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Accumulation of soil microplastics and phthalate esters in nine typical Chinese croplands using plastic mulch film

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KEYWORDS

Agricultural soil, China, microplastics, mulching film additives, phthalate esters

HIGHLIGHTS

- Surface soil microplastics (MPs) in mulched regions of China ranged from 6.2×10^3 to 4.9×10^4 items kg^{-1}.
- Plastic mulch film derived polyolefin account for up to 35% of soil MPs.
- Plastic mulch film contributes < 0.1% to total soil phthalate esters (PAEs).
- Plastic mulch film is not main contributor to soil MPs and PAEs.

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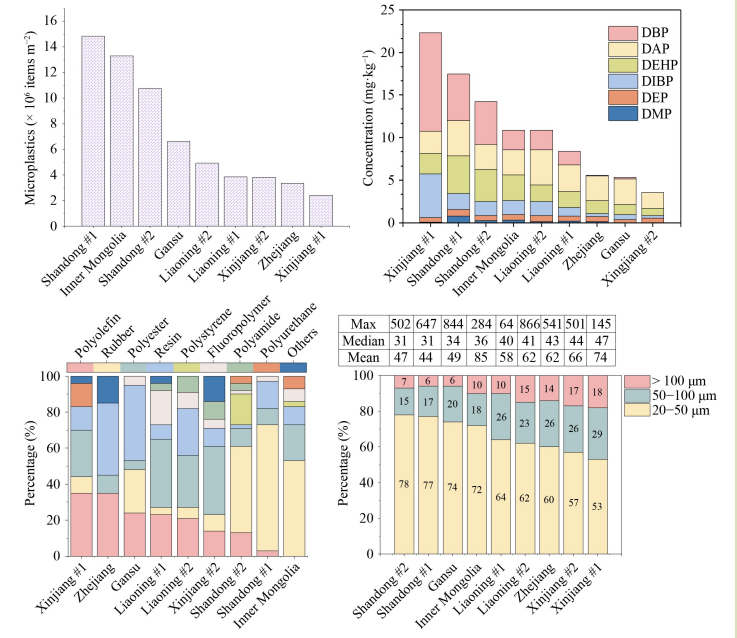
GRAPHICAL ABSTRACT



ABSTRACT

Plastic mulch film (PMF) can release microplastics (MPs) and phthalate esters (PAEs) into agricultural soils. These materials can contaminate the food chain, thereby posing a potential risk to human health. However, inconsistent methodologies hinder cross-regional comparisons of MP and PAE concentrations in agricultural soils, preventing an accurate assessment of the actual risk. To address this knowledge gap, a harmonized analysis of MPs and PAEs was conducted in soil across nine typical mulching region in six provinces of China (Gansu, Inner Mongolia, Liaoning, Shandong, Xinjiang and Zhejiang). The results showed that the abundance of MPs in the 0–30 cm soil layer ranged from 2.4×10^6 to 1.5×10^7 items m^{-2} (equivalent to 5.5×10^3 to 4.9×10^4 items kg^{-1} soil), with the highest abundance in Shandong and the lowest in Xinjiang. These MPs were mainly composed of rubber, polyolefin, polyester, resin, polystyrene, fluoropolymer, polyamide and polyurethane, of which polyolefin (primarily PMF-derived) accounted for up to 35%. Additionally, six PAEs (di(2-ethylhexyl) phthalate, diisobutyl phthalate, dibutyl phthalate, di-n-pentyl phthalate, diethyl phthalate and dimethyl phthalate) were detected, with total residues ranging from 3.6 to 22.3 $mg \cdot kg^{-1}$. It was estimated that the total PAE input from PMF constitutes < 0.1% of the measured PAEs at all the sampling sites. Overall, these findings indicate that PMFs are not the main contributor to MP and PAE contamination in agricultural soils under continued PMF application. While removal and recycling of PMF is essential in reducing PMF-derived MP accumulation in soil, further research into other sources is required to establish impactful mitigation strategies and regulations.

1 Introduction

Global agricultural plastic consumption has reached 12.5 Mt annually, within which plastic mulch film (PMF) is a key component, accounting for about 2.08 Mt[1–3]. Plastic film mulching considerably enhances agricultural productivity by conserving soil moisture, regulating temperature, suppressing weeds or reducing pesticide use, particularly in arid and cold regions[4–7]. However, the slow degradation of PMF (mainly polyethylene based) left behind in the soil after crop harvest has resulted in severe soil contamination[4,8]. The progressive fragmentation of PMF, coupled with inputs from sludge and sewage irrigation and atmospheric deposition, serve as primary pathways for microplastic (MP, < 5 mm) and nanoplastic (< 1 μm) accumulation in agricultural soils[9,10]. At high concentrations, MPs in soils can alter soil properties (e.g., bulk density, porosity and water content) and plant growth[11–13]. In addition, these small plastic particles can vertically migrate into deeper soil layers, posing a risk of groundwater contamination, and nanoplastics can be taken up by crops, thereby entering the food chain and posing potential risks for human health[4,14,15].

In addition to the fragmentation of PMF into smaller particles, PMF can also release associated chemicals and additives throughout the degradation process, potentially increasing the chemical burden placed on agricultural soils. During the production of plastics, various additives are incorporated to enhance the properties of the polymer such as its flexibility, durability and ease of manufacturing[16,17]. Of these, phthalate esters (PAEs) have been widely used as plasticizers within polymer production. PAEs do not chemically react with polymers but instead form loose, non-covalent bonds. As a result, during the breakdown and degradation of PMF, PAEs are readily released into the soil[8,18,19]. This has led to increased PAE concentrations in soils with PMF application compared to soils without PMF[18,20].

China is the largest user of PMF in the world, with 1.41 Mt used in 2018, accounting for approximately 68% of the global total use[3,21]. PMF use in China started as early as 1978, resulting in 47 years of continued PMF use in certain areas[15,22]. This highlights the urgent need to establish the current state of MP and PAE pollution in agricultural soils in China. While multiple studies have reported on the abundance of MPs[15,23–25] or PAEs[26–29] in croplands, at the farm- or regional-scale level in China, the use of various sample processing and analytical methods limits an effective comparison between studies[30].

Therefore, there is a clear need to identify and quantify MPs and PAEs in PMF covered soils across China using a uniform methodology. The objective of this study was to investigate the abundance, size and component distribution of MPs, as well as the occurrence of PAEs in soils from nine typical regions covered by PMF in China. In addition, this study aimed to explore the relationship between MPs and PAEs in Chinese croplands.

2 Materials and methods

2.1 Collection and preparation of soil samples

Nine sampling sites (Table 1) were chosen from six provinces (Gansu, Inner Mongolia, Liaoning, Shandong, Xinjiang and Zhejiang), representing 52% of the PMF covered area in China[3]. There were two sampling sites in Liaoning, Shandong and Xinjiang, and one sampling site in each of the other provinces. The prioritization of site selection was based on the extent of PMF use for a specific cropping system per area, with the dominant PMF cropping systems being prioritized. Three soil layers (0–10, 10–20 and 20–30 cm) were sampled in autumn 2022. Each sampling site consisted of four sampling locations (n = 4) with a distance of at least 200 m between locations. At each location, five points were sampled along a W-shaped transect following the principles of randomization, equal quantity and multi-point mixing (Fig. S1). For each soil layer, soil from the five sampling points within the same replicate was homogenized in equal mass proportions and quartered to form one representative composite sample. In total, 108 composite soil samples (9 sampling sites × 4 replicate locations per site × 3 soil depths) were collected. The collected soil samples were stored in aluminum containers and air-dried for MP and PAE analysis. Detailed descriptions of the sampling sites are given in Table 1, Table S1 and Table S2.

2.2 Microplastic extraction and quantification from soil

The methods of Qi[31] and Ren et al.[32] were used to extract MPs from soil with slight modifications. Briefly, 5.0 g of air-dried soil was added to 200 mL of saturated NaCl solution (1.2 g·cm^{-3}) in a glass beaker, stirred for 30 min, and left to settle for 24 h, after which the supernatant was decanted into a new, clean glass beaker. This process was repeated three times. Following this, 200 mL of 4 mol·L^{-1} NaOH were added to the combined supernatant, and the solution was stirred for 30 min and left to settle for 24 h to digest organic matter. To stain MPs, Nile red was added to the solution at a ratio of 1:2000, homogenized, and incubated in the dark for 30 min. We assumed that polymer particles will be dyed by Nile red whereas other organic/inorganic particles will not. Finally, the solution was filtered through a membrane (mixed cellulose, 0.2 μm, Jinteng Experiment Equipment Co., Ltd, Tianjing, China) using vacuum filtration.

The quantification of MPs on the membrane was carried out using a fluorescence microscope (DM4B, Leica, Wetzlar, Germany) with excitation and emission wavelengths of 360 and 425 nm, respectively, at 50× magnification. The particle distribution on the membrane was visually determined to be homogenous, and 5% of the total membrane area was analyzed, resulting in 13 field of views, which were marked on the membrane for subsequent micro-Fourier transform infrared spectroscopy (μ-FTIR; LUMOS I, Bruker, Bremen, Germany). The obtained images were processed using ImageJ[33] to quantify the total quantity of particles and the average particle diameter. The lower size detection limit for particle quantification was set to 20 μm diameter, as the recognition of particles > 20 μm would have led to considerable uncertainty.

Table 1 Background information for the nine sampling sites

Sampling sites	Coordinates	Crop	Climatic zone	Soil type	Annual average precipitation (mm)	Annual average air temperature (°C)
Gansu	38°28′N, 104°44′E	Maize	Temperate zone	Loessial soil	390	6.4
Inner Mongolia	40°42′N, 107°02′E	Maize	Temperate zone	Irragric anthrosol	138	8.0
Liaoning #1	41°70′N, 119°55′E	Maize	Temperate zone	Cinnamon soil	614	7.6
Liaoning #2	41°69′N, 119°55′E	Millet	Temperate zone	Cinnamon soil	614	7.6
Shandong #1	36°38′N, 120°18′E	Maize	Warm temperate zone	Brown soil	1079	13.5
Shandong #2	36°42′N, 120°20′E	Peanut	Warm temperate zone	Brown soil	1079	13.5
Xinjiang #1	45°20′N, 84°58′E	Cotton	Warm temperate zone	Gleysol	187	7.0
Xinjiang #2	40°63′N, 81°31′E	Cotton	Warm temperate zone	Cambisol	56.8	10.5
Zhejiang	29°76′N, 121°58′E	Strawberry	Sub-tropical zone	Paddy soil	1538	16.2

2.3 Microplastic identification

The μ-FTIR spectrometer was used to determine the infrared spectra of polymers, resolve functional groups and identify polymer types of the particles on the membrane. This process was only conducted for particles in the 0–10 cm soil layer, prioritizing the layer in which we postulated that the most diversity of polymer types. The wavelength range was set from 500 to 4000 cm^{-1}, with a resolution of 4 cm^{-1} and 64 scans per sample spot. On each membrane, three particles were selected and analyzed from each pre-marked area, with three such predefined areas per membrane. These nine particles corresponded to fluorescent spots observed under fluorescence microscopy. Additionally, two random particles were selected from each quadrant on the filter membrane, resulting in a total of 17 particles per membrane (Fig. S2). The lower size detection limit was set to 20 μm, which is the smallest detectable particle size for μ-FTIR under the premise of accurate detection results. After baseline correction, particle spectra were matched to the reference library to determine their chemical composition. The match quality threshold was set to ⩾ 45%, particles below this threshold were classed as unidentified, likely organic or inorganic contaminants. Some particles were not identified as polymers, but rather as additives (e.g., EDTA), minerals (e.g., quartz sand) or metals (e.g., zinc oxide). These data were then used to calculate the accuracy of MPs quantification via Nile red staining and corrections for preliminary quantification were applied (Table S3).

2.4 Phthalate ester determination

Seventeen PAEs were quantified in soil samples by gas chromatography coupled with mass spectrometry (GC-MS; 7000D, Agilent, Santa Clara, CA, USA) with a 60 μm × 250 μm × 0.25 μm column (Agilent 122-5062E). This analysis was only done for the 0–10 cm soil layer based on the assumption that PMF residues will accumulate most in this layer. The target PAEs: di(2-methoxyethyl) phthalate, diisoheptyl phthalate, diethyl phthalate, dipropyl phthalate, di-n-hexyl phthalate, butyl benzyl phthalate, dibutoxyethyl phthalate, dicyclohexyl phthalate, dipentyl phthalate, di-n-octyl phthalate, diisononyl phthalate, di(2-ethylhexyl) phthalate (DEHP), dimethyl phthalate (DMP), diethyl phthalate (DEP), dibutyl phthalate (DBP), diallyl phthalate (DAP) and diisobutyl phthalate (DIBP). Briefly, 5.0 g of soil was added to 30 mL of n-hexane-acetone (1:1 *v/v*), sonicated for 30 min and centrifuged for 3 min at 3000 $r{\cdot}min^{-1}$ to separate the supernatant. The chromatographic column consisted of a fused silica capillary column with a 5% phenyl methylpolysiloxane stationary phase and the GC-MS settings were: 80 °C for 1 min, increased to 280 °C at 20 $°C{\cdot}min^{-1}$ and held for 20 min, with an injection volume of 1.2 μL. The carrier gas was helium with a flow rate of 1 $mL{\cdot}min^{-1}$. The PAEs were quantified by a five-point calibration curve[29,34].

2.5 Quality control

Soil samples were collected using a stainless-steel soil auger and stored in aluminum containers. During sample processing, cotton laboratory coats and gloves were worn, and all experimental materials were made of glass or metal. Further, glass beakers were always covered with aluminum foil to prevent contamination of the samples by airborne particles. All glassware was thoroughly cleaned prior to use with distilled water. Extraction blanks were run alongside samples (n = 3) to account for any process-induced contamination.

2.6 Calculation and statistical analysis

MP abundance (items m^{-2}) in the 0–30 cm soil layer was calculated as follows:

$$A_{0-30} = \sum_{k=1}^{3} (D \times A \times BD_k \times N_k) \quad (1)$$

where, A_{0-30} is the abundance of MPs (items m^{-2}) in the 0–30 cm soil layer, k is the individual soil layer in 0–30 cm layer (1–3 for 0–10, 10–20 and 20–30 cm), D is the depth of individual soil layer (0.1 m), A is the area (1 m^2), BD_k is the bulk density of each individual soil layer ($kg{\cdot}m^{-3}$) and N_k is the number of MPs (items kg^{-1}).

To estimate the contribution of PMF to soil PAEs, we calculated the theoretical release of PAEs from the use of PMF. Then, we calculated the contribution of PMF to soil PAEs through dividing theoretical release of PAEs from PMF by the actual measured PAE content in this study. The theoretical release of PAEs in to soil from PMF use over time (PAE_{film}, $mg{\cdot}kg^{-1}$ soil) was estimated as follows, assuming that 100% of PAEs in PMF residue were released into soil and not degraded (i.e., being the worst-case scenario for PAE accumulation).

$$PAE_{film} = \frac{A_{film} \times T \times \rho \times C_{PAE} \times (1 - R) \times Year}{A_{soil} \times D \times BD} \quad (2)$$

where, A_{film} is the area (1 m^2) covered by the PMF, T is the thickness (m) of the PMF (1×10^{-5} m), ρ is the density

(kg·m^{-3}) of polyethylene PMF (assuming 930 kg·m^{-3})[35], C_{PAE} is the total PAE content (mg·kg^{-1}) in plastic film (15.7 mg·kg^{-1} based on Ding et al.[34]), R is the recovery rate of plastic film by farmers (assuming to be 80%), Year is the number of years of continued PMF use, A_{soil} is the area of soil covered by the PMF (1 m^2), D is soil depth (0–0.1 m) and BD is soil bulk density (kg·m^{-3}) (Table S2).

Statistical analysis was performed using SPSS 26.0 (IBM, Inc., Armonk, NY, USA). Linear regression was used to investigate relationship between PAE and MP abundance. One-way analysis of variance was performed to determine differences in MP abundance between soil layers, with the significance level set at 0.05 (95% confidence interval). Graphical representations were generated using Origin 2024b.

3 Results

3.1 Microplastic abundance, size and chemical composition

The abundance of MPs in the 0–30 cm soil layer ranged from 2.4 × 10^6 to 1.5 × 10^7 items m^{-2} in the nine target provinces with high PMF use across China, with an average of 7.1 × 10^6 items m^{-2} (Fig. 1(a)). MP abundance in Shandong (1.1 × 10^7 to 1.5 × 10^7 items m^{-2}) and Inner Mongolia (1.3 × 10^7 items m^{-2}) were much greater than other regions (Gansu (7 × 10^6 items m^{-2}), Liaoning #2 (5 × 10^6 items m^{-2}), Liaoning #1 (4 × 10^6 items m^{-2}), Xinjiang #2 (4 × 10^6 items m^{-2}), Zhejiang (3 × 10^6 items m^{-2}) and Xinjiang #1 (2 × 10^6 items m^{-2})). The abundance of soil MPs in the 0–10, 10–20 and 20–30 cm soil layer ranged from 6.2 × 10^3 to 4.8 × 10^4, 5.5 × 10^3 to 3.1 × 10^4 and 9 × 10^3 to 2.9 × 10^4 items kg^{-1}, respectively (Fig. 1(b)). We found an inverse relationship between particle size and abundance, that is, the smaller the particle size, the greater the abundance and vice versa (Fig. 1(c)).

MP particles in the size range 20–50 μm accounted for > 50% of the total particle number, while particles sized between 20 and 100 μm accounted for > 90% at almost all sampling sites. Although the maximum particle size was > 500 μm in all sampling points, the median was < 50 μm and the mean < 70 μm.

We detected nine categories of MP in the soil samples (Fig. 1(d)), including polyolefin, rubber, polyester, resin, polystyrene, fluoropolymer, polyamide, polyurethane and others. The categories were classified as follows: polyolefin (polyethylene and polypropylene), rubber (ethylene-propylene rubber and neoprene rubber), polyester (ethylene-vinyl alcohol copolymer, polybutylene terephthalate and polyethylene terephthalate), resin (phenolic resin), polystyrene (polystyrene, polystyrene foam and high impact polystyrene), fluoropolymer (polytetrafluoroethylene), polyamide (nylon66 and nylon12), polyurethane (polyurethane and polyurethane foam) and others (polyether imide and vinyl chloride-vinyl acetate copolymer).

Rubber dominated within the nine categories in Shandong #2 (70%), Shandong #1 (48%) and Inner Mongolia (53%). However, polyolefin (including polyethylene and polypropylene) is the primary components of PMFs and was abundant in multiple regions, particularly in Xinjiang #1 and Zhejiang (both 35%). The proportions of polyolefins in other regions followed the order: Gansu (24%) > Liaoning #1 (23%) > Liaoning #2 (21%) > Xinjiang #2 (14%) > Shandong #2 (13%) > Shandong #1 (3%) > Inner Mongolia (0%).

3.2 Phthalate ester concentrations in soils

Of the 17 targeted PAEs, we identified six PAEs which were abundant in the majority of samples, namely DEHP, DIBP, DBP, DAP, DEP and DMP (Fig. 2). The other 11 PAEs were not detected in our soil samples being not present or below the detection limit. The total amount of Σ_6 PAEs ranged from 3.6 to 22.3 mg·kg^{-1}, with an average of 11.1 mg·kg^{-1} (Fig. 2(a) and Table S4). The average abundance of PAEs in the nine provinces was as follows: Xinjiang #1 (22.3 mg·kg^{-1}) > Shandong #1 (17.4 mg·kg^{-1}) > Shandong #2 (14.2 mg·kg^{-1}) > Inner Mongolia (12.0 mg·kg^{-1}) > Liaoning #2 (10.9 mg·kg^{-1}) > Liaoning #1 (8.4 mg·kg^{-1}) > Zhejiang (5.6 mg·kg^{-1}) > Gansu (5.3 mg·kg^{-1}) > Xinjiang #2 (3.6 mg·kg^{-1}). Only three of the six most abundantly identified PAEs (DEHP, DEP and DMP) are currently regulated in China[36]. The total amount of these Σ_3 PAEs ranged from 1.4 to 6.0 mg·kg^{-1} (Fig. 2(b)).

We also found a regional difference in PAE abundance (Fig. 2(c)), with DBP accounting for 31%–52% of PAEs in Shandong #1, Shandong #2 and Xinjiang #1 (in that order). DAP was most abundant in the remaining provinces, accounting for 38%–56% of PAEs.

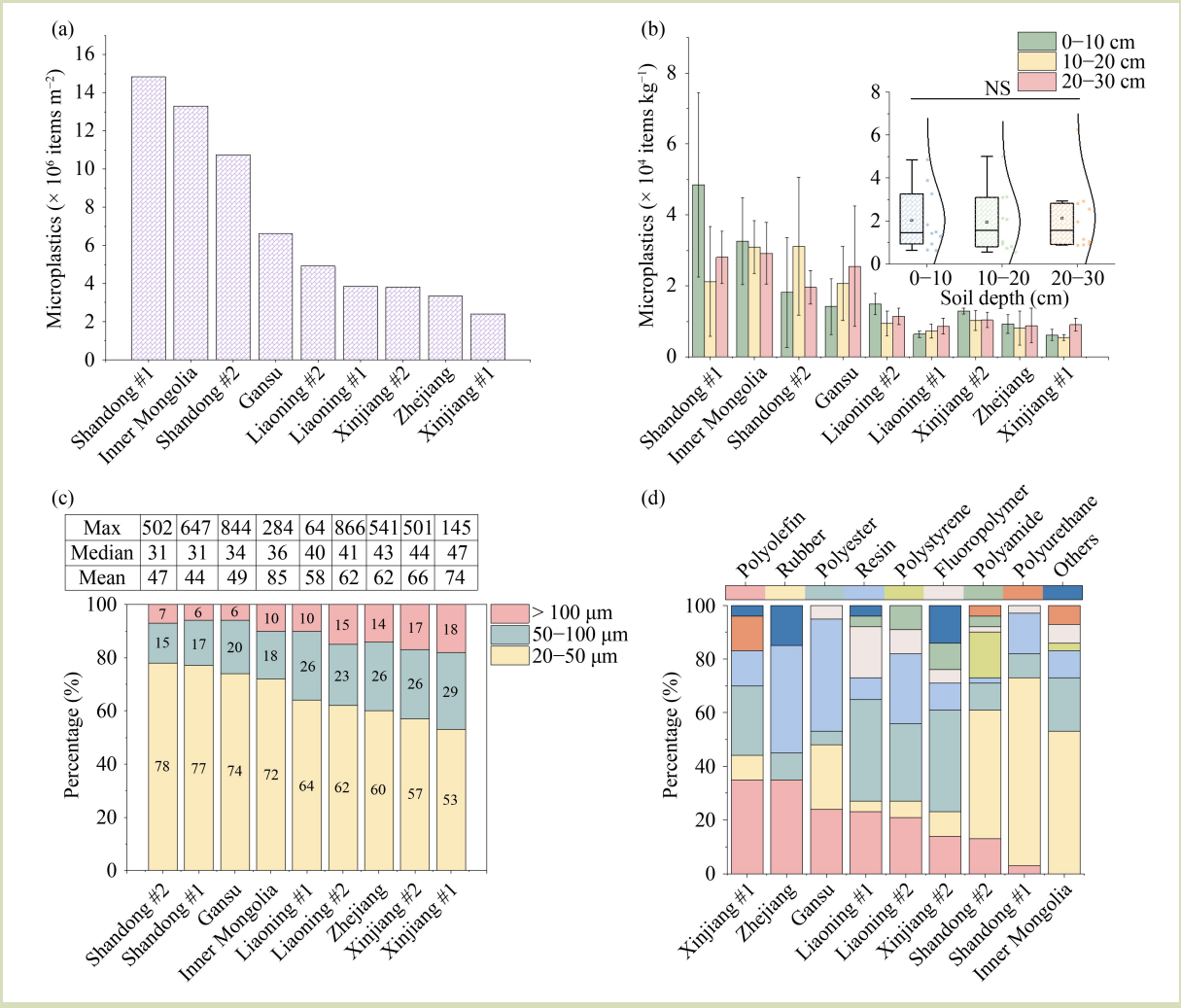


Fig. 1 Abundance (a, b), size distribution (c), and chemical composition (d) of microplastic (MP) at nine sampling sites using plastic film mulch in China. (a) Total number (items m^{-2}) of MP in the 0–30 cm soil layer. (b) MP number (items kg^{-1}) in the 0–10, 10–20 and 20–30 cm soil layers. The inset shows the difference in MP number in the 0–10, 10–20 and 20–30 cm soil layers across the nine regions. (c) Abundance of MP size categories expressed in % (categories: > 100, 50–100 and 20–50 μm). Maximum, median, and mean (μm) microplastics size in the 0–30 cm soil layers in each region are presented in the table above the graph. (d) Percentage of most abundant polymer categories in the 0–30 cm soil layers. Nine categories included polyolefin (polyethylene and polypropylene), rubber (ethylene-propylene rubber and neoprene rubber), polyester (ethylene-vinyl alcohol copolymer, polybutylene terephthalate and polyethylene terephthalate), resin (phenolic resin), polystyrene (polystyrene, polystyrene foam and high impact polystyrene), fluoropolymer (polytetrafluoroethylene), polyamide (nylon66 and nylon12), polyurethane (polyurethane and polyurethane foam) and others (polyether imide and vinyl chloride-vinyl acetate copolymer).

3.3 Relationship between soil phthalate esters and microplastic concentrations

DMP concentration in soil increased with MP abundance in soil (r = 0.64, p = 0.0003, Fig. 3(a)). This was also observed for DEHP (r = 0.62, p < 0.0001, Fig. 3(b)). Abundance of DEP, DBP, DAP and DIBP was not no significantly related to MPs abundance.

4 Discussion

4.1 Accumulation of soil microplastics in Chinese croplands using plastic film mulch

The MP (≥ 20 μm) abundance in the nine typical mulching regions ranged from 5.5 × 10^3 to 4.9 × 10^4 items kg^{-1} and 2.4 ×

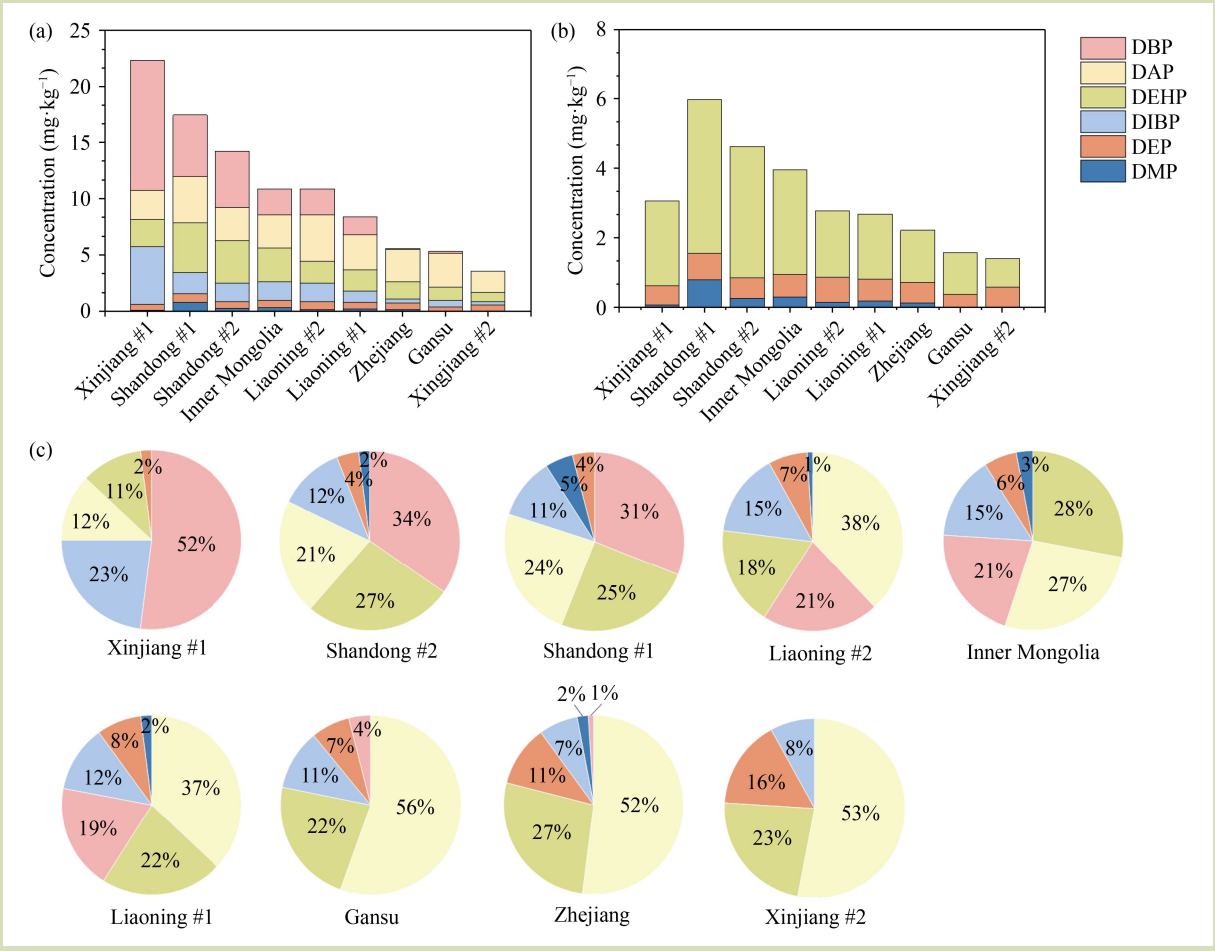


Fig. 2 Phthalate ester (PAE) concentration in the 0–10 cm soil layer at nine sampling sites using plastic film mulch in China. (a) Total concentrations of six PAEs. (b) Concentration of the three regulated PAEs (DEHP, DEP and DMP) in China. (c) Average percentage of individual PAEs in each region. DAP, diallyl phthalate; DBP, dibutyl phthalate; DEHP, di(2-ethylhexyl) phthalate; DEP, diethyl phthalate; DIBP, diisobutyl phthalate; and DMP, dimethyl phthalate.

10^6 to 1.5×10^7 items m^{-2} at 0–30 cm soil depth (Fig. 1). These numbers were higher than the majority of previously reports on the abundance of MPs in Chinese agricultural soils. A previous nationwide survey showed an average MP (size not reported) abundance of 1837 items kg^{-1} across 109 Chinese cities[23], and a regional survey reported MP (> 100 μm) abundance of 77–730 items kg^{-1} in croplands of Northwest China[24]. Notably, a study focusing on cropland with 32 years of continued PMF use reported a MP abundance of 10,586 items kg^{-1} (> 100 μm) at 0–10 cm soil depth[15]. These large differences in reported MP numbers could be attributed to the lower size limit used for MP detection in our study. We included MPs ⩾ 20 μm whereas these earlier studies excluded particles < 100 μm or did not indicate a set analytical size range. In our study, particles sized between 20 and 100 μm accounted for more than 80% of the total MPs abundance in all regions, and the number of particles between 20 and 100 μm is at least one order of magnitude higher than the MP abundance for particles > 100 μm (Fig. 1(c)).

We also found that MPs in agricultural soils are not exclusively derived from PMF. Polyolefins (primarily PMF-derived) accounted for up to 35% of total MPs across all the regions (Fig. 1(d)). Similarly, 33%–56% of MPs in an agricultural field after 32 years of continued PMF application were most likely to have been derived from PMF[15]. Past national surveys of soils

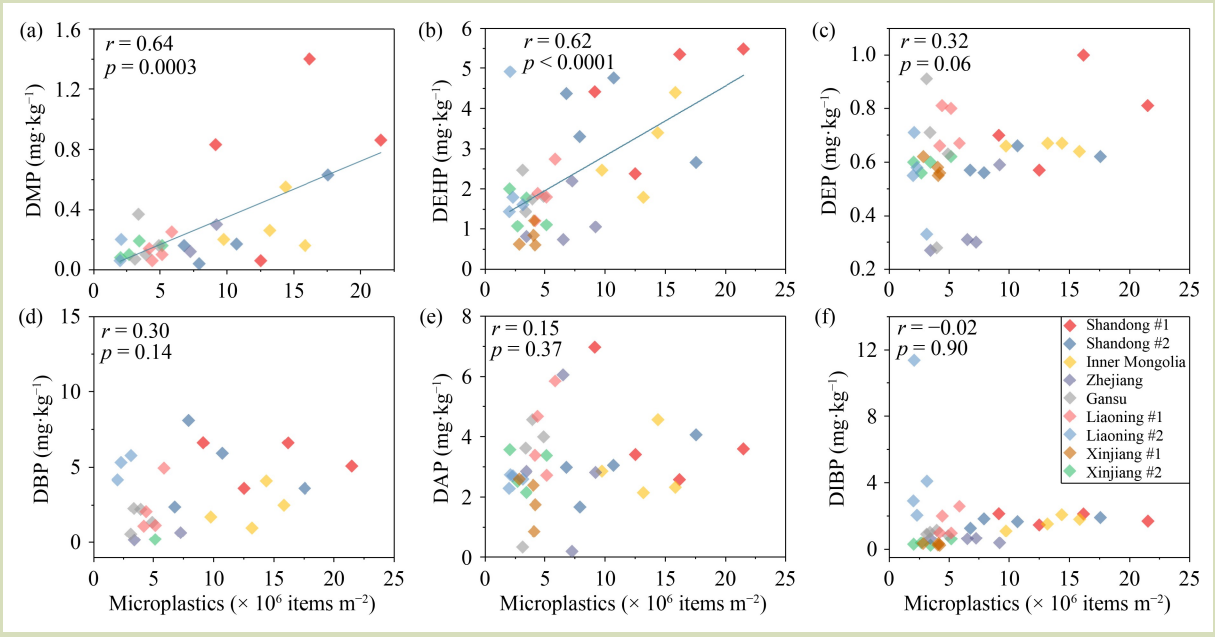


Fig. 3 Relationships between the six phthalate esters and microplastic abundance in the nine sampling sites using plastic film mulch in China. (a) DMP, dimethyl phthalate; (b) DEHP, di(2-ethylhexyl) phthalate; (c) DEP, diethyl phthalate; (d) DBP, dibutyl phthalate; (e) DAP, diallyl phthalate; (f) DIBP, diisobutyl phthalate.

under PMF application reported 31.7%– 64.4% of MPs being likewise attributed to PMFs[25,37,38]. Apart from PMF, atmospheric deposition, irrigation, application of organic amendments (e.g., biosolid, compost and sludge) and coated fertilizers also contribute to MPs in agricultural soils[15]. In Shandong #2, Shandong #1 and Inner Mongolia, rubber dominated the MP chemical composition (Fig. 1(d)). Rubber at these locations most likely originates from tire wear particles transported via atmospheric deposition or wastewater irrigation pathways[39–41].

The discrepancy between PMF use and MP concentrations is most evident in the data from Xinjiang (#1 and #2), where MP concentration was lowest despite the fact that Xinjiang has the highest PMF use compared to other regions and macroplastic (> 5 mm) residue was greatest of all provincial administrations[3]. Our findings align with the results of two previous national soil condition censuses in China, ranking MP concentration in Xinjiang as low in comparison to other provinces[23,25]. It is suggested that the reason for low MP concentrations in Xinjiang is linked to different irrigation sources, as groundwater irrigation is prevalent in this region, whereas other regions with high MP numbers tend to rely on river water[25], which has been shown to contain more MPs compared to groundwater[42]. This further supports our finding, that PMF use represents only a small contributor to MPs in agricultural soils (Fig. 1(d)).

4.2 Accumulation of phthalate esters in Chinese agricultural soils under plastic mulch film application

The total concentration of the six most abundant PAEs in soils ranged from 3.57 to 22.3 mg·kg^{-1} at 0–10 cm soil depth across the nine sampling sites under PMF examined in this study (Fig. 2(a)). Notably, values reported here exceed those from previous studies. A soil survey of 31 Chinese provinces in 2013 reported the total concentration of 15 PAEs as 0.08–6.37 mg·kg^{-1}[26], while another survey of 24 provinces in 2019 reported the total concentration of nine PAEs as 0.07–1.24 mg·kg^{-1}[28]. The two soil surveys did not measure DBP and DAP, however, they accounted for about 57% of the total PAEs in our study (Fig. 2(a)). Similarly, previous studies have reported soil DBP concentrations of 9.8–11.6 mg·kg^{-1} in Shandong[43] and 11.2–57.7 mg·kg^{-1} in Xinjiang[44]. Therefore the lack of quantification DBP and DAP in previous studies is

Table 2 Theoretical phthalate ester (PAE) release from plastic mulch film (PMF) in 0–10 cm soil layer according to calculations presented in section 2.6

Sampling site	Mulching duration (year)	Theoretical PAEs released from PMF (μg·kg^{-1} soil)	Measured PAE content (mg·kg^{-1} soil)	Theoretical PAEs from PMF / measured PAEs (%)
Gansu	1	0.2	5.41	0.004
Inner Mongolia	22	4.6	10.86	0.042
Liaoning #1	7	1.3	8.39	0.015
Liaoning #2	7	1.5	10.87	0.014
Shandong #1	13	3.4	17.43	0.020
Shandong #2	13	3.6	14.21	0.026
Xinjiang #1	25	6.1	22.32	0.027
Xinjiang #2	10	2.5	3.57	0.070
Zhejiang	10	2.3	5.55	0.041

the likely reason for the discrepancy between the two national soil surveys and our study.

Plastic has been established as a dominant carrier for PAEs entering soils, due to PAEs being abundantly used in the plastic production process. DEHP is a dominant PAEs in PMF[45–47], and has long ester chains and strong hydrophobicity, making it readily adsorbed by carbonaceous materials (e.g., biochar and MP) in soil and sediments, and thus making it difficult for it to undergo biodegradation[48–50]. This contributed to the close positive correlation between DEHP concentration and MP abundance in soil (Fig. 3(a)). DBP and DEP had a weak positive relationship with MP abundance in soil (Fig. 3(c,d)), possibly indicating that other sources (e.g., irrigation) contribute to their input apart from PMF. Lastly, DAP and DIBP are rarely used in plastic production, therefore no relation for these with MP abundance was observed (Fig. 3(e,f).

We also estimated the potential contribution of PMF to soil PAEs (Table 2). Even when operating under the worst-case scenario assumption that all PAEs from PMFs persist in soil (i.e., no microbial degradation or leaching), the total PAE input from PMFs would constitute only < 0.1% of the measured PAEs at all the sampling sites. We acknowledge that this assumption is not based on modeled mobility and degradation of PAEs in soil, and is merely used to illustrate the highest potential contribution of PMFs to PAEs in soil after continued PMF use. In reality, the PMF contribution to PAEs in soil is likely to be even lower than the 0.1% calculated here. Accordingly, we conclude that the contribution of PMFs to soil PAE pollution is negligible. In contrast, fertilizers[46,51], sewage sludge[45,52–54] and irrigation water[55] are much more likely to be primary sources of PAEs in Chinese soils.

5 Conclusions

This study investigated MP and PAE abundance from nine typical PMF-covered regions in China. The MP abundance ranged from 2.4×10^6 to 1.5×10^7 items m^{-2} in the 0–30 cm soil layer (equivalent to 5.5×10^3 to 4.9×10^4 items kg^{-1} soil). MP chemical composition included polyolefins, rubber, polyester, resins, polystyrene, fluoropolymer, polyamide, polyurethane and others. Polyolefins (primarily PMF-derived) accounted for up to 35% of the total MP number across all the regions. Six PAEs (DEHP, DIBP, DBP, DEP, DMP and DAP) were detected, with total concentrations ranging from 3.57 to 22.3 mg·kg^{-1}. DEHP and DBP were the dominant compounds, accounting for 76.4% and 9.3% of the total PAE concentration, respectively. Assuming all PAEs from plastic films persist in soil without degradation or leaching losses, the PAE input from PMFs constitutes < 0.1% of the measured PAEs at all the sampling sites. Our study provides evidence that PMFs are not the primary source of MPs and PAEs in Chinese soils under continued PMF application, but that other sources (e.g., atmospheric deposition, irrigation and organic amendments) are likely major contributors that warrant further investigation. For future research, we therefore strongly suggest to further investigate other sources of soil contamination, to implement impactful mitigation strategies and regulations.

Supplementary materials

The online version of this article at https://doi.org/10.15302/J-FASE-2025660 contains supplementary materials (Figs. S1–S2; Tables S1–S4).

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Compliance with ethics guidelines

Yingming Sun, Shitong Li, Fan Ding, Martine Graf, Ruimin Qi, Xuejun Liu, Kai Wang, Tida Ge, Jingkuan Wang, David R. Chardwick, and Davey L. Jones declare that they have no conflicts of interest or financial conflicts to disclose. This article does not contain any studies with human or animal subjects performed by any of the authors.

REFERENCES

1. Food and Agriculture Organization of the United Nations (FAO). Assessment of Agricultural Plastics and Their Sustainability. A Call for Action. Rome: *FAO*, 2009. Available at FAO website on December 7, 2024
2. Shah F, Wu W. Chapter Five—Use of plastic mulch in agriculture and strategies to mitigate the associated environmental concerns. *Advances in Agronomy*, 2020, **16**: 231–287
3. Dai J Z, Hu C, Flury M, Huang Y, Rillig M C, Ji D C, Peng J W, Fei J C, Huang Q, Xiong Y C, Yang N, Jones D L, Wang J K, Ding F. National inventory of plastic mulch residues in Chinese croplands from 1993 to 2050. *Global Change Biology*, 2025, **31**(6): e70297
4. Hofmann T, Ghoshal S, Tufenkji N, Adamowski J F, Bayen S, Chen Q Q, Demokritou P, Flury M, Hüffer T, Ivleva N P, Ji R, Leask R L, Maric M, Mitrano D M, Sander M, Pahl S, Rillig M C, Walker T R, White J C, Wilkinson K J. Plastics can be used more sustainably in agriculture. *Communications Earth & Environment*, 2023, **4**(1): 332
5. Sun D B, Li H G, Wang E L, He W Q, Hao W P, Yan C R, Li Y Z, Mei X R, Zhang Y Q, Sun Z X, Jia Z K, Zhou H P, Fan T L, Zhang X C, Liu Q, Wang F J, Zhang C C, Shen J B, Wang Q S, Zhang F S. An overview of the use of plastic-film mulching in China to increase crop yield and water-use efficiency. *National Science Review*, 2020, **7**(10): 1523–1526
6. Xiao L G, Wei X, Wang C Y, Zhao R Q. Plastic film mulching significantly boosts crop production and water use efficiency but not evapotranspiration in China. *Agricultural Water Management*, 2023, **275**: 108023
7. Yang F K, He B L, Dong B, Zhang G P. Film mulched ridge-furrow tillage improves the quality and fertility of dryland agricultural soil by enhancing soil organic carbon and nutrient stratification. *Agricultural Water Management*, 2024, **292**: 108686
8. Xu B L, Liu F, Cryder Z, Huang D, Lu Z J, He Y, Wang H Z, Lu Z M, Brookes P C, Tang C X, Gan J, Xu J M. Microplastics in the soil environment: Occurrence, risks, interactions and fate—A review. *Critical Reviews in Environmental Science and Technology*, 2020, **50**(21): 2175–2222
9. van den Berg P, Huerta-Lwanga E, Corradini F, Geissen V. Sewage sludge application as a vehicle for microplastics in eastern Spanish agricultural soils. *Environmental Pollution*, 2020, **261**: 114198
10. Ren S Y, Kong S F, Ni H G. Contribution of mulch film to microplastics in agricultural soil and surface water in China. *Environmental Pollution*, 2021, **291**: 118227
11. Qi Y L, Beriot N, Gort G, Lwanga E H, Gooren H, Yang X M, Geissen V. Impact of plastic mulch film debris on soil physicochemical and hydrological properties. *Environmental Pollution*, 2020, **266**(Part3): 115097
12. Zhang J R, Ren S Y, Xu W, Liang C, Li J J, Zhang H Y, Li Y N, Liu X J, Jones D L, Chadwick D R, Zhang F S, Wang K. Effects of plastic residues and microplastics on soil ecosystems: a global meta-analysis. *Journal of Hazardous Materials*, 2022, **435**: 129065
13. Yu Y X, Battu A K, Varga T, Denny A C, Zahid T M, Chowdhury I, Flury M. Minimal impacts of microplastics on soil physical properties under environmentally relevant

concentrations. *Environmental Science & Technology*, 2023, **57**(13): 5296–5304

14. Hua Z D, Zhang T L, Luo J Q, Bai H D, Ma S R, Qiang H, Guo X T. Internalization, physiological responses and molecular mechanisms of lettuce to polystyrene microplastics of different sizes: validation of simulated soilless culture. *Journal of Hazardous Materials*, 2024, **462**: 132710
15. Li S T, Ding F, Flury M, Wang Z, Xu L, Li S Y, Jones D L, Wang J K. Macro- and microplastic accumulation in soil after 32 years of plastic film mulching. *Environmental Pollution*, 2022, **300**: 118945
16. Pfaendner R. A brief history of plastic additives. Part 1: Antioxidants. *Macromolecular Materials and Engineering*, 2025, **310**(8): 2500039
17. Pfaendner R. How will additives shape the future of plastics. *Polymer Degradation & Stability*, 2006, **91**(9): 2249–2256
18. Steinmetz Z, Wollmann C, Schaefer M, Buchmann C, David J, Tröger J, Muñoz K, Frör O, Schaumann G E. Plastic mulching in agriculture. Trading short-term agronomic benefits for long-term soil degradation. *Science of the Total Environment*, 2016, **550**: 690–705
19. Shi M, Sun Y Y, Wang Z H, He G, Quan H X, He H X. Plastic film mulching increased the accumulation and human health risks of phthalate esters in wheat grains. *Environmental Pollution*, 2019, **250**: 1–7
20. Kong S F, Ji Y Q, Liu L L, Chen L, Zhao X Y, Wang J J, Bai Z P, Sun Z R. Diversities of phthalate esters in suburban agricultural soils and wasteland soil appeared with urbanization in China. *Environmental Pollution*, 2012, **170**: 161–168
21. Liu E K, He W Q, Yan C R. 'White revolution' to 'white pollution'—Agricultural plastic film mulch in China. *Environmental Research Letters*, 2014, **9**(9): 091001
22. Yang L L, Heng T, He X L, Yang G, Zhao L, Li Y H, Xu Y. Spatial-temporal distribution and accumulation characteristics of residual plastic film in cotton fields in arid oasis area and the effects on soil salt transport and crop growth. *Soil & Tillage Research*, 2023, **231**: 105737
23. Chen L Y, Yu L, Li Y J, Han B J, Zhang J D, Tao S, Liu W X. Spatial distributions, compositional profiles, potential sources, and influencing factors of microplastics in soils from different agricultural farmlands in China: a national perspective. *Environmental Science & Technology*, 2022, **56**(23): 16964–16974
24. Cao J H, Gao X D, Hu Q, Li C J, Song X L, Cai Y H, Siddique K H M, Zhao X N. Distribution characteristics and correlation of macro- and microplastics under long-term plastic mulching in northwest China. *Soil & Tillage Research*, 2023, **231**: 105738
25. Xue Y H, Li J, Jin T, Liu D S, Zou G Y, Li F, Wang K, Xu L. Meso- and microplastic contamination in mulching cultivated soil at a national scale, China. *Journal of Cleaner Production*, 2023, **418**: 138215
26. Niu L L, Xu Y, Xu C, Yun L X, Liu W P. Status of phthalate esters contamination in agricultural soils across China and associated health risks. *Environmental Pollution*, 2014, **195**: 16–23
27. Zheng S A, Ni R X, Bao Z, Liu D L, Wu Z Y, Gao S B. Occurrence and risk assessment of phthalic acid esters (PAEs) in soils and agricultural products from farmland associated with intensively plastic film mulching, Northwest China. *Environmental Chemistry*, 2020, **39**(7): 1839–1850 (in Chinese)
28. Xu Y W, Jia W Q, Hu A L, Wang J, Huang Y, Xu J M, He Y, Lu Z J. Co-occurrence of light microplastics and phthalate esters in soils of China. *Science of the Total Environment*, 2022, **852**: 158384
29. Liu H J, Liu X T, Wang K, Ma X W, Gao H H, Liu X J, Yan C R. The occurrence and safety evaluation of phthalic acid esters in Oasis agricultural soils of Xinjiang, China. *Ecotoxicology and Environmental Safety*, 2025, **290**: 117593
30. Wang K, Liu X J, Chadwick D R, Yan C R, Reay M, Ge T D, Ding F, Wang J K, Qi R M, Xiao M L, Jiang R, Chen Y L, Ma J, Lloyd C, Evershed R P, Luo Y M, Zhu Y G, Zhang F S, Jones D L. The agricultural plastic paradox: feeding more, harming more. *Environment International*, 2025, **198**: 109416
31. Qi R M. Characteristics and Ecological Effects of Soil Microplastic in Typical Agricultural Region with Plastic Film Mulching in China. Dissertation for the Doctoral Degree. Beijing: *Institute of Environment and Sustainable Development in Agriculture, Chinese Academy of Agricultural*, 2021 (in Chinese)
32. Ren S Y, Graf M, Wang K, Zhang J R, Zhang H Y, Liu X T, Li J J, Zhu T, Ren K G, Sun Y M, Qi R M, Collins B I, Xu L, Jiang X X, Cui J X, Ding F, Yan C R, Liu X J, Jones D L, Chadwick D R. Separation and identification of conventional microplastics from farmland soils. *Journal of Visualized Experiments*, 2025, **217**(217): e67064
33. Schindelin J, Arganda-Carreras I, Frise E, Kaynig V, Longair M, Pietzsch T, Preibisch S, Rueden C, Saalfeld S, Schmid B, Tinevez J Y, White D J, Hartenstein V, Eliceiri K, Tomancak P, Cardona A. Fiji: an open-source platform for biological-image analysis. *Nature Methods*, 2012, **9**(7): 676–682
34. Ding W L, Liu Q, Liu Q Y, Yan C R. Characteristics and safety of phthalates (PAEs) for plastic mulch films in China. *Journal of Agro-Environmental Science*, 2021, **40**(5): 1008–1016 (in Chinese)
35. Stubbins A, Lavender Law K, Munoz E S, Bianchi S T, Zhu L X. Plastics in the Earth system. *Science*, 2021, **373**(6550): 51–55
36. Ministry of Ecology and Environment of the People's Republic of China. Soil Environmental Quality—Risk Control Standard for Soil Contamination of Agricultural Land GB 15618–2018. Beijing: *China Environmental Science Press*, 2018, 13 (in Chinese)
37. Wang J, Li J Y, Liu S T, Li H Y, Chen X C, Peng C, Zhang P P,

Liu X H. Distinct microplastic distributions in soils of different land-use types: a case study of Chinese farmlands. *Environmental Pollution*, 2021, **269**: 116199

38. Xu L, Xu X B, Li C, Li J, Sun M X, Zhang L X. Is mulch film itself the primary source of meso- and microplastics in the mulching cultivated soil? A preliminary field study with econometric methods. *Environmental Pollution*, 2022, **299**: 118915
39. Wik A, Dave G. Occurrence and effects of tire wear particles in the environment—A critical review and an initial risk assessment. *Environmental Pollution*, 2009, **157**(1): 1–11
40. Kreider M L, Panko J M, McAtee B L, Sweet L I, Finley B L. Physical and chemical characterization of tire-related particles: comparison of particles generated using different methodologies. *Science of the Total Environment*, 2010, **408**(3): 652–659
41. Bläsing M, Amelung W. Plastics in soil: analytical methods and possible sources. *Science of the Total Environment*, 2018, **612**: 422–435
42. Wang W F, Ndungu A W, Li Z, Wang J. Microplastics pollution in inland freshwaters of China: a case study in urban surface waters of Wuhan, China. *Science of the Total Environment*, 2017, **575**: 1369–1374
43. Zhang H G, Sun G S, Sun L, Zhou Z F, Zhang S K, Sui H. Preliminary study on phthalic acid esters pollution of typical plastic mulched crops soils. *Environmental Monitoring in China*, 2013, **29**: 60–63 (in Chinese)
44. Guo D M, Wu Y. Determination of phthalic acid esters of soil in South of Xinjiang cotton fields. *Arid Environmental Monitoring*, 2011, **25**(2): 76–79
45. Meng X Z, Wang Y, Xiang N, Chen L, Liu Z G, Wu B, Dai X H, Zhang Y H, Xie Z Y, Ebinghaus R. Flow of sewage sludge-borne phthalate esters (PAEs) from human release to human intake: Implication for risk assessment of sludge applied to soil. *Science of the Total Environment*, 2014, **476–477**: 242–249
46. Wang J, Luo Y M, Teng Y, Ma W T, Christie P, Li Z G. Soil contamination by phthalate esters in Chinese intensive vegetable production systems with different modes of use of plastic film. *Environmental Pollution*, 2013, **180**: 265–273
47. Wang J, Lv S H, Zhang M Y, Chen G C, Zhu T B, Zhang S, Teng Y, Christie P, Luo Y M. Effects of plastic film residues on occurrence of phthalates and microbial activity in soil. *Chemosphere*, 2016, **151**: 171–177
48. Staples C A, Peterson D R, Parkerton T F, Adams W J. The environmental fate of phthalate esters: a literature review. *Chemosphere*, 1997, **35**(4): 667–749
49. Cheng J J, Liu Y A, Wan Q, Yuan L, Yu X Y. Degradation of dibutyl phthalate in two contrasting agricultural soils and its long-term effects on soil microbial community. *Science of the Total Environment*, 2018, **640–641**: 821–829
50. Gao B, Wang P, Zhou H D, Zhang Z Y, Wu F C, Jin J, Kang M J, Sun K. Sorption of phthalic acid esters in two kinds of landfill leachates by the carbonaceous sorbents. *Bioresource Technology*, 2013, **136**: 295–301
51. Mo C H, Cai Q Y, Li Y H, Zeng Q Y. Occurrence of priority organic pollutants in the fertilizers, China. *Journal of Hazardous Materials*, 2008, **152**(3): 1208–1213
52. Zhou Y Q, Liu Y X. Occurrence and fate of phthalates in wastewater treatment plants in Beijing, China. *Environmental Sciences*, 2013, **34**(4): 1357–1362
53. Wang L, Xu X, Zhu D, Li X Y, Zhang G X, Chen G, Liu Q Z, Lin L H, Wang D H. The occurrence and ecological risk assessment of typical organic pollutants in the effluents of urban reclaimed water plants. *Acta Scientiae Circumstantiae*, 2021, **41**(5): 1910–1919 (in Chinese)
54. Li X X, Wang Q, Jiang N, Lv H J, Liang C L, Yang H Y, Yao X F, Wang J. Occurrence, source, ecological risk, and mitigation of phthalates (PAEs) in agricultural soils and the environment: a review. *Environmental Research*, 2023, **220**: 115196
55. Chang H, Ma Y J, Jiang J H, Yan Y N, Tang Z Y, Mao W B, Wang G Y, Dong J, Zhao W C, Gao X, Li X Y. Plasticizer contamination in irrigation water: residue characteristics and toxic effects of DBP and DEHP on wheat growth and metabolism. *Ecotoxicology and Environmental Safety*, 2025, **301**: 118510