



Abundance and morphology of microplastics in an agricultural soil following long-term repeated application of pig manure[☆]



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ABSTRACT

Microplastics occur widely in the terrestrial environment and they currently occur in organic fertilizers applied to agricultural land. However, there is little information available on the accumulation of microplastics in soils fertilized over the long term. Here, we investigate the characteristics of microplastics in both pig manure and soil following long-term manure application in an attempt to assess their accumulation and the potential risk to agricultural soils of repeated application of pig manure. Microplastics were separated from soil and pig manure samples using a sequential flow separation and flotation method. The abundances of microplastics were 16.4 ± 2.7 and 43.8 ± 16.2 particles kg^{-1} in control plots (CK, no manure applied) and plots amended annually with pig manure for 22 years (PM), respectively. The microplastics (especially fragments) were significantly enriched in PM-amended soil compared with the control plots. The average annual abundance of microplastics was 1250 ± 640 particles kg^{-1} in manure. Interestingly, the type and polymer composition of microplastics were very similar in the soil and manure. Differences in color and particle size indicate that microplastics sourced from pig manure may be gradually weathered and degraded after incorporation into the soil. The average accumulation rate of microplastics in the agricultural soil with long-term application of pig manure was estimated to be 3.50 ± 1.71 million particles $\text{ha}^{-1} \text{a}^{-1}$. The microplastics in the manured soil displayed complicated weathered surfaces. The presence of carbonyl groups suggests that the weathered microplastics in soil may have the potential to adsorb contaminants.

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1. Introduction

The widespread occurrence of microplastics in terrestrial ecosystems is of great public concern. The amount of plastics released annually to soils has been estimated to be 4–23 times higher than that released to the oceans (Horton et al., 2017). Agricultural ecosystems are especially likely to be contaminated with microplastics because of the multiple sources of plastics used in agricultural practice (Ng et al., 2018). The application of sewage sludges in agriculture is one example because most of the microplastics

present are retained in sludges after sewage treatment. Repeated application of sludge to agricultural land inevitably results in the accumulation of microplastics in the soil (Li et al., 2018; Berg et al., 2020). The abundance of microplastics in agricultural soil was up to 600–10,400 particles kg^{-1} and increased with increasing frequency of application in Chile (Corradini et al., 2019). Widely-used plastic mulch films are also an important source of microplastics in agricultural soils (Huang et al., 2020) and the residual film can be further fragmented into microplastics. For example, agricultural soils on the coastal plain of Hangzhou Bay in east China showed a higher average abundance of microplastics in the surface of mulched soils than in non-mulched soils with 571 and 263 particles kg^{-1} , respectively (Zhou et al., 2020). In general, microplastics in agriculture are mainly derived from biosolids, organic manures, polyethylene mulching films, atmospheric deposition, irrigation, and surface runoff (Luo et al., 2018). However, little is known about

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the accumulation of microplastics in organic soils receiving repeated applications of organic manures.

Organic manures are important sources of plant nutrients in agricultural production and they can also improve and maintain soil quality (Ro et al., 2016; Karimi et al., 2018). Furthermore, long-term manure application increases the abundance of bacteria, fungi, and other microorganisms (Wang et al., 2020). However, repeated application of organic manures can result in soil pollution with potentially toxic elements and antibiotic residues (Guo et al., 2018). The (micro)plastic pollution of organic fertilizers has usually been ignored. Gajst (2016) found up to 1200 mg plastics kg^{-1} in organic compost in Slovenia. Bläsing and Amelung (2018) estimated that the amount of visible plastic discharged into farmland with an annual application of compost would be 0.016–1.2 kg ha^{-1} (based on an annual rate of compost application of 7 t ha^{-1}). Surprisingly, most countries allow the application of plastics in organic fertilizers which are allowed to contain 0.1 wt% of plastics in Germany, one of the countries with the most stringent quality control of organic fertilizers (Weithmann et al., 2018). Microplastics may be more abundant than plastics in organic fertilizers. The source of the (micro)plastics in manure may be feces and the surrounding environment during the composting process. Previous studies show that microplastics occur in animal and human feces (Yan et al., 2020). In addition, microplastics can be transported in air and deposition in outdoor and indoor environments (Enyoh et al., 2019). Evaluation of the impact of the application of organic fertilizers on the accumulation of microplastics in agricultural land is therefore urgently required.

Here, we investigate the abundance, distribution, and morphology of microplastics in 'red soil' (Acrisols according to the FAO classification system) with repeated application of pig manure.

The aim is to understand the accumulation and surface weathering of microplastics in agricultural land following long-term repeated application of pig manure.

2. Materials and methods

2.1. Site description and sample collection

The long-term field plots were established in 1996 at the National Agricultural Ecosystem Observation and Research Station at Yingtan, Jiangxi province, southeast China ($28^{\circ} 12' \text{ N}$, $116^{\circ} 55' \text{ E}$; Fig. 1). The test soil is derived from Quaternary red clay. The experiment contained control plots (CK, no fertilizer applied) and experimental plots to which pig manure was applied annually for 22 years (PM). The pig manure was obtained from a nearby pig farm and was applied annually at a rate of 1.69 t ha^{-1} (dry weight basis) before planting peanut. Conventional field management was carried out during the growing period.

Soil samples were collected on July 22, 2018 to a depth of ~20 cm using a steel soil sampler and roots and stones were removed. Duplicate samples were collected from randomized plots of each treatment (CK, 2 plots; manure amended soil, 10 plots). Each sample was taken randomly using a multipoint mixed method. The soil samples were homogenized in the laboratory to avoid cross-contamination. Soil Moisture content was calculated by the oven-drying method (Zhou et al., 2018) (Table S1 in Supplementary data).

2.2. Separation of microplastics

A microplastic separation method was established based on a continuous air-flow flotation separation device (Zhou et al., 2018).

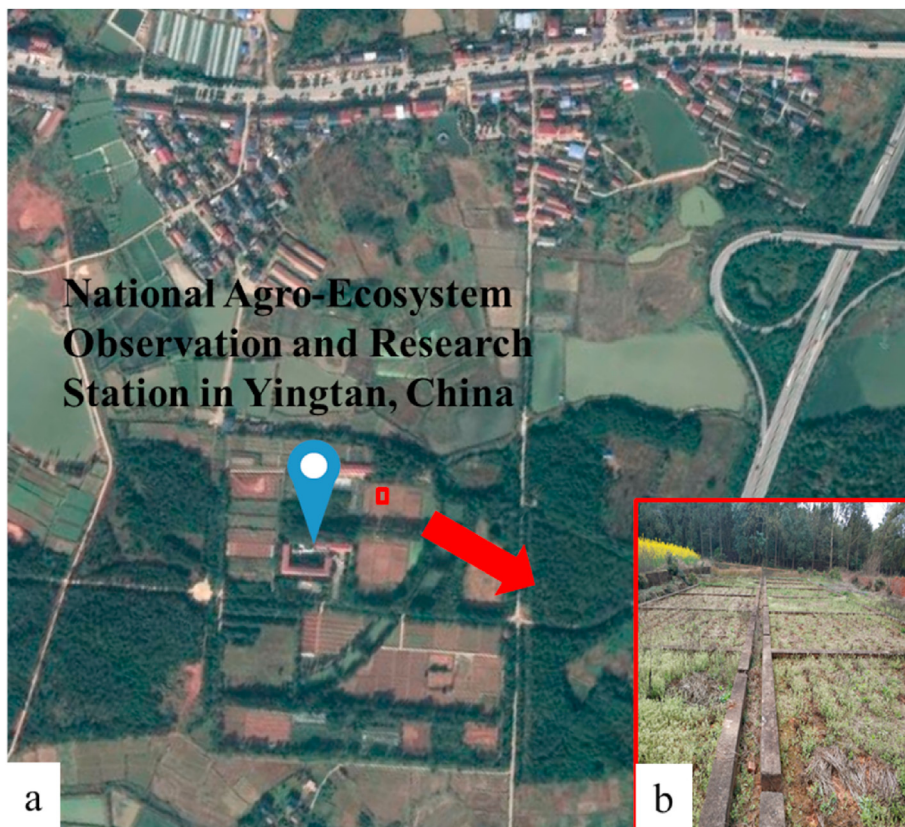


Fig. 1. (a) Satellite imagery of the field experiment established at the National Agricultural Ecosystem Observation and Research Station in Yingtan, China; (b) close-up of the long-term manure application field.

A vacuum filter pump was used instead of wet sieve filtration to save time and avoid the loss of microplastics during transfer that can occur using the traditional flotation method. The soil samples (250 g) were weighed into dry glass beakers. Saturated NaCl solution (ρ 1.20 g cm⁻³) was added and the suspensions were stirred and shaken using ultrasound for 5 min. Large clods of soil were crushed using a glass rod to mix and disperse the soil thoroughly. After density separation, the residues were further floated using a saturated NaI solution (ρ 1.60 g cm⁻³) if they contained many solid particles. Finally, the sample filtered with a 20- μ m pore nylon fiber membrane (Millipore, Burlington, MA), washed with 30% H₂O₂ to dissolve the organic matter on the electric hotplate (72 h, 60 °C). The samples were observed after filtration.

The same separation method was used for dried pig manure samples collected from 2011 to 2018 (2014–2016 samples were unavailable or contaminated). Due to the small amount of stored pig manure samples, duplicate 5-g manure samples were weighed into dry glass beakers. The pig manure did not contain solid soil particles and secondary flotation in saturated NaI solution was not required. There were more residues in the manure than in the soil. Particular care was taken to pick out suspected microplastic particles using microscopic observation.

2.3. Microplastic identification and analysis

Macroplastics (>5 mm) were excluded from the study. Microplastics were counted and measured after flotation using a stereomicroscope (Model S9i, Leica AG, Wetzlar, Germany). Microplastics (2–5 mm, not including fibers) were identified using Fourier transform infrared spectrometry (Nicolet iS10, Thermo Fisher, Waltham, MA) fitted with an ATR accessory (ATR-FTIR). The spectral range was 650–4000 cm⁻¹ at 32 scans. Representative particles (<2 mm) and fibers were identified by μ -FTIR (Nicolet iN10, Thermo Fisher, Waltham, MA). The spectral range was set at 650–4000 cm⁻¹ and samples were scanned 16 times. A scanning electron microscope (SEM) (S-4800, Hitachi, Tokyo, Japan) was used to observe the surface morphology of the microplastics, and the elemental composition of selected surface micro-domains was analyzed by combination with an energy dispersive spectrometer (EX-350, Horiba, Kyoto, Japan).

The microplastics were classified into four categories based on their characteristics and morphology, namely fibers (elongated strings), fragments (hard angular pieces), films (soft transparent flakes), and granules (irregular stereo particles) (Fig. S1). Here, the category polypropylene comprised poly(ethylene)propylene copolymers, polyethylene consisted of low-density and high-density polyethylene, and polyester (PES) encompassed polyester fibers and polyethylene terephthalate (PET).

2.4. Statistical analysis

Microplastics were counted according to color, size, composition, and type. The abundance of microplastics in the soil and manure samples was expressed as particles kg⁻¹ dry weight. The data are shown as mean \pm standard deviation and were recorded and drawn using Microsoft Excel 2016 and OriginPro 2017. The data were subjected to analysis of variance and independent sample *t*-test using the SPSS 18.0 software package and differences at *p* < 0.05 were considered to be statistically significant.

2.5. Quality assurance (QA) and quality control (QC)

Separation and extraction of microplastics were conducted in a laboratory without cross-contamination. Deionized water and dustproof clothing were used in all experiments to avoid cross-

contamination throughout the process. Two procedural blanks were set up for soil and manure samples. Occasional cotton fibers were found in the soil blanks and three polyester fibers were observed in the manure blanks. Results were corrected by subtracting background values (Table S2 and Table S4 in the Supplementary data).

A recovery experiment was set up using triplicates based on the procedures described in section 2.2 above. Representative microplastics were chosen to include analytical grade polymers (polyamide (PA), polypropylene (PP), polystyrene (PS), polyethylene (PE), polyvinyl chloride (PVC), expandable polystyrene (EPS)), PA fiber, and PE film. The average recoveries ranged from 81.7 to 100%, with PA fiber giving the lowest recoveries (Table S3 in the Supplementary data).

3. Results and discussion

3.1. Characteristics and accumulation of microplastics in the soil with repeated applications of pig manure

Microplastics were detected in all soil samples examined and transparent microplastics accounted for a larger percentage (72.8% in manured (PM) than in control plots (50.0%)) (Fig. 2 (a)). Lv et al. (2019) found that white, translucent, and black were the dominant colors of microplastics in rice-fish co-culture system soils in Shanghai, China. Another investigation found that white microplastics (62.5%) predominated in agricultural land in southeast Germany (Piehl et al., 2018). Thus, the color of microplastics in different areas and different soil uses are not consistent. Microplastics <1 mm accounted for 50.0 and 54.4%, respectively, in the control and manured plots (Fig. 2 (b)). The abundance of microplastics in the soil was inversely proportional to particle size and this agrees with previous studies (Liu et al., 2018; Zhou et al., 2018). Microplastics with smaller particle sizes (<1 mm) might be more readily ingested by soil organisms (Lwanga et al., 2016). A study of vegetable fields in suburban Wuhan in central China found that 95% of the microplastics were < 1 mm (Chen et al., 2020). Zhang and Liu (2018) also found that 95% of the microplastics in an arable soil near Dian Lake in southwest China were <1 mm.

Four microplastic polymers were detected after excluding cotton and other organic substances (such as sorbitan monooleate and polygalacturonic acid sodium) from the extracted fibers and particles. These were polyester (PES), polyethylene (PE), polypropylene (PP), and rayon (Fig. S2). The percentages of microplastic polymers in the manured soil (high to low) followed the sequence PP (47.8%), PES (39.1%), rayon (7.07%), and PE (5.98%). PES, PP, and rayon also occurred in the controls (Fig. 2(c)). Rayon fibers are common in agricultural land and coastal mangrove sediments (Zhou et al., 2020a; 2020b). PP (50.5%), PE (43.4%), and PES (5.91%) occurred in a vegetable soil covered by agricultural mulching film (Liu et al., 2018). Microplastics in traditional agricultural soils also consist of polyethylene and polypropylene (Piehl et al., 2018). These microplastic types may be derived from the degradation of large pieces of conventional plastics contaminating the environment.

The abundance of microplastics was significantly different in the control and manured samples (Fig. 2(d)), with 16.4 ± 2.7 and 43.8 ± 16.2 particles kg⁻¹, respectively. Moreover, only fibers were present in control samples and the abundance of fragments in manured samples was significantly higher than in the controls. The experimental area is remote from residential areas and relatively unaffected by urban activities. This indicates that repeated applications of pig manure had significantly increased the abundance of soil microplastics. Here, the surface soil was sampled to a depth of 20 cm to better reflect the level of accumulation of microplastics in this cultivated soil. The abundance of microplastics was low

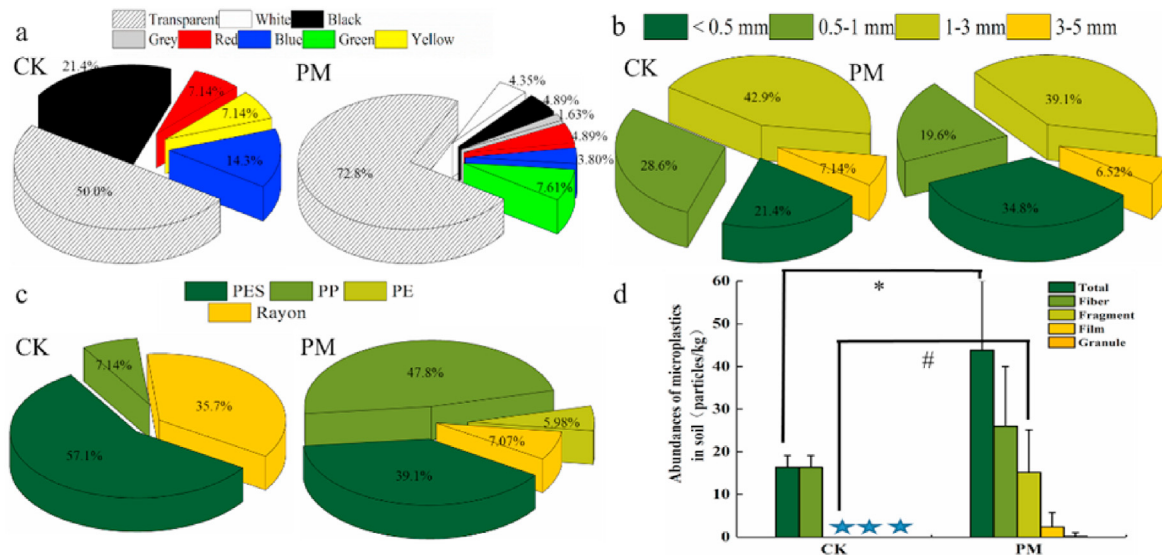


Fig. 2. Percentages of microplastics by (a) color, (b) size, and (c) composition in control and manured treatments (PP, polypropylene; PES, polyester; PE, polyethylene); (d) abundance of different shapes of microplastics in control and manured treatments (pentagram indicates 0 particles kg⁻¹; * and # represents significant differences in the abundance of microplastics between control (CK, n = 4) and manure (PM, n = 20) treatment (independent sample *t*-test, *p* < 0.05)). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

compared with other studies, perhaps due to the distance from residential areas or the relatively deep sampling. The abundance of microplastics in soil can be negatively related to sampling depth (Xu et al., 2019). The abundance of microplastics at depths 0–3 cm and 3–6 cm in an agricultural soil in suburban Shanghai was 84.8 ± 13.2 and 66.0 ± 13.9 particles kg⁻¹ dry soil (Liu et al., 2018). Piehl et al. (2018) found that the abundance of microplastics at a depth of 5 cm in agricultural land where only traditional agricultural methods were practiced was 0.34 ± 0.36 kg⁻¹. Human activities have a distinct impact on the abundance of microplastic in soils. The use of agricultural films or fertilizers will increase the abundance of microplastics in agricultural land.

Here, the average accumulation of microplastics in agricultural land via pig manure was estimated to be 1.25 ± 0.61 particles kg⁻¹a⁻¹. The soil bulk density at the experimental site is calculated at 1.40 g cm⁻³ (without regard to the effect of manure applications). The average accumulated amount of microplastics in the surface soil under repeated applications of pig manure may reach 3.50 ± 1.71 million particles ha⁻¹a⁻¹. This indicates that the organic manure is an important source of soil microplastics and is similar to the accumulation of soil microplastics from repeated applications of sewage sludge. Previous studies report that the abundance of microplastics accumulated in a Chilean soil with application of sludge ranged from 1100 to 3500 particles kg⁻¹ (Corradini et al., 2019). One recent study found that the abundance of microplastics in soil was 545.9 and 87.6 particles kg⁻¹ after repeated application of sludge compost (30 and 15 t ha⁻¹a⁻¹, respectively), values significantly higher than in the unamended soil (5 particles kg⁻¹) (Zhang et al., 2020). Very large application rates of sludge containing numerous microplastics will result in the accumulation of microplastics in the soil.

3.2. Surface morphology and attachments of microplastics in the soil with repeated application of pig manure

The surface morphology of microplastics in this soil is shown in Fig. S3. Our observations indicate that microplastics show complicated morphological features after long-term weathering in the soil. Fibers and granules exhibited protrusions and depressions on

the surfaces. The surfaces of the fragments exhibited some cracks and several filaments around the cracks. In addition, there were large micropores (~1 μm) on the surfaces of the thin films (Fig. S3 (f, i)). The surface features differ from previous studies in which strong weathering influences on microplastic surfaces were observed in coastal beach soils (Cooper et al., 2010; Zhou et al., 2018). Strongly weathered microplastics indicate that the morphologies are related to mechanical, chemical, and biological processes (Zhou et al., 2018; 2020b). The arable soil in this area may be less subject to abiotic degradation (such as UV radiation and mechanical erosion) due to the soil depth and vegetation cover. Some very small micropores and filaments in fragments and films may reflect the important role of microorganisms. Microorganisms can colonize the surfaces of films in soils (Huang et al., 2020). Zhang (2019a) also found numerous microbes inhabiting micropores on the surface of a mulching film. Microplastics have the potential to attract degradative microorganisms. It is important to conduct further research to determine whether or not organic manures lead to the formation of secondary microplastics in soils.

Elemental analysis of the surfaces of the fragments and thin films is shown in Fig. 3. The surfaces of the microplastics contained Si, Al, Fe, Mg, and other elements present in the environment as oxides. Soil mineral colloids presumably exist on the surfaces of microplastics. Jian et al. (2018) also found clay minerals attached to the surfaces of microplastics in the bottom mud of Poyang Lake. This indicates that plastic-mineral complexes occur widely in the environment. PP fragments and PE films from the long-term manured soil were analyzed by ATR-FTIR and compared with commercial PP woven bags and PE film (Fig. 4). Interestingly, peaking at 1034 cm⁻¹ was likely due to clay on the surfaces. This is consistent with the aging results of PE film in the soil of Briassoulis et al. (2015). Moreover, the hydroxyl and carbonyl absorption peaks of microplastics in the environment occur around 3450 and 1700 cm⁻¹, respectively. This suggests that the microplastic surface generates functional groups such as carboxyl, aldehyde, and hydroxyl (Fairbrother et al., 2019). The appearance of these functional groups was related to the degradation of microplastics. Microplastics may thus act as carriers to adsorb pollutants, making it easier to transfer contaminants and cause harm (Xu et al., 2019).

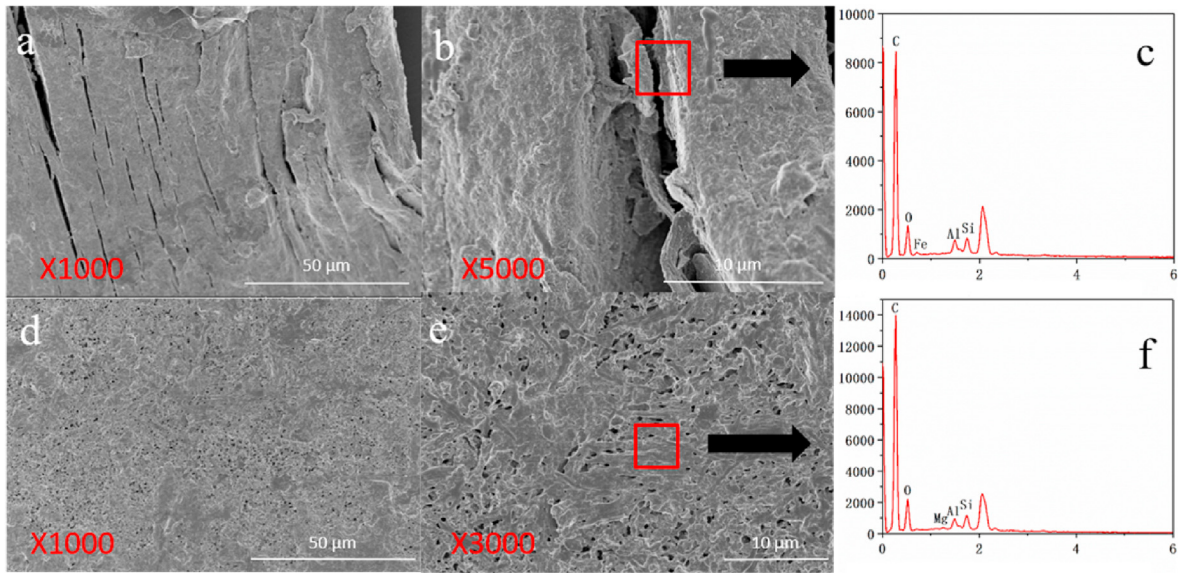


Fig. 3. SEM images (a & b, PP fragments; d & e, PE films) and the energy spectrum diagram for substances adsorbed on the surfaces (c, PP fragments; f, PE films) of microplastics from the long-term manured soil.

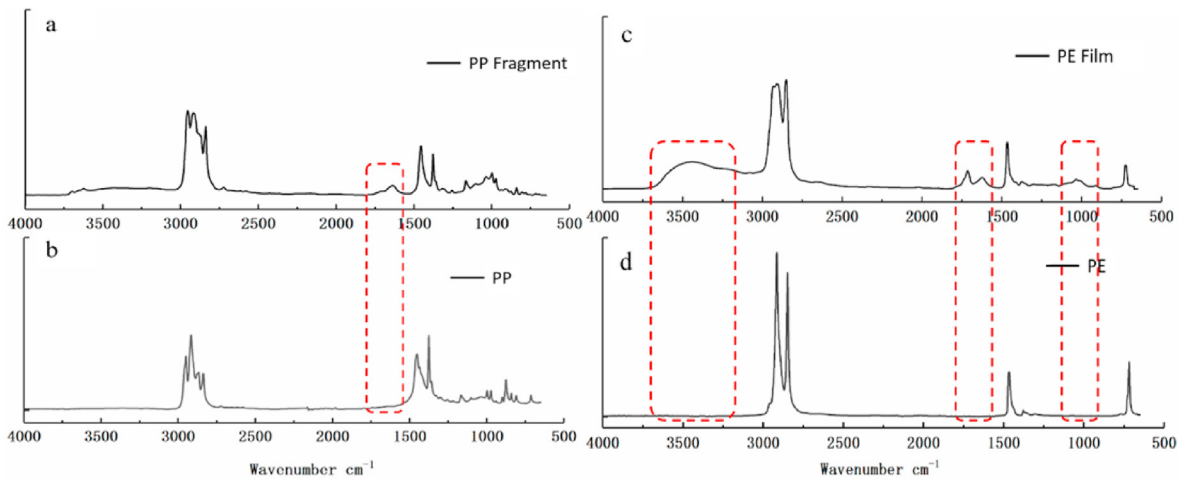


Fig. 4. Comparison of (a, b) PP fragments and (c, d) PE films from the soil with those of commercial substances by ATR-FTIR spectra.

Degraded microplastics also release more harmful additives (Bandow et al., 2017).

3.3. Characteristics of microplastics in pig manure and their contribution to microplastic accumulation in the soil

Aggravated pollution by microplastics in agricultural land from the application of pig manure was further explored. The distribution of microplastics in pig manure in different years was examined. Microplastics occurred in all manure samples examined (Fig. 5, Table S4). Fig. S4 shows photos of typical microplastics that have been extracted from pig manure. Classification by color, size, and composition (Fig. 5(a, b, c); Fig. S5) gives detailed information on the manures in different years. A total of seven colors of microplastics were found, with black (32.8%) and blue (27.2%) predominant (transparent 12.8%, red 12.8%, white 11.2%, green 1.60%, purple 1.60%). Microplastics in pig manure had a wider range of colors than those in manured soil. The average percentage of microplastics >1 mm in manure was 63.2% and was higher than in the

experimental soil. These results indicate that microplastics can be broken down and undergo abiotic and biotic degradation in the soil. Feuilloley et al. (2005) found PE plastic film breaking down into smaller plastics in the natural environment. The composition of microplastics from pig manure was similar to that from the soil (Fig. 5(c)), comprising mainly PP, PES, and small amounts of PE and rayon). Weithmann et al. (2018) found PES, PP, and PE in organic fertilizer. These are also the most commonly used plastics (PlasticsEurope, 2019). The experimental plots, far from residential areas, had a similar composition indirectly indicating that microplastics in the soil were derived from the pig manure.

The average annual abundance of microplastics was 1250 ± 640 particles kg⁻¹ in manure (Fig. 5 (d)). The percentage of fibers in pig manure was relatively high (79.2%). The average input of microplastics from pig manure was estimated to reach 2.11 ± 1.08 million particles ha⁻¹ a⁻¹. This is broadly consistent with the calculated average amount of microplastics accumulated in the surface soil. The long-term application of pig manure would have made the dominant contribution to the microplastic abundance of the soil in

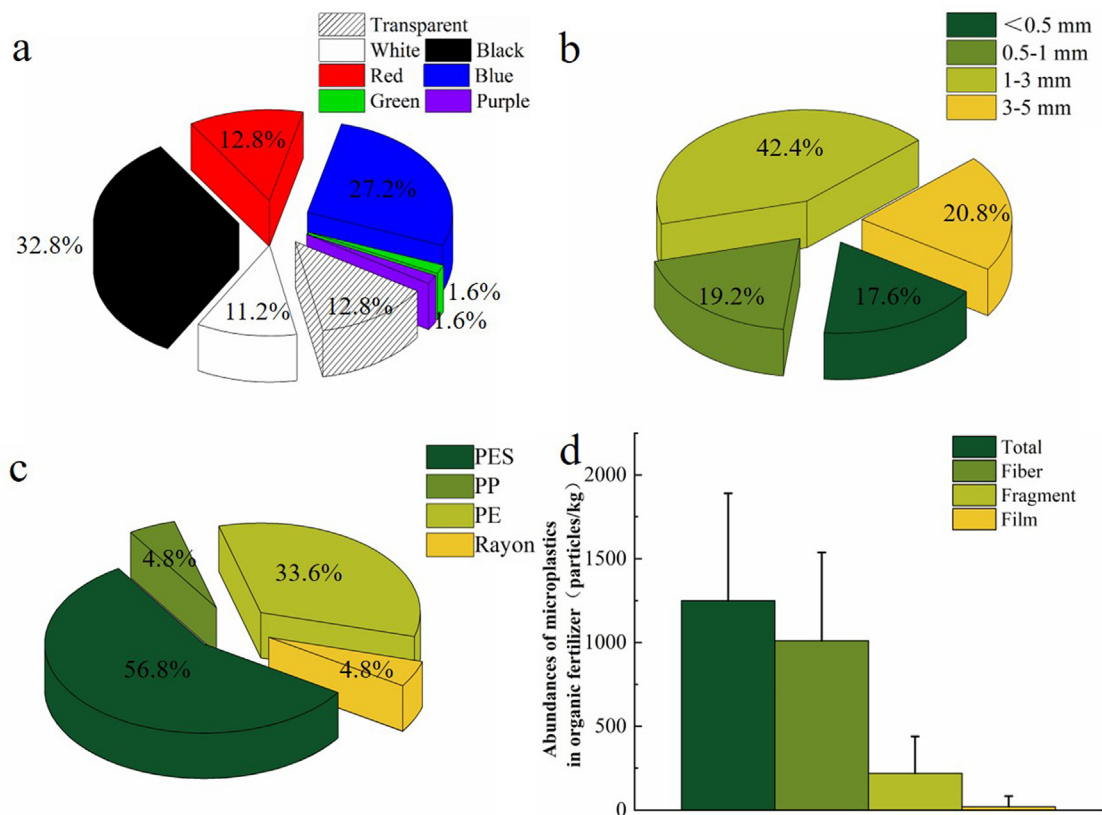


Fig. 5. Average percentages of microplastics in manure by (a) color, (b) size, and (c) composition; (d) average abundance of microplastics in the pig manure. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

this area. Weithmann et al. (2018) investigated different organic fertilizers and found that the abundance of microplastics (1–5 mm) in biowaste digestion was 14–895 particles kg⁻¹. Some of the microplastics in manure may be derived from feces. Liu et al. (2019) reported that microplastics in feces of birds (Poyang Lake, China) reached 4.93 ± 4.25 particles g⁻¹. Another investigation of chicken feces found an abundance of microplastics of up to 129.8 ± 82.3 particles g⁻¹ (Lwanga et al., 2017). The occurrence of microplastics in feces indicates that they have entered the bodies of animals and humans, possibly from contaminated air, food, and drinking water (Yan et al., 2020). In addition, microplastics are widespread in the air (Enyoh et al., 2019). Exposure to air during composting is another potential source of microplastics in manure. Although fibers and fragments found in pig manure differed in different years (Fig. S5(d) and Fig. S6.), only the fragments in soil receiving repeated applications of sludge showed significant differences from unamended soil. Fibers may more difficult to separate from agricultural soil aggregates than are fragments due to the linear shape of fibers entangling soil aggregates (Zhang et al., 2019b).

4. Conclusions

This is the first report showing the distribution of microplastics in soil following long-term repeated application of pig manure. Manure application significantly enriched the soil with microplastics compared with unamended soil. Differences in the color, particle size and consistency of composition suggest that microplastics may break down to form smaller particles when pig manure is applied repeatedly to agricultural land. Moreover,

surface morphology indicates that microorganisms may play an important role in the degradation of microplastics. The appearance of oxygen-containing functional groups and clay minerals may result in microplastics more readily adsorbing pollutants and acting as carriers of pollutants. Clearly, the contamination of soils by microplastics via manures is an important topic that merits further investigation.

Author statement

Jie Yang: Data curation, Methodology, Writing original draft, Visualization, Formal analysis. **Ruijie Li:** Data curation, Investigation, Methodology. **Qian Zhou:** Investigation, Methodology. **Lianzhen Li:** Writing - review & editing. **Yuan Li:** Writing - review & editing. **Chen Tu:** Writing - review & editing. **Xinyue Zhao:** Investigation. **Kuanxu Xiong:** Software. **Peter Christie:** Writing - review & editing. **Yongming Luo:** Conceptualization, Supervision, Writing - review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no conflicts of interest.

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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.2020.116028>.

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