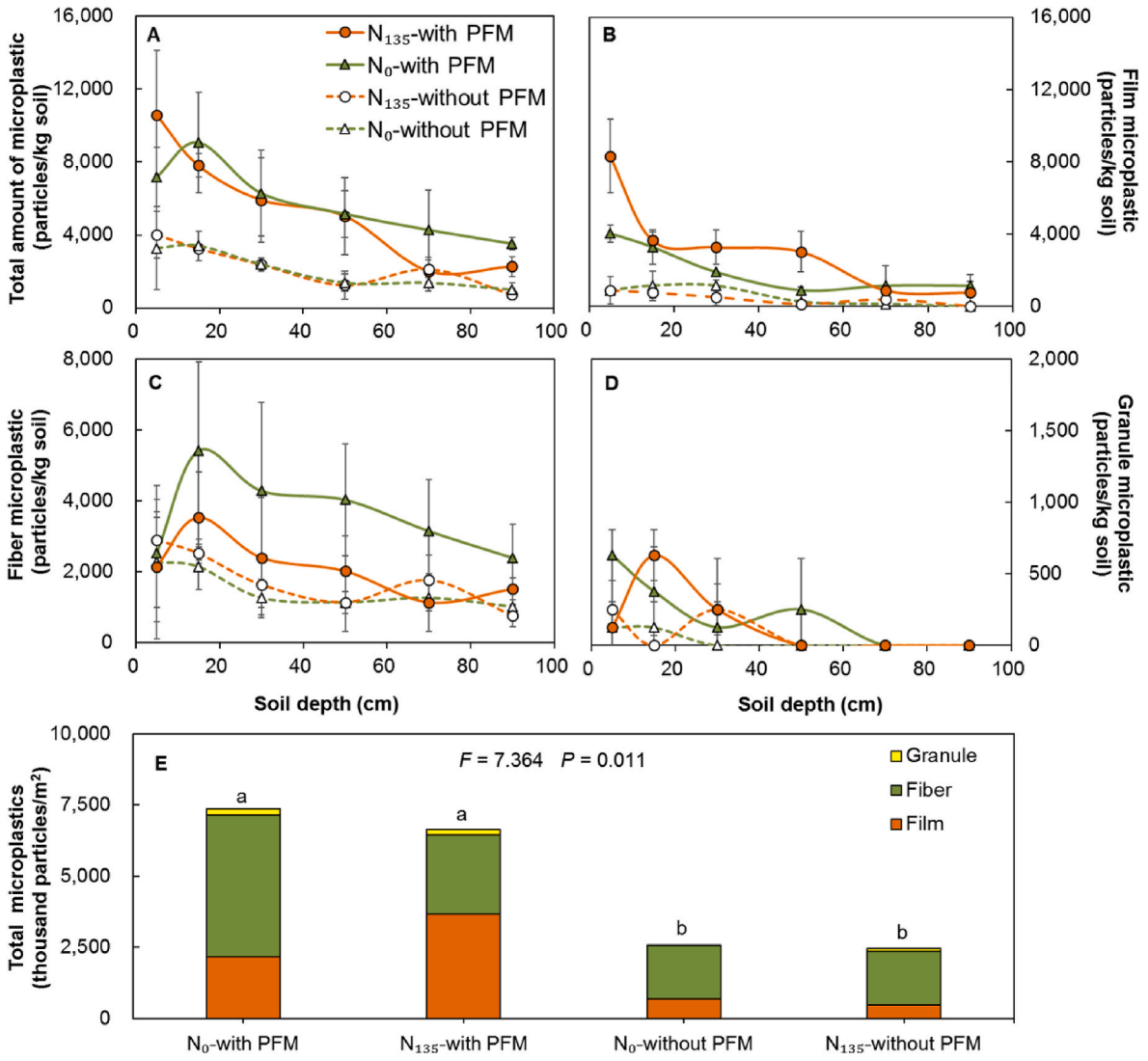

Fig. 2. Influence of N fertilization on the amount, type and depth distribution of microplastics in the soil profile (0–100 cm) of a long-term plastic film mulching experiment under maize cultivation. A: total amount of microplastic; B: films; C: fibers; D: granules; E: cumulative amount of films, fibers, and granules microplastics in the 0–100 cm profile. N_0-with PFM: no N fertilizer under plastic film mulching; N_{135}-with PFM: N fertilizer applied with 135 kg N ha^{-1} yr^{-1} under plastic film mulching; N_0-without PFM: no N fertilizer; N_{135}-without PFM: N fertilizer applied with 135 kg N ha^{-1} yr^{-1}. Error bars represent standard deviations ($n = 3$). Different letters in Figure E indicate significant difference in total microplastic abundance among the treatments.

China where plastic mulch film has been used for a comparable amount of time.

At our experimental site, the average abundance of microplastics in the surface soil (0–10 cm) was 10,586 particles/kg and 7183 particles/kg in the N_{135}-with PFM and N_0-with PFM, respectively (Fig. 2A). The number was larger than or comparable to most previous investigations of plastic film mulched land. In Southeast Spain, the abundance of microplastics in 0–10 cm depth was 2116 particles/kg in vegetable fields mulched with plastic film for 10 years (Beriot et al., 2021). Similarly, a study of horticultural soils in Tunisia also found microplastic concentrations of 476 particles/kg after several years of mulch films use (Boughattas et al., 2021). In Xinjiang province of China, the abundance of microplastics in cotton fields was 80, 308, and 1076 particles/kg, after continuous plastic film mulching for 5, 15, and 24 years, respectively (Huang et al., 2020). A large-scale investigation across Yunnan Province of China showed an average microplastic abundance of 9800 particles/kg for 100 sampling sites with plastic film mulching (Huang et al., 2021).

Lower abundance of plastics would be expected when mulch films are thicker and therefore less likely disintegrate during removal after harvest (Zhang et al., 2016). Indeed, in Europe, where mulch films are generally ticker than in China (25–50 μm vs 8 μm), lower plastic concentrations have been reported for comparably long duration of plastic mulch use (Beriot et al., 2021).

4.2. Impact of N fertilization on macro- and microplastics presence in soil

Long-term N fertilization aggravated the amount and weight of macroplastic (Fig. 1A and B) and the amount of plastic film-derived microplastic (Fig. 2B) in the soil profile in PFM plots. The recovery rate of plastic film after crop harvest from the N_0-with PFM plots (92.2% ± 3.4%) was indeed higher than those from the N_{135}-with PFM plots (89.7% ± 1.8%), although the difference was not significant ($P = 0.412$, Figure S1). Nonetheless, the small difference can lead, over 32 years of accumulation, to a tenfold and twofold difference in the amount of macroplastic (Fig. 1A) and film microplastic (Fig. 2B), respectively, between N_{135}-with PFM and N_0-with PFM. The difference in recovery rates may be explained by enhanced root growth under N fertilization (Ding et al., 2021), which will increase the entanglement of plastic film and roots and impede the removal of plastic films after harvest. In

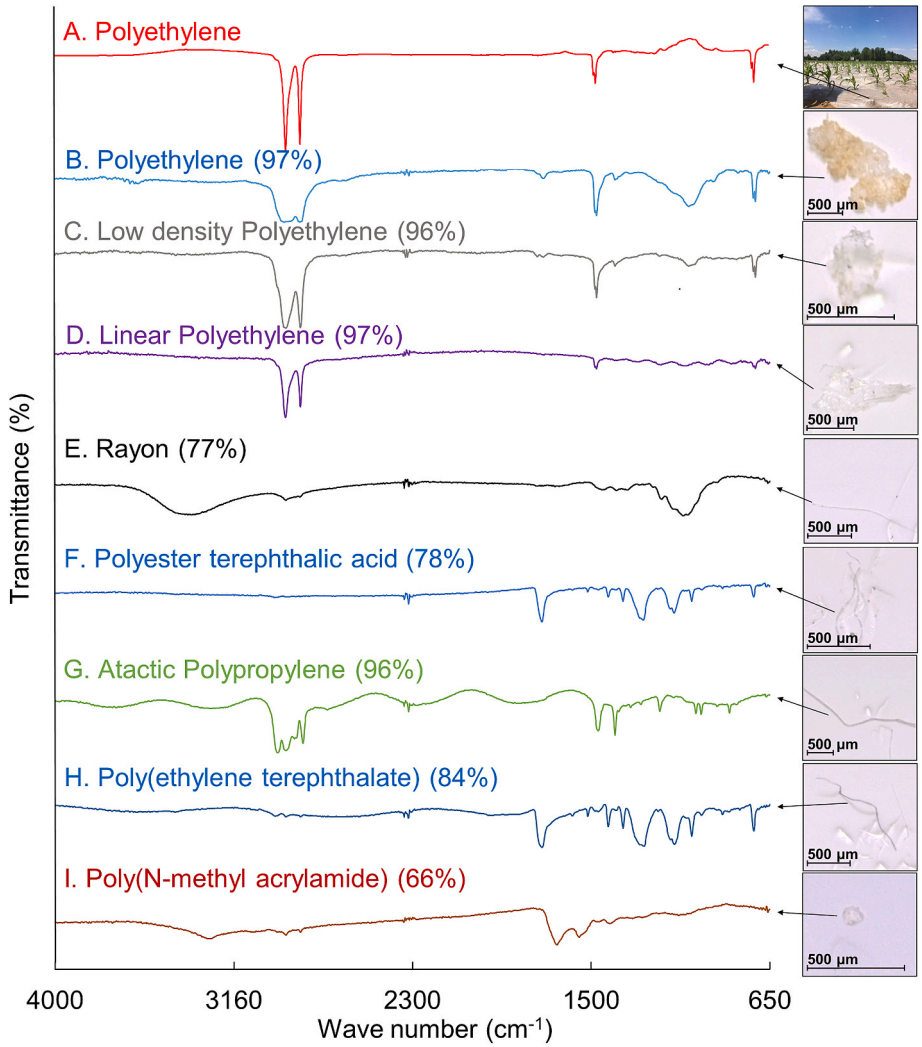

Fig. 3. FTIR spectra and microscopy images of microplastics from long-term plastic mulching experiment. A: original polyethylene plastic mulch film; B, C, and D: film microplastic; E, F, G, and H: fiber microplastic; I: granular microplastic. The numbers in % indicate the match of the measured spectra compared to the FTIR spectral library.

addition, N fertilization may impact polyethylene degradation indirectly through affecting soil microorganisms. In a laboratory incubation experiment, it was observed that N addition increased the biodiversity and abundance of several dominant genera of soil microorganisms capable of degrading low-density polyethylene, thereby accelerating the fragmentation of polyethylene particles from large to small particle sizes (Zhang et al., 2020). This is consistent with our results that the plastic film fragmentation was stronger in the N_{135}-with PFM than in the N_0-with PFM (Fig. 1C).

Fertilizer can also be a source of microplastics. However, there was no difference in total microplastic concentrations between non-fertilized and fertilized non-mulched plots (Fig. 2E), indicating that there was no significant input of microplastics through fertilizer application.

4.3. Sources of microplastic pollution in soil

Microplastics in long-term plastic film mulched soil come from a wide range of sources. Film microplastic is derived from plastic film, i.e., polyethylene (Fig. 3). This is also supported by Huang et al. (2020) who investigated microplastics in agricultural soils in Xinjiang, China. In our experiments, the proportion of film microplastics compared to total microplastics in PFM plots (Fig. 2E) was 33%–56%. A recent study reported the contribution of mulch film to microplastics to be 10%–30%, based on 69 soil samples from 19 provinces in China (Ren et al., 2021). The smaller contribution reported in Ren et al. (2021) was probably due to the shorter duration of plastic mulch film mulching for their sampling sites.

Compared with the PFM plots, there was a substantial amount of plastic also found in the non-PFM plots, mainly consisting of fibers. This suggests that fibers were most likely deposited by atmospheric deposition (Dris et al., 2016). The presence of film microplastic in the non-PFM plots, however, suggests that there was cross-contamination from the PFM plots, likely caused by wind. The exogenous macroplastics in the non-PFM plots likely deteriorate into microplastics, contributing to an elevated film microplastic concentration. Fiber microplastics, which were made up of rayon (Fig. 3E), polyester terephthalic acid (Fig. 3F) and poly (ethylene terephthalate) (Fig. 3H), likely originated from textiles, which were deposited through atmospheric fall out. Granular microplastics were mainly composed of poly (N-methyl acrylamide, Fig. 3I), which can come from pesticide formulations, binders in seed coating agent, or flocculants in paper (Exon, 2006).

4.4. Microplastic translocation in soil

We observed plastic film-derived microplastics in deeper soil depths. The top 20 cm of the soil were mixed by tillage, so the plastic

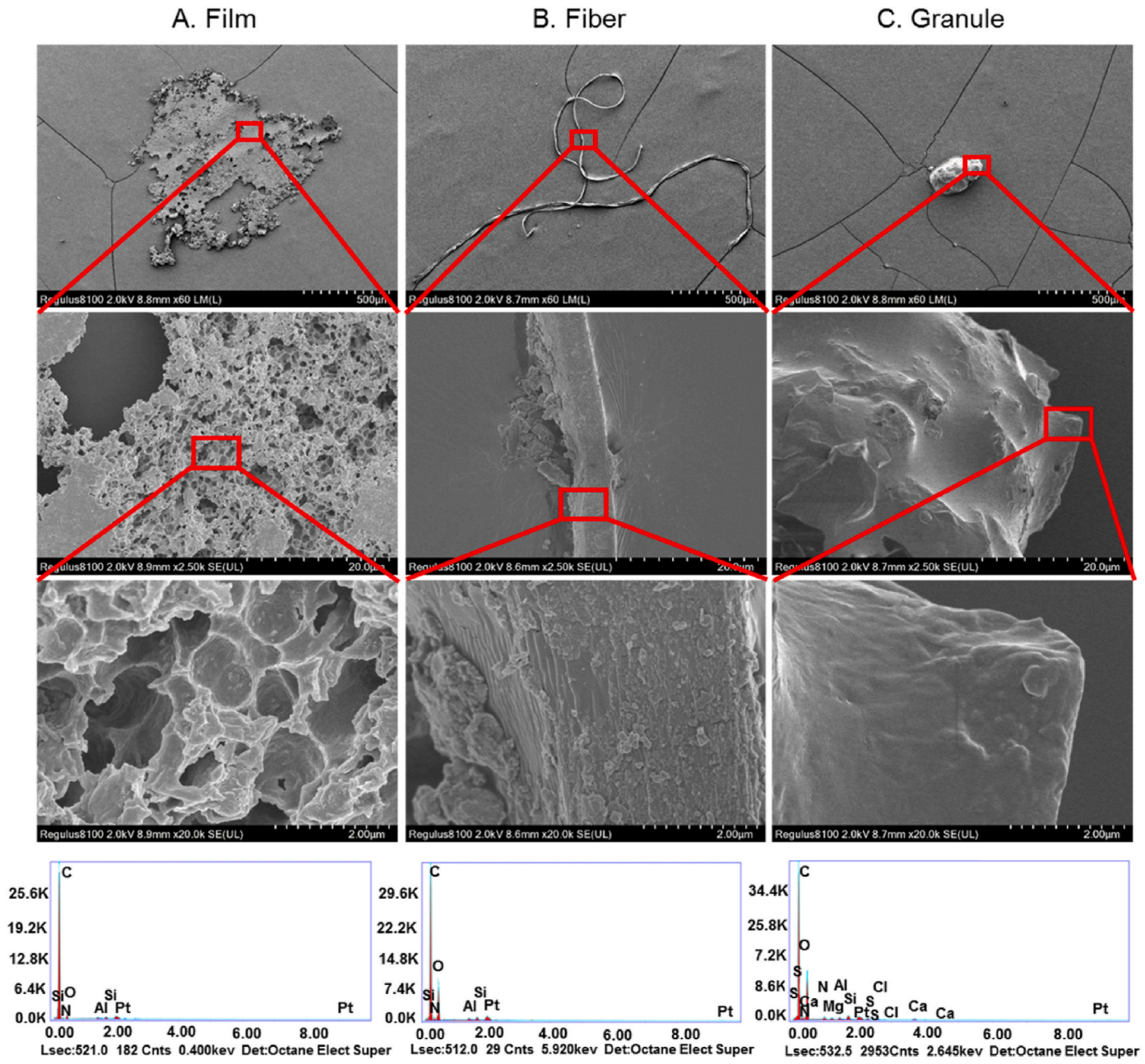

Fig. 4. Scanning electron microscopy images of microplastics. A: films, B: fibers, and C: granules at three different magnifications. Elemental spectra are energy-dispersive X-ray spectra.

concentrations will have been homogenized in this layer. Nonetheless, we observed substantial amount of microplastics below the plow layer. The total number of microplastics was 2268–3529 particles/kg soil in the 80–100 cm soil layer (Fig. 2A) and film microplastics amounted to 436 particles/kg in the PFM plots (Fig. 2B), indicating that microplastics (including plastic film-derived microplastic) can migrate down to at least 100 cm depth. A recent investigation in Xinjiang reported microplastic concentrations of 112 particles/kg at 40–80 cm depth after continuous film mulching for 10 years, but microplastics only included fibers and granules and no film microplastic was found at this depth (Hu et al., 2021). They explained that film microplastics had a larger particle size and were therefore less susceptible to downward migration as compared to other types of microplastics. The absence of plastic film derived microplastics at deep soil layers in their study was probably also due to their shorter duration of plastic film mulching and the dry climate (precipitation: 50 mm and evaporation: 2000 mm). Nevertheless, our study shows that plastic film derived microplastics can move through soil to at least 100 cm depth and thus may have the potential to contaminate groundwater. Further, the potential to remediate plastic pollution in deep subsoils will be much more difficult than in topsoils; therefore, it is likely that this plastic legacy will remain in soil for centuries.

5. Conclusion

Long-term use of plastic mulch films can cause accumulation of plastic residues in soils. As conventional plastic mulch films are made of low-density polyethylene, a material that does not readily degrade in soil, the accumulation of these plastic residues causes long-term pollution of soils. Macroscopic plastic residues will disintegrate into micro- and nanoplastics over time, thereby not only increasing the number concentrations of the plastics, but also increasing their bioavailability.

Further, as shown in this study, micro- and nanoplastics have the potential to migrate through the soil profile, and distribute into deeper soil layers, where degradation is minimal.

To minimize plastic pollution of soils, better methods and protocols should be developed to more efficiently remove agricultural plastic films from soils after harvest. Increasing the thickness of plastic mulch films renders the films less fragile, so that they do not readily break apart during removal operations. This will facilitate the removal of plastic films from soils. Nonetheless, complete removal of plastics is not possible even with thicker plastic films. A promising alternative to conventional plastic mulch films are biodegradable plastics, which do not have to be removed from the fields after harvest, but rather can be tilled into the ground where they biodegrade.

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

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.118945.

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