Science of the Total Environment 703 (2020) 134722

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Review

Behavior of microplastics and plastic film residues in the soil environment: A critical review

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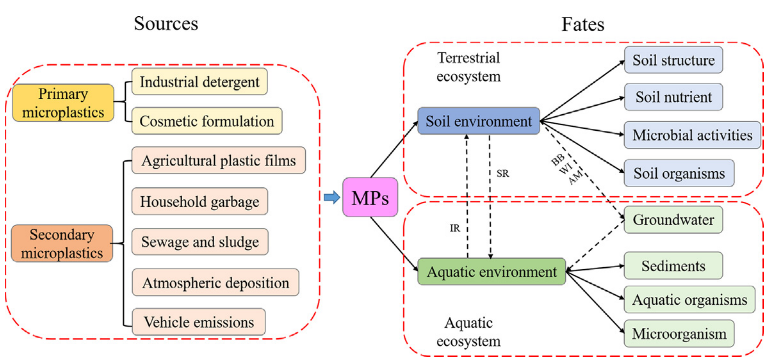
HIGHLIGHTS

- Micro- and nano-plastics in agricultural soils represents a major environmental threat.
- Standard methods for microplastic separation and detection are urgently needed.
- Microplastics affect agroecosystem functioning by accumulating heavy metals or organic contaminants.
- Plasticizers represent a major source of pollution within microplastics.
- Adoption of biodegradable films will help reduce soil microplastic accumulation.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 18 July 2019
Received in revised form 23 September 2019
Accepted 27 September 2019
Available online 17 October 2019

Editor: Jay Gan

Keywords:
Environmental risk
Nanoplastics
Soil heath
Mulching film
Plasticizer

ABSTRACT

It is now widely acknowledged that microplastic pollution represents one of the greatest anthropogenically mediated threats to Earth-system functioning. In freshwater and marine ecosystems the presence of large amounts of microplastic appears almost ubiquitous, with frequent reports of negative impacts on aquatic health. In contrast, however, the impact of plastic in terrestrial environments remains poorly understood. In agroecosystems, microplastics (particles < 5 mm) can enter the soil environment either directly (e.g. from biosolids application, irrigation water, atmospheric deposition), or indirectly through the *in situ* degradation of large pieces of plastic (e.g. from plastic mulch films). Although we have encouraged the use of plastics over the last 50 years in agriculture to promote greater resource use efficiency and food security, the legacy of this is that many soils are now contaminated with large amounts of plastic residue (ca. 50–250 kg ha^{-1}). Due to difficulties in separating and quantifying plastic particles from soil, our knowledge of their behavior, fate and potential to transfer to other receptors (e.g. surface and groundwater, air) and enter the human food chain remains poor. This information, however, is critical for evaluating the risk of soil-borne microplastic pollution. In this critical review, we systematically summarize (i) the distribution and migration of microplastics in soils, (ii) highlight the separation, extraction, and identification methods for monitoring microplastics in soils, (iii) discuss the ecological effects and pollution mechanisms of soil microplastics, (iv) propose mitigation strategies to help prevent and reduce microplastic pollution, and (v) identify the most important future challenges in soil microplastics research.

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https://doi.org/10.1016/j.scitotenv.2019.134722
0048-9697/

Contents

1. Introduction

Microplastics have often been defined as particles smaller than 5 mm in size. These mainly originate from tiny plastic particles (such as abrasives in detergents and cosmetics) that are released directly into the environment, or indirectly from the degradation of large piece plastics (such as plastic film, household garbage, atmospheric deposition, and vehicle emissions) (Andrady, 2011; Cole et al., 2011; Gesamp, 2015). Microplastics are now recognized as an important environmental pollutant, being almost ubiquitous in the atmosphere, water, soil, and other environmental media. Due to their small particle size and very slow biodegradation rate they can be easily absorbed by organisms and subsequently transported through food webs (Horton et al., 2017a; Hurley and Nizzetto, 2018; Liu et al., 2018; Rillig, 2012). In addition, they may act as a vector for other contaminants (e.g. human pathogens, organic pollutants, heavy metals). For these reasons, microplastic pollution was recently listed as one of the top 10 environmental problems by the United Nations Environment Programme (UNEP, 2014). Consequently, plastic pollution is now considered to be a major factor responsible for the global decline in biodiversity and represents a major threat to the Earth-system functioning and human health (Gall and Thompson, 2015).

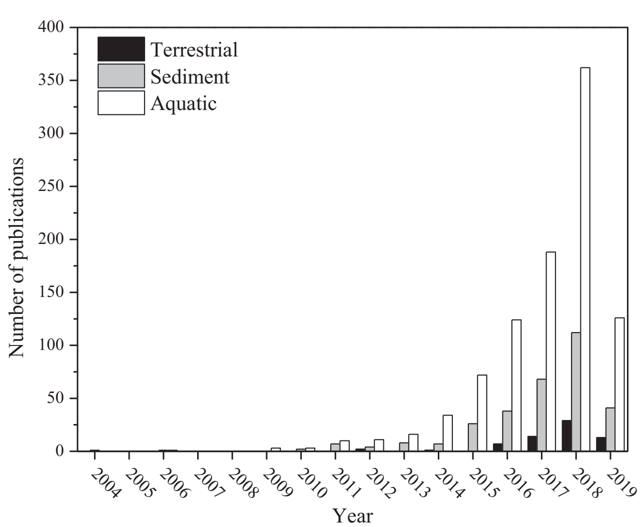

Fig. 1. Global scientific publications on microplastic pollution after the concept of microplastic was first proposed by Richard C. Thompson in 2004. The bars represent the annual number of papers published in each research area from 2004 to February of 2019 (Scopus database).

Microplastics are emerging pollutants that have been extensively detected in aquatic ecosystems, especially oceans. However, there is a knowledge gap regarding microplastic pollution in agricultural soils and terrestrial ecosystems (Horton et al., 2017a; Huerta Lwanga et al., 2016). We surveyed the publications related to microplastic contaminants in the Scopus database (https://www.scopus.com/) using the keywords "microplastic or microplastics" in combination with "terrestrial or soil" and "sediment, beach, or sludge" and "water, river, lake, sea, ocean, or marine." The literature retrieved included 1331 publications covering the period between 2004 and February 2019. Among these publications, 71% focused on marine environments, freshwater lakes, rivers, and other aquatic ecosystems, 24% concentrated on sediments (from aquatic environments, beaches, and sludge), and 5% were devoted to terrestrial ecosystems (Fig. 1). Since Rillig (2012) identified the problem of microplastic pollution in the soil environment, increasing attention has been paid to plastic pollution in soils and the potential dangers of soil microplastics (de Souza Machado et al., 2018; Horton et al., 2017; Huerta Lwanga et al., 2016; Nizzetto et al., 2016; Scheurer and Bigalke, 2018; Yang et al., 2018; Zhang and Liu, 2018). The number of studies on microplastics related to terrestrial ecosystems is increasing compared to a previous surveys, however, it is still lagging behind work in aquatic habitats (Andrés Rodríguez-Seijo, 2018).

Globally, the most frequently polymers found in the soil environment are polyethylene and polypropylene with lesser amounts of polyvinyl chloride and polyethylene terephthalate also present. Although these plastic polymers are relatively simple in structure and may be relatively environmentally benign, plastics may also contain a wide range of additives which may greatly enhance their ecological toxicity (Koelmans et al., 2019). For example, plasticizers such as phthalic acid esters (PAEs) represent a main additive of plastic films commonly used in agriculture and have been implicated in the contamination of vegetables and fruits (He et al., 2014; Kong et al., 2012). After entering the soil, macroplastic residues typically disintegrate into micro- and nano-plastics and absorb a variety of heavy metals or release organic pollutants into the soil, especially PAEs, which pose potential risks to soil biology and human health (Kasirajan and Ngouajio, 2012; Steinmetz et al., 2016; Wang et al., 2013, 2016). Interestingly, however, the presence of microplastics and PAEs have been reported in agricultural soils where microplastic-containing fertilizers and agricultural plastics have never been used (Kong et al., 2012; Piehl et al., 2018). It is still unclear therefore whether microplastics and PAEs in soil and crop products actually originate from plastic mulching film and whether they pose an actual risk to soil organisms and soil quality.

It is necessary to systematically study the changes and safety of microplastics in soils where plastic mulching film has been intensively used in agriculture. In this context, the main objectives of this study were to (1) summarize the distribution and migration of microplastics in soils; (2) highlight the separation, extraction, and identification methods for monitoring microplastics in soils; (3) discuss the ecological effects and pollution mechanisms of soil microplastics; (4) identify potential solutions to mitigate microplastic pollution; and (5) identify the future challenges in soil microplastics research.

2. Classification and migration of microplastics in soil

2.1. Classification and source of microplastics in soil

Microplastics can be divided into primary microplastics and secondary microplastics based on the original manufactured particle size. Primary microplastics mainly include plastic microbeads and nanoparticles directly used in a variety of industrial processes, such as industrial detergents and cosmetics. In addition, they may enter soil from atmospheric deposition (Allen et al., 2019). Secondary microplastics originate from large plastic products that have broken down *in situ* (e.g. plastic film residues, household garbage). This may occur at the soil surface in response to solar UV irradiation or within the soil profile due to physical abrasion (abiotic) and biological attack (Andrady, 2011; Cole et al., 2011; Gesamp, 2015). The types of microplastics can be divided into fibers, fragments, thin films, and particles depending on the plastic shape. Depending on the source, fibers often represent the predominant form if they enter soil from biosolids or irrigation waters derived from municipal wastewater (Jabeen et al., 2017; Liu et al., 2018). In contrast, the breakdown of plastic mulch films leads to a predominance of heterogeneous fragments, while plastic coated fertilizers leads to a predominance of thin films. Microplastics are further divided into small microplastics (<1 mm), medium microplastics (1–3 mm), and large microplastics (3–5 mm) according to their particle size (Andrady, 2011; Andrés Rodríguez-Seijo, 2018; Gesamp, 2015; Horton et al., 2017a; Liu et al., 2018; Rillig, 2012). Nanoplastics are typically referred to as being 1–1000 μm in size, while picoplastics are <1 μm in size. The reason that categorizing microplastic size is important is that it affects their potential for transport in soil and their potential to be taken up by cells. For example, microplastics >150 μm are unlikely to be taken up by most plants and soil organisms (with the exception of mesofauna) and are not thought to pose a risk to human health (WHO, 2019). In contrast, nanoplastics are more likely to pose an environmental risk as they have the potential to be taken into cells in a similar way to other nanoparticles (e.g. by endocytosis; Kuhn et al., 2014). In the case of nanoparticles, the rate of uptake is known to be dependent on the size, shape and surface chemistry of the material (Ma et al., 2013). Unfortunately, nanoplastics are rarely quantified in soil and their uptake into plants and soil microorgansims has not been evaluated from a risk assessment perspective. However, based on studies in marine systems we expect this exposure route to be significant (Al-Sid-Cheikh et al., 2018).

The main sources of macro- and microplastics entering agricultural soils includes plastic mulch films, municipal waste (e.g. municipal solid waste, compost), biosolids (sewage sludge and anaerobic digestate), plastic-coated fertilizers and atmospheric deposition (Andrés Rodríguez-Seijo, 2018; Blasing and Amelung, 2018; Liu et al., 2018; McCormick et al., 2014; Nizzetto et al., 2016b). Of these, agricultural films and compost application are the probably the most important (Blasing and Amelung, 2018; Hurley and Nizzetto, 2018). Plastic mulching is an important technique used to promote agricultural production in many regions of the world. Specifically, plastic mulches greatly enhance water and nutrient resource efficiency as well as providing thermal insulation and early planting and/or harvest cropping (Yin et al., 2014; Liu et al., 2019; Gao et al., 2019). In addition, plastic mulch films may reduce soil erosion and reduce the disease burden of the crop and allow the more efficient use of pesticides (Yan et al., 2010; Ruíz-Machuca et al., 2015). It is therefore not surprising that the use of plastic mulch films has been widely promoted by industry and agri-extension agencies to promote greater food security, sustainable food production and improve livelihoods (Liu et al., 2014; Yan et al., 2014; Steinmetz et al., 2016). However, in the long-term, the application of plastic film may cause serious pollution problems. One of the key debates is therefore whether the short term gains from using plastic films to promote food security outweigh the potential long-term risk to soil health. One of the major issues is that these films are extremely thin (ca. 8–50 μm thick) making their physical extraction from soil at the end of the growing season very difficult (Liu et al., 2013). In addition, there is a lack of recycling facilities capable of handling soil-contaminated plastic making recovery from the soil uneconomic as well as impractical. Inevitably, this has led to the progressive accumulation of large amounts of residual film in farmland (Fig. 2). Aided by tillage, UV irradiation and biodegradation, this residual mulch slowly fragments forming a continuum of macro, micro and nano plastics in soil (Ramos et al., 2015; Steinmetz et al., 2016).

The recycling of biosolids to land is widely advocated as a way of closing the nutrient cycling loop as well as replenishing organic matter in highly intensive cropping systems (Singh and Agrawal, 2008; Sullivan, 2015). While it is known that biosolids may contain a range of metals and organic pollutants (Smith, 2009; Semblante et al., 2015; Sharma et al., 2017), it is now becoming evident that they may contain significant amounts of plastic pollution (Gatidou et al., 2019). Typically, between 70 and 99% of the microplastics present in domestic wastewater are recovered in the sludge fraction during water treatment (Carr et al., 2016) leading to microplastic concentrations in sludge of 10^3–10^5 particles kg^{-1}. Consequently, large amount of microplastics will accumulate in the soil, particularly after repeated applications of sewage sludge to agricultural land (Andrés Rodríguez-Seijo, 2018; Nizzetto et al., 2016b). One of the key debates is therefore whether the effective recycling of nutrients and organic matter back to land via biosolids outweighs the risk of plastic contamination.

Municipal solid waste landfills may also represent point sources of microplastic pollution affecting the underlying soil and groundwater (Andrés Rodríguez-Seijo, 2018; Duis and Coors, 2016; Hopewell et al., 2009; Zubris and Richards, 2005). In the leachate fraction, microplastic particles range from 100 to 1000 μm in size with a concentration of 1–25 particles l^{-1} (He et al., 2019). As landfill leachate is rarely applied to agricultural land this probably represents a minor source of contamination globally (Jones et al., 2006).

2.2. The distribution and migration of microplastics in soil

With the development of the plastics industry, the global production of plastics has rapidly increased from 1.5 million tons in 1950 to 348 million tons in 2017 (Liu et al., 2018; Statista, 2018; PlasticsEurope, 2018). Since 1950, the total cumulative global production of plastic has been estimated at ca.10 billion tonnes, of which 55% has been sent to landfill or discarded either on land

Fig. 2. Large amount of plastic mulching film residues in farmland in China. A and B show recycled film residues, while C and D show residual film in agricultural soils. The photos were taken in Gansu Province by Yan Changrong in 2018.

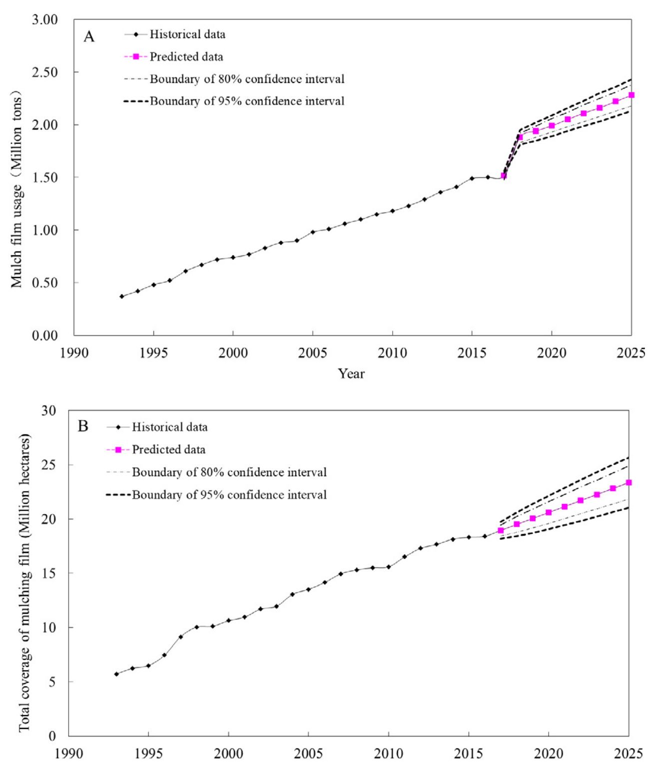

Fig. 3. Past and future prediction of the application of plastic mulching film to agricultural land in China according to the Autoregressive Integrated Moving Average model. A and B indicate the amount and coverage areas of plastic mulching film, respectively.

or in the oceans (Geyer et al., 2017; Roland et al., 2017). At the global level, best estimates suggest that approximately 80% of ocean plastics come from land-based sources (Li et al., 2016a).

Although packaging represents the biggest consumer of plastic (ca. 150 million tonnes y^{-1}), the global consumption of plastics by agriculture is also significant at 8 million tonnes per year with an estimated annual market value of $6 billion (Scarascia-Mugnozza et al., 2011). Of this, approximately 427 and 300 thousand tons of plastic mulching are used each year to cover farmland in Europe and North America, respectively (Nizzetto et al., 2016b,c). China also represents one of the largest producers and consumers of plastics in the world, accounting for around 30% of global use. The use of agricultural film in China (including mulching film and greenhouse film) has now reached up to 2.6 million tons y^{-1}, of which mulching film accounts for 1.5 million tons covering a total area of 18.4 million ha (Yan et al., 2014; National Bureau of Statistics of China, 2017). Using an Autoregressive Integrated Moving Average model we have predicted the future amount and coverage of plastic mulching film in China (Fig. 3). Our findings suggest that the use of plastic mulching film will reach 1.99 million tons in 2020 and 2.28 million tons by 2025 (Fig. 3A). We also predict that the area covered by mulching film in China will continue to increase, and will reach 21.0 million ha in 2020 and 23.4 million ha by 2025 (Fig. 3B). As it is impossible to completely remove plastic films from soil, and the rate of degradation is very slow, this will inevitably lead to a progressive accumulation of plastic in soil (Liu et al., 2014). An investigation of the major plastic mulching film usage areas in China has shown that the most severe pollution occurred in cotton fields in Xinjiang Province, with an average plastic film residue level in soil of 259 kg ha^{-1} and maximum plastic film residue of 381 kg ha^{-1} in some areas (Yan et al., 2014). Large amounts of macroplastic residues also inevitably leads to a significant accumulation of microplastics in agricultural soils over time (Ramos et al., 2015; Steinmetz et al., 2016). In the case of biosolids, assuming an average microplastics contamination of 10^4 -particles kg^{-1} and a typical land application rate of 1–15 t ha^{-1} y^{-1} this would add between 10^6 and 10^9 particles ha^{-1} y^{-1} leading to a topsoil contamination level of ca. 4–150 particles/kg for each year of application. It is therefore not surprising that contamination level of 670 fibers kg^{-1} have been reported in the topsoils from

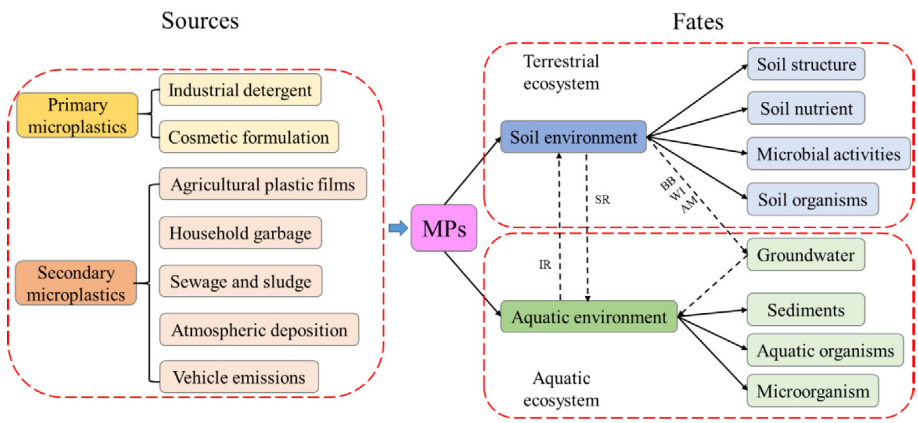

Fig. 4. Schematic showing the main sources and fates of microplastics in the environment. *Notes:* MPs represents microplastics; BB represents bioturbation; WI represents water infiltration; AM represents agricultural management; IR represents irrigation and SR represents surface runoff.

European farmland (Barnes et al., 2009; Ren et al., 2018), and that microplastics have been detected in 90% of Swiss floodplain soils (Scheurer and Bigalke, 2018). It should also be noted that the abundance of microplastics in the terrestrial environment is much higher than that in marine ecosystems (Horton et al., 2017a; Nizzetto et al., 2016b; van Sebille et al., 2015).

Many studies have documented the distribution and pollution mechanisms of microplastics in coastal and marine environments (Bergmann et al., 2017; Cole et al., 2011), particularly in lakeside and coastal areas where there is a high intensity of human activity (Duis and Coors, 2016; Horton et al., 2017b; Jabeen et al., 2017; Wang et al., 2017; Zhou et al., 2018). However, there are few studies on the distribution and migration of microplastics from agricultural soils into the wider environment (de Souza Machado et al., 2018; Zhou et al., 2018); several studies have shown that the abundance of microplastics varies at different soil depths, reaching up to 7% by weight in highly polluted topsoils (Fuller and Gautam, 2016; Liu et al., 2018). Liu et al. (2018) showed that microplastics were found in both shallow and deep soils from a range of agricultural sites in China, but that the abundance of microplastics in shallow soils was much higher than that in deep soils.

Soil is not only a sink of microplastics, but may also represent a source of microplastics to groundwater and the aquatic environment (Fig. 4). This risk of loss is expected to be much greater in agricultural soils with artificial drainage, large amounts of macropores and when surface runoff occurs. Currently, microplastic contamination of groundwaters are low but have shown to reach levels of 12 particles l^{-1} suggesting that transfer does occur (Panno et al., 2019). However, it is likely that this contamination may also be derived from human-derived wastewater (e.g. septic tanks) rather than from agricultural plastics. Microplastics can be also transferred directly through the soil via bioturbation (Rillig, 2012; Rillig et al., 2017a, 2017b), tillage operations (Liu et al., 2018), and water infiltration (Luo et al., 2018). In addition, losses from soil may occur via wind erosion, surface runoff or during crop offtake (Zhang et al., 2018b). Microplastics can also be transported and dispersed by soil animals and livestock, either through attaching to the outside of the organism or through transfer from ingestion and defecation (Cao et al., 2017; Rillig et al., 2017b). Microplastics can also be transferred to aquatic ecosystems by surface runoff (Blasing and Amelung, 2018; Brodhagen et al., 2015; Hurley and Nizzetto, 2018; Kyrikou and Briassoulis, 2007; Steinmetz et al., 2016). The migration of microplastics through surface runoff is related to the particle size and density of the microplastic. The bulk density of common plastics typically varies from 910 to 970 kg m^{-3} depending on the nature of the material. Therefore plastics without much soil mineral contamination (density of 2650 kg m^{-3}) readily float. In addition, the migration is easier for smaller particles as there is less likelihood of physical trapping in the soil matrix or surface vegetation (Nizzetto et al., 2016a; Li et al., 2019a,b). Additionally, the shape, type, and surface characteristics of microplastics are also important factors which are likely to affect their migration in soils. Thus, it is vital to further study the weathering process, adsorption capacity, and migration of microplastics, especially those with a particle size <1 mm (Zhou et al., 2018). These studies will not only be beneficial to understanding the distribution and migration of microplastics in marine and terrestrial ecosystems, but also provide an important reference for protection and governance of marine, freshwater and terrestrial ecosystems. Given the numerous potential ways in which microplastics can move in soil, it is also critically important that we determine the quantitative importance of each pathway. This will enable the better parameterization of risk models and also the implementation of more targeted monitoring and mitigation programmes.

3. Separation and detection of microplastics in soil

Separation of microplastics, especially nanoplastic, from the soil environment is more complex than from aquatic ecosystems owing to the complexity of the soil environment and characteristics of microplastics (Alimi et al., 2018; Hurley and Nizzetto, 2018). Despite efforts to establish effective analytical procedures (Gigault et al., 2016; Velzeboer et al., 2014), the comparative analysis of microplastics in different components of the Earth system remains challenging as no unified standard methods exist for microplastic separation and identification (Song et al., 2015; Tagg et al., 2015).

In accordance with methods for separating microplastics from marine sediments and aquatic environments, microplastic separation from soil can be divided into the process of microplastics extraction and impurity removal. Microplastics extraction methods for soils includes: (1) air flotation; (2) heating (3–5 s at 130 °C); and (3) density suspension. Notable problems with these approaches, however, include: (i) the recovery rate is often not reproducible and the extraction efficiency is low; (ii) it is difficult to capture the high degree of spatial heterogeneity in soil plastic contamination; (iii) current methods are not designed to capture nano- and picoplastic particles; (iv) the heating method is not feasible for large numbers of samples and cannot readily detect the quantity and size of microplastics, which is not suitable for the analysis of samples in complex environments; (v) the density suspension method is a common method to extract microplastics from sediment and sludge, but its suitability in soil remains to be validated. One of the major challenges is the removal of other contaminants from the samples (e.g. organic matter). Impurity removal methods include: (1) acid digestion; (2) alkali digestion; (3) enzyme digestion, and (4) chemical oxidation. The problems with these approaches include: (i) some plastics react with strong acids or alkali; (ii) enzymatic digestion is not feasible for large numbers of samples and/or large-volume samples, due to prohibitive costs and is very poor at removing stable organic matter; (iii) the impact of wet oxidation on microplastics and the potential to inadvertently remove co-contaminants (e.g. plasticizers) remains to be verified.

A major consideration in microplastic research is the robust collection of the samples from the field. It is recommended that collection of samples in plastic materials be avoided where possible to minimize contamination (Koelmans et al., 2019). In addition, contamination of samples in the laboratory due to airborne polymer particles and fibers has been described as a major problem in microplastic analysis (Torre et al., 2016). Further, adequate negative controls (blanks) should be included to demonstrate absence of contamination during sample processing (e.g. during sieving, filtration, digestion, transfer and analytical identification steps; Hermsen et al., 2018). In addition, the recovery of pure microplastics of different sizes (i.e. reference standard, positive control) added to the samples should be used to validate plastic recovery.

The development of techniques for extraction and analysis of microplastics from soil media is in its infancy. Extraction using density suspension and the removal of organic matter on the surface of microplastics using a suitable oxidizer solution are the most commonly used methods for separating microplastics from soils (Liu et al., 2018; Nuelle et al., 2014; Qiu et al., 2016; Thompson et al., 2004; Zhang et al., 2018b). According to the methods for separation of microplastics from marine sediments and aquatic environments, a saturated sodium chloride solution, sodium polytungstate solution, or seawater can be used for density separation of plastic particles from soils (Scheurer and Bigalke, 2018). Sodium iodide and zinc chloride solutions are also commonly used density suspensions (Claessens et al., 2013; Corcoran et al., 2015; Fok and Cheung, 2015). However, different types of microplastics have different densities, and other interfering substances can be easily suspended in high density liquids, while some types of microplastics cannot be suspended in low density solutions. Zhang et al. (2018b) used pure water to float polyethylene and polypropylene microplastics (density 0.90–0.96 g cm^{-3}) from soils, however, this necessitates removal of mineral contaminants. Theoretically therefore, to improve the extraction efficiency of microplastics, suspension of different concentrations can be used to extract different types of microplastics from soil samples step-by-step. Nuelle et al. (2014) used an air-induced overflow method to extract microplastics from soil samples using a low-density suspension solution first, and then used a high-density solution for the subsequent flotation step. The extraction efficiency, which was dependent on the shape, size, and source of microplastics in the whole procedure, reached up to 91%–99%.

It is more difficult to extract microplastics from soil environments than from aquatic ecosystems because the surface of soil microplastics often develop biofilms that absorb impurities, such as mineral particles and organic matter. These impurities need to be removed for further studies. Solutions of acid (e.g. HCl, HNO_3, H_2SO_4), base (e.g. KOH, NaOH) and oxidants (e.g. $KMnO_4$, H_2O_2) in isolation or combination have all been used to remove surface impurities (Liu et al., 2018; Qiu et al., 2016). It was reported that a 35% H_2O_2 solution was more conducive to the removal of biological organic matter than 37% HCl and 20%, 30%, 40%, and 50% NaOH (Nuelle et al., 2014). Tagg et al. (2015) showed that a 30% H_2O_2 solution could not only effectively remove organic matter, but also improve the filtration efficiency, which is beneficial to identifying different types of microplastics by Fourier transform infrared spectrometry (FTIR). Cole et al. (2014) found that enzymatic digestion was a more effective method for the removal of organic impurities compared with acid or alkali digestion. Unfortunately, there is no uniform standard to remove organic impurities from the surface of microplastics.

The main identification methods of microplastics include visual screening, stereoscopic microscopy, scanning electron microscopy (SEM), FTIR, and Raman spectroscopy. In the assessment process, FTIR and Raman spectroscopy have been applied for qualitative assessments, while microscopy, including SEM, has been used for the quantification of microplastics (Qiu et al., 2016; Scheurer and Bigalke, 2018; Song et al., 2015; Zhou et al., 2018). The visual method is also used to detect microplastics, but it is difficult to identify small plastic particles, and underestimation or overestimation of the abundance of microplastics in the ecological environment is likely to occur (Song et al., 2015; Shim et al., 2017).

4. Impact of microplastics on soil ecosystems

4.1. Impact of microplastics on the soil structure

A loss of soil structure commonly occurs when large amounts of macroplastics are present in the soil. This is deleterious as it reduces the infiltration of rainwater and irrigation water, negatively affects the soil's water holding capacity and may induce anoxia (Liu et al., 2014). It has also been reported that residual plastic mulch film damages the structure of soil aggregates and reduces soil aeration and water permeability, thereby reducing root growth and overall plant productivity (Jiang et al., 2017; Zeng et al., 2013; Zhang et al., 2018a). In contrast to macroplastics, there are relatively few reports on the relationship between microplastics and the soil structure and aggregates (Zhang and Liu, 2018), and no studies have clearly shown the influence of microplastics on soil structure. Further studies are required to determine where microplastics are physically located in the soil matrix and how this affects their fate and behavior.

4.2. Effect of microplastics on soil physical and chemical properties

Several studies have reported that microplastics have a negative impact on soil organic carbon (C) and nitrogen (N) cycling, soil microbial activity, and nutrient transfer (Cao et al., 2017; Liu et al., 2017; Rillig, 2012, 2018; Rillig et al., 2017b). Liu et al. (2017) showed that the addition of microplastics can stimulate soil enzyme activities and the accumulation of soluble nutrients in soil. In addition, plastic mulch residues inadvertently contribute to increasing the size of the stable soil organic C pool. At a typical plastic contamination level of 5–25 kg ha^{-1} y^{-1}, this equates to a C addition rate of ca. 4–20 kg C ha^{-1} y^{-1}. It should be noted that this is low in comparison to rates of organic C loss from most intensive agricultural systems and therefore should not be viewed in a positive light. Hodson et al. (2017) found that microplastics can improve the bioavailability of zinc and increase the contact between earthworms and zinc as a medium, but little is known about the potential risk to earthworms. In addition, the underpinning mechanisms responsible for this increase in micronutrient bioavailability remain unknown.

In agriculture, measurements of soil physical and chemical quality indicators have been used as indicators to evaluate the advantages and disadvantages of agricultural plastics. In some cases, plastic mulch films improve specific soil quality indicators whilst in others a decline is apparent (Jiang et al., 2017; Liu et al., 2014; Steinmetz et al., 2016). As no integrated soil quality assessment system currently exists it remains difficult to ascertain whether the benefits of plastic mulches outweighs the potential disadvantages (Sarmiento et al., 2018). Some studies have concluded that residual plastic accumulation negatively impacts on the soil's physicochemical properties and will subsequently lead to unsustainable farmland use and environmental damage (Andrés Rodríguez-Seijo, 2018). However, rarely are critical limits for excessive microplastic contamination defined (i.e. tipping points) at which these negative impacts are observed. This makes it difficult to evaluate the spatial scale of the problem, give guidance on microplastic loading rates and predict the carrying capacity of agroecosystems. Ramos et al. (2015) found that plastic residues can accumulate pesticides from soil leading to changes in the soil habitat. Several studies have also indicated that the soil microbial biomass C and N contents significantly decrease with an increasing amount of film residues (Moreno and Moreno, 2008; Wang et al., 2016). It is necessary to confirm whether the abovementioned results were caused by the plastic residues themselves, their intrinsic primary pollutant load (e.g. plasticizers) or secondary pollutant load (e.g. pesticides). In addition, it is necessary to differentiate between the potential toxic effects of the macroplastic vs. microplastic components. Consequently, the evidence base on the impact of plastics on many soil properties is somewhat contradictory and often incomplete. In addition, the focus of previous research has not been on soil function and has not taken an ecosystem services approach. Thus our ability to evaluate their impact on soil health remains very difficult. Another major issue is that the results are not put into a wider context. For example, it is difficult to assess whether the observed changes in soil quality due to microplastics are any different from other waste materials commonly added to land (e.g. biochar, manure, sludges).

4.3. Impact of microplastics on soil organisms and plants

Based on the negative impacts of plastic pollution on marine organisms, there is increasing focus on the dangers of microplastics to soil organisms (Cao et al., 2017; Chae and An, 2018; de Souza Machado et al., 2018b; Huerta Lwanga et al., 2016, 2017a, 2017b; Rillig et al., 2017a, 2017b). Mesofauna (e.g. earthworms, mites, collembola) are known to be vital in maintaining soil quality, however, intensive agricultural systems typically lead to a loss in mesofaunal abundance (George et al., 2017). Consequently, a further loss of these keystone organisms by plastics could represent a major threat to long-term agroecosystem functioning. Huerta Lwanga et al. (2016) studied the survival and fitness of earthworms exposed to microplastics in litter at concentrations of 7%, 28%, 45%, and 60% dry weight. After incubation for 60 d, the earthworms in the 28%, 45%, and 60% microplastic conditions in the litter had a higher mortality rate and significantly lower growth rate compared with those of the control and 7% treatment. The research also confirmed the concentration-transport and size-selection mechanisms of microplastics, which may have important implications for the fate and risk of microplastics in terrestrial ecosystems. It should be noted, however, that the concentrations of plastic used in those studies were 1000-fold higher than seen in most plastic-contaminated agricultural soils. Cao et al. (2017) stated that a low soil microplastic concentration (<0.5%) has little impact on earthworms, but when the microplastic concentration rose to 1% and 2%, it significantly inhibited the growth of earthworms and increased their mortality. The adverse effects of microplastics on soil organisms may be mainly caused by the significant accumulation of microplastics in the gut and stomach of organisms, which can damage their immune systems and affect their feeding behavior and development. Studies in marine organisms indicate that organic pollutants sorbed to microplastics, however, do not readily transfer to the host (Bakir et al., 2016; Ziccardi et al., 2016), albeit this xenobiotic exposure route still needs testing in soil organisms. Bandopadhyay et al. (2018) indicted that biodegradable plastic mulches affect soil microbial communities indirectly by changing the soil microclimate, soil physical structure and through the addition of contaminants adhering to the film fragments. Given the high degree of functional redundancy and diversity within the soil microbial community, it is highly likely that plastic mulch films will affect the composition of the microbial community as it will create new ecological niches within the soil. What is critical, however, for future studies is whether microplastics negatively affect keystone microbial species that are fundamental to the delivery of key soil functions (e.g. nitrifiers, arbuscular mycorrhizas) or whether they increase the prevalence of disease causing organisms (e.g. plant and animal pathogens).

Agricultural plants are known to take up a range of nanoparticles and consequently it is likely that microplastics may enter the food chain through this route (Jassby et al., 2019). Li et al. (2019a,b) has reported that polystyrene microplastics (0.2 μm) can be absorbed and enriched in the root of raw vegetables, and migrate from the root to the shoots. A comparison of microplastics transported from the soil to the edible parts of the plant versus that deposited directly onto the shoots from atmospheric deposition or wastewater irrigation remains unknown. Based on current evidence it is unlikely that this exposure pathway constitutes a major risk to human health or other parts of the food chain (Mateos-Cárdenas et al., 2019). Qi et al. (2018) indicated that the microplastics derived from starch-based plastic mulching film showed stronger negative effects on wheat growth compared to polyethylene. This may be ascribed to the biodegradable plastic being composed of 44.6% polyethylene terephthalate and 18.3% polybutylene terephthalate, or more likely to shifts in microbial communities or starch-induced N immobilization in the soil. The mechanisms by which macro- and micro-plastic contamination in soil affects plant growth remains largely unknown and further work is required to explore this for a wide range of crop plants, especially for edible root crops.

4.4. Influence of microplastics on the groundwater environment

Some studies have indicated that microplastics in marine environments originate from terrestrial ecosystems (Horton et al., 2017b; Luo et al., 2018; Wagner et al., 2014). Soil microplastics can be transported from land to the groundwater environment via long-distance movement, such as animal disturbance, surface runoff, and water infiltration (Blasing and Amelung, 2018; Brodhagen et al., 2015; Hurley and Nizzetto, 2018; Kyrikou and Briassoulis, 2007; Steinmetz et al., 2016), thereby affecting the underground aquatic environment and even disturbing the marine ecosystem. There are scarce reports on the effects of microplastics on the groundwater environment (Chae and An, 2018), even though studies on the marine environment widely exist (McCormick et al., 2014; Zettler et al., 2013).

5. Soil microplastics and phthalic acid esters

One of the key challenges in microplastic research is separating the environmental effects of the plastic polymer from primary and secondary contaminants present in the plastic. Plastic mulch films contain large amounts of phthalate esters (PAEs) which can be released into the soil. As they are endocrine disrupting chemicals, PAEs are often known as "environmental hormones," and have the potential to severely impair human health owing to their reproductive toxicity, developmental toxicity, carcinogenicity, and other toxic responses, particularly if there is prolonged exposure (He et al., 2014; Kong et al., 2012). Six of these PAEs (namely, butyl benzyl phthalate (BBP), di-(2-ethylhexyl) phthalate (DEHP), dimethyl phthalate (DMP), diethyl phthalate (DEP), di-*n*-butyl phthalate (DBP), and di-*n*-octyl phthalate (DOP)) found in mulch films are listed as environmental priority pollutants by the United States Environmental Protection Agency (USEPA, 2013). Concentrations of PAEs in soil exposed to much films have been found in the range 1.8 to 3.5 mg kg^{-1} (Shi et al., 2019). In addition, di-n-butyl phthalate (DBP) and di-(2-ethylhexyl) phthalate (DEHP) were found to be highly enriched in grain samples in the same study (4–12 mg kg^{-1}). Consequently, PAEs not only have negative effects on soil properties, but also accumulate in the food chain, thereby posing a major threat to ecosystem and human health. In order to avoid this threat, it is particularly important to reduce and eliminate the sources of PAEs and offset the existing pollution of PAEs in the environment (He et al., 2014). Microbial degradation is one of the most important methods to remove PAEs in the environment, but it cannot completely remove PAEs from soil or aqueous solutions in the short term (Zhang et al., 2007). It is therefore vital that we reduce or prohibit the use of PAEs in industrial processes where there is an appreciable environmental risk.

It has been reported that PAEs can reduce microbial activity by inhibiting soil respiration and enzyme activity (Guo et al., 2010; Xie et al., 2009). Additionally, PAEs can also affect soil invertebrates, such as earthworms (Chen et al., 2004). Studies have shown that plastic film residues can readily release PAEs into soil (Chen et al., 2012; He et al., 2014; Liu et al., 2014; Wang et al., 2013, 2016) and that these toxic compounds may change the behavior of organic pesticides in soils (Ramos et al., 2015), inhibit enzyme activities (He et al., 2014; Wang et al., 2009; Xie et al., 2010; Zhou et al., 2005) and alter soil microbial communities (Chen et al., 2013; Wang et al., 2016). Thus, they may pose a potential danger to soil functioning (Kasirajan and Ngouajio, 2012; Steinmetz et al., 2016). Worryingly, Wang et al. (2016) have shown that the content of PAEs in soil continues to accumulate with the repeated application of plastic mulch. This suggests that PAEs are relatively recalcitrant in soil and may pose a long-term risk. However, it is unclear whether PAEs in soil and crop products actually originate from plastic films or from another source (Duis and Coors, 2016; Lambert and Wagner, 2016; Rillig, 2012; Wang et al., 2016). For example, DBP may exist in the atmosphere at concentrations of 3–59 ng m^{-3} while in aquatic environments it can be present at concentrations of 1–30 ng l^{-1} (Xie et al., 2005; Wang et al., 2008). There is therefore a clear need to determine the source-partitioning of PAE in agricultural systems. In addition, it is known that PAE removal from soil can be stimulated by the addition of organic materials in the laboratory (e.g. compost; Chang et al., 2009), however, this still needs testing under realistic conditions in the field to facilitate the design of effective mitigation strategies.

6. How to solve microplastic pollution

Microplastics, which are stable and non-degradable, will quasi-permanently remain in the environment, and thereby pose a long-term risk to ecosystems (Horton et al., 2017b; Hurley and Nizzetto, 2018; Liu et al., 2018). An important measure to reduce microplastic pollution in soil is to minimize or avoid using plastics within food production systems. Plastic mulching film, as an important microplastic source in agriculture soils, yet it is indispensable in agricultural production. These films increase crop yields by more than 30%, thereby making a great contribution to the security of agricultural products in countries like China (Fan et al., 2017). Li et al. (2016b) indicated that the use of agricultural film led to 42.3% greater water use efficiency. Give that water supplies are dwindling in soil countries and becoming more uncertain with climate change, it is unlikely that the use of plastic mulch films will stop (van Ittersum et al., 2013).

Thus, reasonable usage and forced recycling of traditional polyethylene mulch films represents a promising solution to reduce plastic pollution in agriculture. The government and relevant departments should also strictly control the entry of low grade plastic film (i.e. high contaminant load) from entering the market. Additionally, we should also promote the multi-year use of agricultural plastic films.

The use of bioplastic, which is a material that can be partially or completely degraded by microorganisms, is an important direction for the plastic industry and agricultural development. The use of biodegradable especially bio-based mulching film is becoming another significant method to solve the problem of plastic film residue and microplastic pollution at source. However, compared to polyethylene film, biodegradable mulch films (especially those which are bio-based) have significant limiting factors for large-scale application owing to their lower mechanical strength, higher cost and greater C footprint associated with production. In the future, it is necessary to reduce the production cost, improve the properties of these products, and optimize raw materials and control degradation time of biodegradable plastic mulching film.

7. Conclusions and perspectives for future work

As an emerging research field, microplastic pollution in terrestrial ecosystems is gaining increasing scientific and media attention due to the long-term threat to agroecosystem functioning, food security and human health. The study of microplastics in soil has proven more difficult than in aquatic ecosystems owing to difficulties in separating plastics from the soil matrix. It is clear from current evidence, however, that a comprehensive mechanistic investigation of the properties and behavior of microplastics in agricultural soils is urgently required. Without this, it will not be possible to evaluate the true risk that microplastics pose to the environment and also will prevent the formulation of effective legislation and policies which enable the safeguarding of human health and help protect the soil and wider environment. Based on the current evidence we have identified 12 priorities for future research:

(1) **Standardization of microplastic separation and detection methods.** Standardization of techniques and methodologies is vital for the quantitative comparison of microplastics in different environments and for the assessment of their potential risks. This also needs to incorporate good quality assurance (QA) procedures to avoid contamination from other sources. Microplastics may decompose or degrade into nanoplastics, which can be ingested in large quantities by marine or terrestrial organisms, thereby posing a serious threat to the organismal and ecosystem health. Hence, it is not only necessary to identify and standardize the separation and identification methods for microplastics, but also to explore and establish methods for the separation, detection and pollutant load assessment of nanoplastics to better understand their ecological significance.

(2) **Microplastics and soil aggregates.** Soil aggregates, as the most basic structural unit of soil and the foundation of soil fertility, are an important site for soil organic matter decomposition and accumulation, nutrient transfer, and transformation. The study of the spatial distribution and behavior of microplastics within soil aggregates in both top and subsoils will be vital in predicting the likelihood of transport, uptake and transformation within the soil profile.

(3) **Microplastics, heavy metals and organic pollutants.** Microplastics, which have a large specific surface area, strong adsorptivity, and strong hydrophobicity, can sorb large amounts of heavy metals, organic pollutants, and pathogens from soil. Research on the sorption/desorption relationships of heavy metal ions and pesticides with microplastics is needed. In addition, the role of plastic biofilms on the retention of plant and animal pathogens is required. This information will greatly inform the likely risk of these pollutants to be transported in soil as well as their potential for long term accumulation in soil, biotransformation and bioaccumulation.
(4) **Microplastics and plasticizers.** Plasticizers, especially phthalic acid esters (PAEs), are now widespread contaminants in agricultural soils and can negatively affect soil enzyme activity, microbial diversity, crop yield, and crop quality. Knowledge of plasticizer concentrations in soil remains fundamental to undertaking holistic risk assessments for microplastics in soils. Work is therefore needed to study the behavior of PAEs in residual agricultural film (macroplastics) and microplastics in a range of soil types and under a wide range of conditions. We also need to evaluate the extent to which PAEs can affect soil food webs and enter the human food chain.
(5) **Long-term fate of plastics in soil and their legacy.** Vast areas of the world's agricultural land are now contaminated with plastics. While it is widely acknowledged that we should gradually transition from using non-renewable plastics to more bio-based plastics within agriculture, the legacy of these conventional plastics is likely to remain for hundreds of years. Consequently, we need to better understand the rates by which macroplastics fragment into micro- and nano-plastics over time and whether problems will still exist even when renewable mulch films are in use.
(6) **Microplastic tipping points.** At present, no critical limits for microplastic pollution in soil have been determined. This limits our ability to be able to quantify the present and future degree of environmental damage caused by microplastic pollution. Consequently, more ecotoxicological dose-response studies are required in which a broad range of soil functions are assessed. This should also focus on the behavior of keystone organisms which regulate critical functions in soil.
(7) **The use of appropriate quality control in microplastic experiments.** In many respects, there are great similarities between research looking at the impact of biochar and microplastics on soil quality. In the former, it is now widely recognized that a standard set of measurements are needed to characterize the biochar used in experiments to allow comparison between studies. Biochar is known to vary greatly depending on origin and this has been shown to greatly affect how it affects the plant-soil-microbial system. Microplastics also vary in size, shape, composition, crystallinity, impurity/contaminant load and consequently a standard set of reporting criteria should be developed to facilitate inter-study comparisons.
(8) **Representative soil conditions and unbiased reporting.** Many previous studies on the impact of microplastics on the soil ecosystems have used extremely high doses which do not reflect real world conditions. These are not useful from a risk assessment perspective. In addition, the studies using excessive doses has led to misinterpretation and over-sensationalization of the results by the media. It is important that researchers provide a balanced opinion and place the results in a wider context when reporting.
(9) **Longer-term field trials.** Most controlled studies using microplastics have been performed in the laboratory over a period of weeks or months, however, this only provides an initial snapshot of the response. It is important therefore that controlled trials are set up which allow the longer-term impact of microplastics to be evaluated. These should preferably be performed at the field scale with sufficient replication and the appropriate controls. Field-scale trials will also improve our understanding of how UV weathering of plastics affects its persistence in soil.
(10) **Cleaning-up plastic pollution in soil using bio- and phytoremediation.** A range of microorganisms have been isolated from soils and waters which possess the ability to degrade plastic polymers. These are normally present at low abundance suggesting that their realized niche in soil is small. Although bio-inoculants typically have low rates of persistence in soil, there is a need to look at the potential to inoculate soils with these organisms to facilitate plastic depolymerization. In addition, it is clearly possible to genetically modify plants and associated symbionts to produce exoenzymes (e.g. PETase and MHETase) which can degrade plastic residues.
(11) **Comparison of the impact of macro- and microplastics.** It is still unclear whether micro- and nano-plastics are more environmentally damaging than macroplastics. More work is therefore needed to directly compare these different size fractions. In addition, the effect of microplastics are typically only compared against an unamended control sample. The use of a negative control (e.g. addition of inert quartz sand) or a positive control (e.g. compost, biochar) could be used to better contextualize any changes observed following the addition of microplastics.
(12) **Microplastic modelling at the field and landscape scale.** It is widely reported that many microplastics entering the ocean are derived from the land. The role of agriculture and plastic much films in particular in this transfer process remains unknown. More landscape-level monitoring studies are therefore needed where source apportionment can be made between the different microplastic sources and sinks. This will be greatly aided by soil profile-based models which can predict the long-term movement and persistence of microplastics in soil.

In conclusion, microplastics are becoming almost ubiquitous in soils and they are likely to increase in abundance for the foreseeable-future. Based on their recalcitrance, it is also likely that they will still be present in soil for generations to come. Therefore, it is necessary and urgent that we prohibit the addition of microplastic particles to cosmetics, detergents, and other industrial products which may enter soil via biosolids and irrigation water. It is also important that we continue to develop more environmentally friendly biodegradable mulches to replace conventional plastic films. Plastic has proved to be vital in the green revolution, however, we need a second green revolution in which mulch films are truly biodegradable and which leave no toxic, visible or distinguishable residues following degradation. These products will ensure that we are able to maintain the sustainable development of agricultural systems and restore soil health.

Declaration of Competing Interest

The authors declare that they have no conflict of interest.

Acknowledgments

This work was supported by the Zero-Waste Agricultural Mulch Films for Crops in China project (Newton Fund: Newton UK-China Agritech Challenge 2017; No. 2017YFE0121900), National Natural Science Foundation of China General Program (No. 31871575), Science and Technology Innovation Project of CAAS (2018-2020) and the UK Global Challenges Research Fund (BU2019). We gratefully acknowledge the anonymous reviewers for their valuable comments on our manuscript.

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