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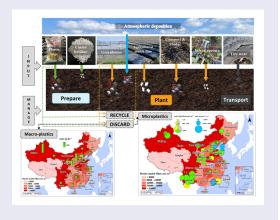
Potential sources and occurrence of macro-plastics and microplastics pollution in farmland soils: A typical case of China

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ABSTRACT

Plastic debris (including macro-plastics, microplastics (MPs), and nanoplastics), defined as an emerging contaminant, has been proven to significantly affect soil ecosystem functioning. Accordingly, there is an urgent need to robustly quantify the pollution situation and potential sources of plastics in soils. China as the leading producer and user of agricultural plastics is analyzed as a typical case study to highlight the current situation of farmland macro-plastics and MPs. Our study summarized information on the occurrence and abundance of macro-plastics and MPs in Chinese farmland soils for the first time based on 163 publications with 728 sample sites. The results showed that the average concentration of macro-plastics, and the abundance of MPs in Chinese farmlands were 103kg ha⁻¹ and 4537



items kg⁻¹ (dry soil), respectively. In addition, this study synthesized the latest scientific evidence on sources of macro-plastics and MPs in farmland soils. Agricultural plastic films and organic wastes are the most reported sources, indicating that they contribute significantly to plastic debris in agricultural soils. Furthermore, the modeling methods for quantifying macro-plastics and MPs in soils and estimating the stock and flow of plastic materials within agricultural systems were also summarized.

KEYWORDS Abundance; farmland soils; macro-plastics; microplastics; quantitative method; source apportionment **HANDLING EDITORS** Hyunjung Kim and Jörg Rinklebe

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1. Introduction

Plastics are widely used across almost all sectors of society due to their versatility relatively cheap cost, light weight and durability (Jambeck et al., 2015; Plastics Europe-The Facts, 2022). The production of plastics is increasing with global cumulative production predicted to reach up to 33 billion tons by 2050 (Sharma et al., 2020). As most plastics have a relatively short functional lifespan, the disposal of plastics represents a major global problem with a large proportion of plastics not being recycled (Luan et al., 2021; Wang et al., 2021). Currently, it is estimated that 79% of plastic waste enters either landfills or the natural environment where it represents a threat to terrestrial, freshwater, and marine ecosystems (Geyer et al., 2017). Although macro-plastics (particles size > 5 mm) represent the primary type of waste entering the environment, they gradually degrade into smaller fragments in response to ultraviolet (UV) irradiation, mechanical abrasion, and biodegradation (Barnes et al., 2009; Yang et al., 2022). Microplastics (MPs) are defined as particles < 5 mm and > 1 µm, including fragments, fibers, particles, foams, and films, while plastic particles with the size between 1 nm and 1 µm are defined as nanoplastics (NPs) (Frias & Nash, 2019; Thompson et al., 2004). Since MPs and NPs are small in size, and present in large quantities and degrade slowly, they are easily absorbed, inhaled, or ingested by organisms, leading to bioaccumulation (Barnes et al., 2009; Leslie et al., 2022; Wu et al., 2022). Studies have found the presence of MPs and NPs in plants, soil fauna, human feces, and blood (Leslie et al., 2022; Li et al., 2020; Lwanga et al., 2017; Zhang et al., 2021). Current evidence indicates that MPs can be transferred to the human body through the food chain as well as via inhalation and are likely to give rise to a range of cytotoxic effects that are now becoming evident, albeit still incomplete (Brachner et al., 2020; Hua et al., 2022; Wu et al., 2022).

Previous reports have focused mainly on aquatic ecosystems with MPs as an emerging contaminant (Cozar et al., 2014; Rochman et al., 2016). Recently, researchers have expanded their focus to terrestrial environments (Kumar et al., 2020; Li et al., 2020). Our recent meta-analysis study quantified the effect of plastic residues and MPs on indicators of global soil ecosystem functioning (i.e., soil physicochemical properties, plant and soil animal health, abundance, and diversity of soil microorganisms) (Zhang et al., 2022). The results showed that plastic residues and MPs can alter plant growth and soil physicochemical properties. For example, plastic residues and MPs decreased root biomass, plant height, soil dissolved organic carbon, and soil total nitrogen (N) content by 14%, 13%, 9%, and 7%, respectively (Zhang et al., 2022). It should be noted, however, that significant bias may occur in meta-analyses as neutral/non-significant results are often not reported in the literature (Coursol & Wagner, 1986). Further, many studies investigating the effect of MPs on soil ecosystem responses have used high rates of MPs contamination in soil (> 1% w/w) (Meng et al., 2021; Ng et al., 2021), which may not reflect levels (much lower than 1% w/w) that occur in typical agronomic field conditions (Huang et al., 2020; Liu et al., 2019). In addition, plastic contaminants and their additives have been shown to inhibit the growth and development of soil animals. The decrease in growth rate, movement rate (e.g. frequency of body bending and head thrashing), feeding rate, and reproduction rate reveals the disturbed locomotor behaviors of animals caused by plastic residues and MPs (Wang et al., 2021; Zhang et al., 2022). Furthermore, several studies have reported that plastic particles within the size of 0.08-2.00 µm (i.e., NPs) can penetrate the stele of rice, cucumber, wheat, and lettuce, leading to efficient uptake of smaller plastic particles (Li et al., 2021; Liu et al., 2022). It indicates that MPs and NPs can be transferred to livestock and the human body through the terrestrial food chain, causing a potential threat to livestock and human health, and natural ecosystem food webs (Lwanga et al., 2017; Zhou et al., 2021).

MPs can also be vectors for the attachment and transmission of other contaminants (e.g. hydrophobic organic contaminants, heavy metals, harmful microorganisms), posing a potential threat to (human, livestock, soil fauna) organismal health and the wider environment (Brennecke et al., 2016; Wang et al., 2019; Zettler et al., 2013). For example, the heavy metals cadmium (Cd) and lead (Pb) were detected in MPs samples (n = 924) from two beaches in Southwest

England with maximum concentrations of 3.4 and 5.3 mg g⁻¹, respectively (Massos & Turner, 2017). In addition, they evaluated the maximum bio-accessible concentrations of Cd and Pb in the proventriculus-gizzard of seabirds (Fulmarus glacialis) and found concentrations exceeded the safe dietary intake limit by a factor of about 50 and 4, respectively (Massos & Turner, 2017). Furthermore, high concentrations of zinc (Zn, 9407 mg kg⁻¹) and polycyclic aromatic hydrocarbons (PAH, 47 mg kg⁻¹) have been detected in MPs samples recovered from earthworms and the surrounding soil, and MPs exposure resulted in the steep rise of the abundance of pathogenic microorganisms in the worm intestinal tract (Ding et al., 2020).

Many studies have indicated that the additives contained in MPs may represent a greater threat to terrestrial ecosystems than the plastic polymer itself (Hahladakis et al., 2018; Halden, 2010). There have been very few studies on the effects of MPs on human health, however, some of the additives used in plastics manufacturing, e.g. plasticizers and heavy metals, have been shown to interfere with gene expression, cell metabolism (e.g. signal transduction, enzyme function), and animals and humans development as well as reproduction (e.g. endocrine disrupting properties) (Rist et al., 2018). For instance, the common plasticizer of bisphenol A (BPA), as an endocrine disruptor, could disrupt the endocrine system and various functions of organisms, including the thyroid, reproductive system, and metabolism (Halden, 2010).

As plastic waste (including macro-plastics and MPs) in terrestrial environments poses a potential threat to food security, human health, and the health of our natural environment, it is important to control plastic input and manage legacy plastic in soils. Greater effort is needed to quantify the sources of macro-plastics and MPs and the fate of different plastic fragments. A study by Jambeck et al. (2015) reported that 80% of marine plastic residuals arise from land, suggesting that soil is not only an important sink of MPs but also an important source (Rachman, 2018). The accumulation of macro-plastics and MPs waste in soils is the result of various human activities and environmental origins, such as discarded plastic litter (Rillig, 2012), plasticulture practices (e.g. plastic mulch films, greenhouse films, irrigation pipes, and associated infrastructure) (Bläsing & Amelung, 2018, Gündoğdu et al., 2022; Huang et al., 2020; Wang et al., 2022), sewage sludge application (Long et al., 2019), coated fertilizers (Katsumi et al., 2021), organic fertilizer and agricultural compost (Weithmann et al., 2018), atmospheric deposition (Allen et al., 2019), digested food waste (Porterfield et al., 2023), and rubber tire wear (Evangeliou et al., 2020). Of these, plastic films represents an important source of macro-plastics and MPs in agricultural soils and has attracted extensive research and discussion (Qi et al., 2020). Plastic films are used extensively throughout the world in both horticulture and arable cropping (e.g. mulch films and polytunnels) as well as within livestock production (e.g. silage wrapping). Typically, these plastics are not recycled efficiently for several reasons including (i) difficulty in removing them from the soil after use, (ii) contamination by soil and vegetation residues, (iii) loss from the field due to wind erosion, (iv) lack of recycling infrastructure, and (v) poor financial incentives (Li et al., 2021; Mekonnen et al., 2016). These barriers to recycling have led to the accumulation of legacy plastic in soils.

An increasing body of research has focused on the abundance and distribution, migration pathways, and ecological environmental impact of MPs. Several recent reviews have covered these topics, but there is little information about the sources of macro-plastics and MPs, generation rates of MPs, and movement of MPs in and through the soil environment, especially for farmland soils (Qi et al., 2018; Yang et al., 2021). At the current rate of increase in plastic production between 2020 and 2021 (about 4%) (Plastics Europe-The Facts, 2022), understanding the sources and consequences of macro-plastics and MPs represents a priority in terms of understanding the potential risks as well as the design and implementation of effective mitigation strategies. Therefore, the aims of this review are to provide the latest understanding of the sources, abundance, and distribution of both macro-plastics and MPs in agricultural farmlands with a focus on quantitative methods and knowledge gaps. Furthermore, China as an example, which has become the world's biggest consumer and disposer of plastic films to be mapped the different sources and abundance of macro-plastics and MPs in the Chinese farmland soils based on the data from published literature (Figures 1 and 2).

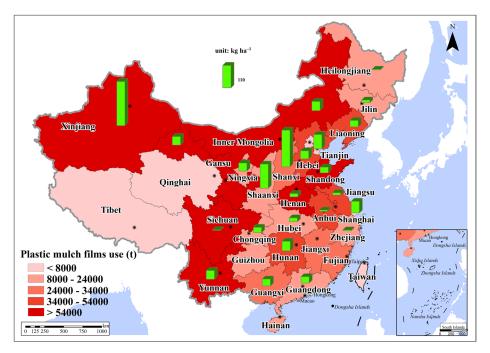


Figure 1. Concentration and distribution of macro-plastics in Chinese farmland soils with the depth of 0 to 80 cm. Data were collected from the literature published before 31st jan 2022. The green bars refer to the concentration of macro-plastics (kg ha⁻¹). The red background color refers to the total amount (t) of plastic mulch film used in different provinces of China in 2021.

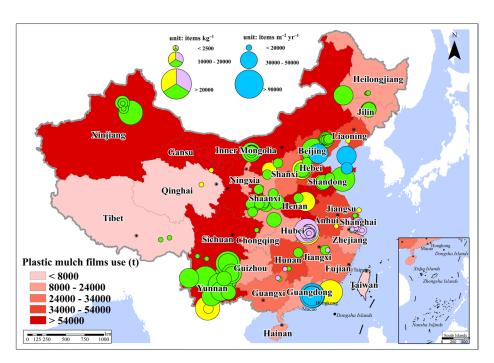


Figure 2. Abundance and distribution of MPs from different sources in Chinese farmland. The different colors refer to the potential sources of MPs entering farmland that are mentioned in the article: agricultural plastic film (green, unit: individual items kg—¹); compost and sewage sludge (purple, unit: individual items kg—¹); atmospheric deposition (blue, unit: individual items m-²yr-¹); the unknown source (yellow, unit: individual items kg—¹). The circles represent the abundance of MPs and the red background color refers to the total amount (t) of plastic mulch film used in different provinces of China in 2021.



2. Distribution and potential source of macro-plastics and MPs in Chinese farmlands

China is suffering from serious plastic pollution, particularly within agricultural environments where recycling is problematic (Plastics Europe-The Facts, 2022; Qi et al., 2020). In 2021, the use of plastic mulch film (PMFs) in China was 1.3 million tons, representing 75% of global plastic mulch use (NBSC, 2011-2022; Yan, 2022). Further, the area covered by plastic mulch film in China in 2021 was 17.3 million hectares, equivalent to 70% of the area of the United Kingdom (NBSC, 2011-2022). However, the recovery rate of plastic mulch film from the field at the end of cropping was < 60% (Zhao et al., 2017). Because of the low plastic film thickness of 6-8 µm commonly used in China, it is difficult to completely retrieve it from soil (Yan et al., 2006), and what is retrieved is contaminated with soil, limiting opportunities for recycling. Based on previous studies, summary information on the occurrence and potential sources of macro-plastics and MPs is important to control them in agricultural soils.

2.1. Literature search and data collection

To understand the distribution of soil macro-plastics and MPs in China's agricultural soils, this review searched the literature from three scientific databases (i.e., Web of Science, EI Compendex, and China Knowledge Resource Integrated Database) for "search terms" including (plastic residue or plastic debris) and (macro-plastic or microplastic or nanoplastic) and (soil or terrestrial) (specific search strings in Table S1). The literature search was limited to papers published before the 31st of January 2022. In summary, these papers were chosen according to the following selection criteria: (a) the study must be practical measurement data, without extra addition of nonagricultural plastics and MPs; (b) these sampling data must have a specific location name (to cities or counties) or longitude and latitude; (c) the samples must be collected in the farmland soil of China; (d) must be collected in bare soil, rather than in the greenhouse. Finally, 163 articles (including 123 studies for macro-plastics and 40 studies for MPs) were selected from more than 4,800 publications using strict inclusion criteria (Tables S2 and S3). Details of search strings and the number of publications were presented in Tables S1 and S4 in Supporting Information. The database covered 30 provinces with 728 data points to show the situation of macro-plastics and MPs in Chinese agricultural soils (Figures 1 and 2). However, there is a limitation that due to the longitude and latitude of some points being located close together, they overlapped and appeared to only be one location presented in Figure 2. This situation was shown in Jiangsu (data num = 5), Guangdong (13), Jilin (7), Hebei (16), Heilongjiang (4) and Inner Mongolia (13) Province.

2.2. Distribution and occurrence of macro-plastics and MPs in Chinese farmlands soil

2.2.1. The occurrence of macro-plastics in Chinese farmlands soil

The concentration of macro-plastics was investigated in 24 provinces of China collected from 123 articles (see the list of articles in Table S2) as shown in Figure 1. The concentration of macro-plastics found in soil (0-80 cm depth) varied from 0.2 to 421.6 kg ha-1 with an average value of 103.3 kg ha⁻¹, with the median of 54.7 kg ha⁻¹. The province's highest concentration (421.6 kg ha⁻¹) was found in Xinjiang province (Northwestern China). The highest average concentration of macro-plastics (142.7 kg ha⁻¹) was found in Northwestern China (e.g. Xinjiang, Ningxia, and Gansu province), followed by 37.4 kg ha⁻¹ in Northern China (e.g. Hebei, Shandong, and Shanxi province), and 30.7 kg ha-1 in the soils of Southern China (e.g. Hubei, Yunnan province and Shanghai) (Table S2). This result was consistent with previous results that investigated 384 soil samples collected from 19 provinces, and the macro-plastics concentrations in the typical areas covered with mulch films soil samples ranged from 0.1 to 324.5 kg ha⁻¹ and the highest concentration was observed in Northwestern China (i.e., Xinjiang, Gansu province

and Inner Mongolia) (Huang et al., 2020). More than 40% of soil samples (n=51) were from the plow layer, i.e., a depth of $0-30 \,\mathrm{cm}$. Of these samples, 49% (n=25) were divided into three soil layers: 0-10, 10-20, and 20-30 cm. Based on an assumed film thickness of 6-8 µm and density of 0.910 - 0.925 g cm⁻³ (low-density polyethylene, LDPE), a weight of 103 kg ha⁻¹ macro-plastics corresponded to an area of $1.4-1.9\times10^4$ m² ha⁻¹ (Yan et al., 2006). The region in Northwestern China is a typical area where plastic films are used widely, e.g. Shaanxi province (21 thousand tons of plastic mulch films used in 2021) and Gansu province (122 thousand tons of plastic mulch films used in 2021) (NBSC, 2011-2022). The preference for using plastic mulch films in these regions is to conserve soil water and increase soil temperature in maize, cotton, peanut, potato, and other vegetable cropping (Dong et al., 2015; Gao et al., 2019). As the most important cotton-producing area in China, Xinjiang province uses the largest amount of plastic mulch film (240 thousand tons per year in 2021, NBSC, 2011-2022). The region of Southern China has adequate water and mild temperatures for crop growth without the need for plastic mulch films. Nevertheless, there are some exceptions. For example, Yunnan province, which is located in the Southwestern of China with the largest flower and tobacco production, used 90 thousand tons of plastic mulch films for the cultivation of cash crops cultivation in 2021, exceedingly more than 80% Chinese provinces (NBSC, 2011-2022). The average amount of macro-plastics in this area was relatively high at 44 kg ha⁻¹. It should be noted that the median value (54.7 kg ha⁻¹) of macroplastics in current study was consistent with the average value of plastic residues (55 kg ha⁻¹) from the second national pollution source census of China, while the average value (103 kg ha⁻¹) of macroplastics in current study was higher compared to 55 kg ha-1 observed in the second national pollution source census of China, (Atlas of the Second National Agricultural Pollution Source Census, 2022). This may be due to differences of study area and data sources. Our study included 24 provinces with the data published from 1991 to 2021, while the event of second national census on pollution sources census of China included 31 provinces and the data was based on the soil samples collected in 2017.

The chemical composition of most macro-plastics generated from plastic mulch film was suggested to be polyethylene (PE) since the majority of plastic mulch film was made of PE in China. In addition, chemical compositions of polyvinyl chloride (PVC) and ethylene vinyl acetate copolymer (EVA) were also reported in several studies as shown in Table S2.

2.2.2. The occurrence of MPs in Chinese farmlands soil

The abundance of MPs was investigated in 28 provinces of Chinese farmland through the data collected from 40 articles (see the list of articles in Table S3) shown in Figure 2. The abundance of MPs varied from 1.6 to 6.2×10^5 items kg⁻¹ (dry soil) with an average abundance of 4536.6 items kg⁻¹, with the median of 1640.0 items kg⁻¹. The highest average MPs abundance (4,817.9 items kg⁻¹) was found in Southern China, followed by 4,156.1 items kg⁻¹ in Northern China, 3,602.7 items kg⁻¹ in Northwestern China, and 82.3 items kg⁻¹in the Qinghai-Tibet Plateau (Table S3). Interestingly, Northwestern China with a higher concentration of macro-plastics was not the highest abundance of MPs. This might be due to the lower temperatures and plow frequency in Northwestern China, causing the lower generation rate from macro-plastics to MPs. The most commonly researched regions for MPs were Hubei province (abundance of MPs from 327.5 to 6.2×10⁵ items kg⁻¹), and Shanghai city (abundance of MPs from 1.6 to 2153.0 items kg⁻¹) (see more information in Table S3). The number of sampling points in these two regions was the highest, with 59 (Hubei province) and 123 (Shanghai city), indicating that more research of MPs has been carried out in Hubei province (Wang et al., 2021; Zhou et al., 2019) and Shanghai City (Lv et al., 2019; Zhou et al., 2020) compared to other Chinese regions. However, there were only a few studies of MPs in Northwestern China (such as Xinjiang and Gansu provinces), where the concentration of macro-plastics was relatively high. Therefore, further studies are recommended that focus on quantifying terrestrial MPs pollution in Northwestern China.

As the types of MPs are important to their environmental fate and ecotoxicity (Zhang et al., 2022), the chemical composition of MPs in Chinese farmland soils investigated in the literatures are summarized in Table S3. The mainly components of MPs in Chinese farmland soils were PE, polypropylene (PP), polyester (PES), polystyrene (PS) and polyamide (PA). There were 2-27 types of plastic materials in the Northern China and PP and PE were mostly widely used, while more types (2-60) of plastic materials were investigated in the Southern China. This may be due to the well-watered condition and higher population density in the region, resulting in a complex source of polymers (Chen et al., 2022). 10 of 11 literatures (which studied the chemical composition of MPs in the Northwestern China) showed that the main plastic type was PE. This was probably attributed to the large consumption of plastic mulch films in this area, and the majority of this film was made of PE in China. Therefore, PE mulch films probable be an important source of soil MPs in Chinese farmland soils.

2.3. The potential sources of MPs in Chinese farmlands soil

Most of the macro-plastics in farmland soils come from the damage of agricultural plastic products. However, the sources of MPs in soils are much more various. In the last 10 years, the agricultural sector has become increasingly important as a source of MPs in soils. The most potential agricultural sources (i.e., agricultural mulch films, compost and sewage sludge, and atmospheric deposition) have been focused and mapped the distribution and abundance of MPs from these sources in China (Figure 2 and Table S3). It should be noted that the data points of MPs caused by other sources, such as coated fertilizer and food waste (which are not well investigated in previous literature but may contribute to MPs in soils) are presented in yellow circles in the revised Figure 2.

This research explored the distribution and abundance of MPs from the agricultural plastic films with 146 sampling points (Figure 2(green)). It highlighted that most of the research on agricultural plastic films has focused on the regions of Northern and Northwest China (Wang et al., 2021; Zhang et al., 2021). In contrast, most of the research on compost and sewage sludge was concentrated in Southern China as shown in Figure 2(purple) with 57 soil samplings, where there are more wastewater treatment plants and larger quantities of sludge production (Yang et al., 2021). Only a few cities have measured MPs in the atmosphere, including Shanghai, Dongguan, Dalian, Tianjin, Wenzhou, and Yantai, with a total of 15 samples as shown in Figure 2(blue). The average abundance of MPs from agricultural plastic films, composts and sludge, and atmospheric deposition were 4,231.1 items kg⁻¹, 1,002.3 items kg⁻¹, and 7.9×10⁴ items m⁻²yr⁻¹. Taking into account all the potential sources on MPs, plastic mulch films represent the most important source in Chinese farmland soils.

3. Contribution of different sources to soil macro-plastics and MPs wastes

The sources of macro-plastics mainly consist of input from improper disposal of agricultural plastic practices and solid waste (e.g., domestic waste) from the surroundings of farmland soil (Hurley & Nizzetto, 2018; Qi et al., 2020). In contrast, the sources of MPs are more complex. MPs are generally categorized into two types: primary and secondary MPs. Primary MPs mainly refer to micro-size plastic particles that are manufactured intentionally for commercial uses, and they often act as raw materials for industrial production (Bläsing & Amelung, 2018; Yang et al., 2021). The rapid growth in the fibers from man-made textiles, microbeads from personal care products, and fragments produced during the plastic manufacturing process mean that they are significant sources of primary MPs (Fu & Wang, 2019). Furthermore, increasing quantities of primary MPs are being introduced to agricultural soils via organic wastes and wastewater residue input (Yang et al., 2021). However, the most common MPs in the environment are secondary MPs (Cole et al., 2011; Qi et al., 2020). Secondary MPs are generated from the degradation and decomposition of larger macro-plastics products and debris into micro-nanoplastics by abiotic (e.g. high temperature, wind-blown and ultraviolet radiation, (Horton et al., 2017; Rezaei et al., 2019) and biotic factors (e.g. microbial decomposition, Yuan et al., 2020). Compared to primary MPs, secondary MPs are more difficult to determine the source and rates of generation. The following sections of this study provide a summary of the potential sources and quantitative estimate modeling methods for macro-plastics and MPs.

3.1. Plasticulture practices

3.1.1. Plastic mulch films

Plastic mulch films are used for several reasons that improve crop yields, e.g. for increasing the soil temperature and water use efficiency, promoting seed germination, inhibiting weed growth, and reducing soil erosion (Gao et al., 2019). However, with the increasing use of agricultural plastics film, especially in several developing countries (e.g. China, India, Egypt, and Vietnam), there is an increasing legacy of plastic mulch film residue accumulation, including macro-plastics and MPs (Maraveas, 2020; Plastics Europe-The Facts, 2022).

In Asia, the largest usage of agricultural films is China with consumption of 2.5 million tons, accounting for > 70% of Asia's, and almost 50% of the worldwide in 2018 (FAOUN, 2021; Le Moine, 2018; NBSC, 2011–2022). The use of plastic mulch film in China has increased nearly three times from 375 thousand tons in 1993 to 1320 thousand tons in 2021, however, the recovery rate after crop harvest is under 60% (Zhao et al., 2017). Additional, according to 1.18 million tons of accumulated plastic residue in Chinse farmland soils (Atlas of the Second National Agricultural Pollution Source Census, 2022) and the assumed film thickness of 6—8 μ m as well as the LDPE film density of 0.910–0.925 g cm⁻³, 0.16–0.22 million km² of legacy plastic has been left in the soil in China.

A study by Ren et al. (2021) reported that the amounts of MPs in Chinese surface farmland soils (0—10 cm) ranged from 4.9×10^6 to 1.0×10^7 tons in 2018, and agricultural mulch films contributed 10-30% of the total inventory of MPs in the Chinese farmland. Of this, it was estimated that $1.2 \times 10^5 - 2.2 \times 10^5$ and $3.4 \times 10^4 - 6.6 \times 10^4$ tons of MPs from Chinese farmland soils entered the surface water and ocean each year, respectively. In addition to the plastic polymer, chemicals added to agricultural films during their production, e.g. phthalates (phthalic acid esters, PAEs) can be released into farmland soil by plastic debris (Wang et al., 2016). According to the study by Zhang et al. (2021), 91.5 tons of PAEs migrated into Chinese soils from agricultural films in 2017, with a risk of these being taken up by vegetables and entering the human body *via* the food chain.

Biodegradable plastic mulches (BDMs) have been developed as substitutes to conventional PE mulch films and are formulated to reduce the persistence of residues in soil (Yang et al., 2022). Because of the growing awareness of the persistence of synthetic plastic mulch films in the environment, BDMs have gradually entered the mulch film market in China (He et al., 2018; Plastics Europe-The Facts, 2022). However, studies have reported that MPs formation is more rapid from biodegradable mulch than from traditional non-degradable mulch films. For example, plastic films were exposed to UV irradiation of 2.1 MJ m⁻² in a lab experiment by Yang et al. (2022). This level of UV exposure simulated the cumulative irradiance level of 70 days of natural summer solar light in Northern China. The average quantity of MPs released from biodegradable, and non-degradable mulch films were 475, and 155 particles cm⁻², respectively.

In summary, the mulching duration, amounts of mulch films and plastic material are important factors that affect the plastic fragmentation. As an important source of macro-plastics and MPs in agricultural soil, the level of plastic accumulation by agricultural plastic mulch films, especially BDMs, is alarming but has received relatively little attention to date. A few studies have shown that BDMs can contribute more MPs to soil compared with conventional PE mulch films at the



same time period (Yang et al., 2022; Zhou et al., 2023). Therefore, further research is needed to investigate the fate and generation process of MPs from mulch films, including BDMs.

3.1.2. Greenhouse films

Greenhouses represent the largest proportion of agricultural plastic films used in plant production worldwide (Le Moine, 2018). It was estimated that the global average quantities of greenhouse films used 3500 kg ha-1 in 2019, which represented 47% of agricultural film demand (FAOUN, 2021; Le Moine, 2018). Greenhouses are used to prolong the growing season in temperate regions of the world, and most plastic greenhouses are concentrated around Asia (FAOUN, 2021). Since there is no direct amount of greenhouse films used in China and agricultural films is mainly used as mulch films and greenhouse films in China, the amount of greenhouse films is represented by the difference between the amount of agricultural films and mulch film (Zhang et al., 2021). In 2021, China's use of greenhouse films reached 1.04 million tons, accounting for 44% of the total amount of agricultural film (NBSC, 2011-2022).

A study by Wang et al. (2022) investigated MPs contamination from three different types of greenhouses (abandoned greenhouse, normal greenhouse, and simple greenhouse). The degree of MPs contamination was found to follow the order: abandoned greenhouse (2215.56 items kg^{-1}) > normal greenhouse (891.11 items kg^{-1}) > simple greenhouse (632.50 items kg^{-1}). The composition of MPs from these different greenhouses was also different. The most important components of the abandoned greenhouses were rayon (RY) (10.3%) and poly (ethylene terephthalate) (PET) (7.7%). In the simple greenhouses, poly (1-tetradecene) (PTD) (14.2%) and RY (10.0%) were more common. The most abundant polymer type was PP, PE, and polypropylene polyethylene copolymer (PP: PE) in all the three greenhouses. These polymers accounted for > 50% of the total (Wang et al., 2022).

The environmental concern about MPs from greenhouse plastic film covers is less compared to PMFs. There is a tradition of recycling greenhouse films in China, since the greenhouse films (thickness of 8-50 µm) is durable and easier to be recycled than mulch films (thickness of 6-8 µm). The recovery rate of plastic mulch films was less than 60% in China (Zhao et al., 2017), and the target recovery rate of agricultural films will be 85% by 2025 (Development and Reform Commission of the People 's Republic of China 2021). It can be inferred that the recovery rate of greenhouse films would be higher than 85% by 2025.

3.1.3. Irrigation pipes and associated infrastructure

The source of macro-plastics and MPs in farmland soil is not limited to the use of plastic films, also included abandoned irrigation pipes, mismanaged agrochemical containers, and disposable crop protection packaging (Gündoğdu et al., 2022).

A recent study in Turkey showed the concentration of MPs from disposable greenhouse plastic films and irrigation pipes in agricultural soil ranged from 0.3 to 32 particles kg⁻¹ with an average of 11.1 particles kg-1 (Gündoğdu et al., 2022). Furthermore, MPs with additives could act as vectors for pollutants, e.g. dibutyl phthalate which has been shown to be released from PVC pipe fragments in water, representing an added risk to both organisms and the environment (Ye et al., 2020). In addition, the pollution level is highly correlated with the amount of disposable drip irrigation pipes and greenhouses in the contaminated sites (Katsumi et al., 2021; Ye et al., 2020). Results showed that irrigation pipes and associated infrastructure could be potential sources of MPs in irrigated farmland soils, which should be taken into account when estimating the concentration of MPs in soil (Gündoğdu et al., 2022; Pérez-Reverón et al., 2022). Furthermore, the topics of lifespan and waste management of agricultural plastic infrastructure and source identification of MPs in soil based on their physical properties (e.g. size, shape, and the type) are also worthy of further study.

3.1.4. Coated fertilizer use

Polymer-coated fertilizer (PCF) comprises a nutrient core wrapped by a polymer coating and is designed to release nutrients to plants at a gradual and controlled rate (Du et al., 2006). PCF is composed of microcapsules with a thickness of $10-80\,\mu m$ and a diameter of 2–5 mm, which are not recovered after use. These microcapsules are primary MPs and can further degrade into NPs (Bian et al., 2022; Katsumi et al., 2021; Trenkel, 2010). The use of PCF in China is increasing at a rate of 10%-15% per year (Li et al., 2022). It is expected that the output of the Chinese PCF will reach $7.6\times10^6-11.3\times10^6$ tons by 2025, with the microcapsules input to soils potentially amounting to $0.4\times10^6-0.6\times10^6$ tons (Yang et al., 2009).

Although PCF can reduce nutrient leaching loss and ammonia emissions, the fate and impact of the residual polymer coating are attracting the attention of fertilizer companies and environmental researchers (Lian et al., 2021). For example, Katsumi et al. (2021) investigated the accumulation of microcapsules derived from coated fertilizer in 19 paddy fields in Japan with concentrations found to range from 6-369 mg kg⁻¹ (mean 144 mg kg⁻¹). The result showed that legacy plastics from microcapsules will continue to accumulate in farmland soil as long as conventional PCF is used. The spatial distribution of MPs from PCF is also strongly affected by irrigation, and the soil around drainage outlets has been found to be a hot spot (Katsumi et al., 2021). Several studies have also measured the release of macro-plastics in the environment by PCF made from different co-polyesters (Lubkowski et al., 2016). For example, experiments have shown that the residual amount of PCF film shells left in the soil was 50 kg ha⁻¹ every year, which accounting for 50% of the average annual nutrient consumption input in the European Union (100 kg ha⁻¹) (Lubkowski et al., 2016). Furthermore, it is estimated that the concentration of macro-plastics from PCF in the soil can reach 500 kg ha⁻¹ after continuous application of PCF for 10 years (Li et al., 2022). Whilst previous research has focused on the benefits of PCF on plant growth, soil properties, soil microbial communities, and reduced risk of nutrient losses to water and air, further attention is needed to assess the contribution of this source to plastic pollution in China (Bian et al., 2022; Lian et al., 2021). The release of MPs may become a potential food safety problem for the long-term application of PCF in farmland.

3.2. Organic wastes and wastewater residue input

3.2.1. Sewage sludge

During wastewater treatment, most MPs are removed from the wastewater stream and become concentrated in the sludge (biosolids) fraction (Ziajahromi et al., 2016). In many countries, this nutrient-rich semi-solid waste product is applied to agricultural land as a soil improver and fertilizer (Corradini et al., 2019; Hurley & Nizzetto, 2018). Agricultural soils in Europe and North America may receive more than 63,000 and 44,000 tons of MPs per year through sludge applications, respectively (Nizzetto et al., 2016). However, very little is known about the fate and transport of MPs in sludge in the terrestrial environment (de Souza Machado et al., 2018; Ng et al., 2018).

In Europe and North America, about 50% of sewage sludge is processed for agricultural use, and it is estimated that 125-850 tons of MPs per million peoples are added annually to European agricultural soils either through direct application of sewage sludge or as processed biosolid (Nizzetto et al., 2016). A recent study by Lofty et al. (2022) estimated a maximum application rate of $4.8\,\mathrm{g}$ of MPs m⁻²yr⁻¹ or 1.15×10^4 MPs particles m⁻²yr⁻¹ in Europe from sewage sludge applied to agricultural soil by measuring the MPs content of sewage sludge at wastewater treatment plants (WWTPs). These studies strongly suggest that the practice of spreading sludge on agricultural land could potentially make them one of the largest global reservoirs of primary MPs pollution (Lofty et al., 2022).

In China, the situation is similar to other regions of the world. It was estimated that more than seven million tons of dry sludge were generated from wastewater treatment in China in

2020 (MEPC, 2017). However, > 80% of this sludge is disposed of improperly (i.e., dumped) with only 2.4% of the sludge applied to land (Yang et al., 2015). Li et al. (2018) investigated the occurrence of MPs in sludge by analyzing 79 sewage sludge samples collected from 28 WWTPs in 11 Chinese provinces. The results showed that on average, the concentration of sludge-based MPs entering the soil and the wider environment in China was estimated to be 1.56×10^{14} particles per year, which is the same order of magnitude of MPs released into European farmland soils (i.e., $8.6 \times 10^{13} - 7.1 \times 10^{14}$ particles per year) (Li et al., 2018; Lofty et al., 2022). Yang et al. (2021) investigated the contributions of three types of sludge (i.e., fresh municipal sludge, mainly industrial sludge, and dry heat-treated municipal sludge), which were repeatedly applied to farmland soil for nine years in Jiangsu province, Southwest China. The results showed that the input of sludge led to an accumulation of MPs in the soil, as high as 149.2 particles kg⁻¹ (compared with the control treatment). These findings confirm that sewage sludge recycling to land represents an important source of plastic pollution in the environment.

At present, the reported treatment methods of MPs in sewage sludge are generally divided into two types: i) physical and chemical methods, and ii) anaerobic digestion methods (Wu et al., 2022). Of these, physical and chemical methods often cause MPs to break into smaller plastic fragments. Whilst there is evidence that some MPs (such as PET and polyurethane reactive, PUR) can be partially degraded under anaerobic digestion (Mahon et al., 2017), most MPs are not degraded, mainly depending on their chemical structure, molecular weight as well as the type of plastic additives in MPs (Moharir & Kumar, 2019). In the future, the technique of efficiently removing MPs from sewage sludge should be developed to reduce the pollution caused by MPs.

The Chinese government has proposed that the daily capacity for harmless treatment of sludge (with moisture content >80%) should be no less than 2.0×10^4 tons by 2025 (Development and Reform Commission of the People 's Republic of China 2022). The harmless treatment rate of urban sludge is expected to reach above 90%. These policies would affect the sources and occurrence of MPs, which can be further investigated in the future study.

3.2.2. Compost

Organic resources such as compost are rich in plant nutrients and organic carbon and are hence widely used as soil amendments to improve soil properties and soil nutrient content (Cherif et al., 2009). However, there is increasing evidence that soils receive plastic input through the application of compost (Bläsing & Amelung, 2018). Because of improper disposal and insufficient waste separation of plastic from organic matter, macro-plastics in compost can accumulate in the soil and risk entering the food chain via crop plants. In China, Zhang et al. (2022) investigated the abundance, shape, composition, and size of MPs from organic fertilizers using attenuated total Fourier transformed infrared spectroscopy. The results showed that mature compost application to agricultural fields goes along with MPs load of $3.5 \times 10^{12} - 6.6 \times 10^{12}$ items per year. Another study in Germany showed that compost application led to an annual input of > 1 mm plastic plastics to arable fields that reached up to 35 billion - 2.2 trillion (Weithmann et al., 2018). In recent years, several countries have strongly encouraged farmers to use organic fertilizers and successively formulated subsidy policies for composts. For instance, the Chinese Ministry of Agriculture provides a subsidy for households of 1,500 RMB ha⁻¹ (equal to 215 US dollars ha-1) to use >3,750 kg ha-1 commercial composts in some pilot areas (Ministry of Agriculture and Rural Affairs of the People's Republic of China, 2018). However, more attention needs to be paid to the potential contribution of compost to macro-plastics and MPs in agricultural soils.

The analysis result shows that plastic mulch films are the most studied potential sources of macro-plastics and MPs in Chinese farmland soils, while compost and sewage sludge may be important sources in Europe, since this type of fertilization is commonly used in European counties.

3.2.3. Food waste digestate

Diverting food waste from landfills to anaerobic digestion can facilitate the conversion of energy into usable forms and produce nutrient-rich soil improvers (Cheong et al., 2020; Xu et al., 2018). However, concerns arise due to the presence of plastic packaging in many food waste streams, which may inadvertently introduce macro- and micro-plastics into agricultural soils (Porterfield et al., 2023). For example, the abundance of MPs was 3.0×10^5 pieces kg⁻¹ in food waste collected from grocery stores in the USA (Golwala et al., 2021), and it was 4.1×10^3 particles kg⁻¹ in the food compost sample in Lithuania (Sholokhova et al., 2022). In addition, some biodegradable plastic packages (e.g. Polylactic acid, PLA) are widely used in food packaging and disposable tableware and the usage of biodegradable plastic packages are increasing (Lu et al., 2022). However, the aging and fragmentation process of PLA also could be enhanced within thermophilic anaerobic digestion with kitchen waste, generating large amounts of macro-plastics and MPs. Research on the occurrence and relative importance of MPs from food waste is in an early stage and this potential pathway of macro-plastics and MPs to agricultural soils needs further clarification (Porterfield et al., 2023).

3.3. Other sources of plastic contamination

3.3.1. Atmospheric deposition

Atmospheric transport and deposition of MPs is one of the major pathways for plastic fragments entering the soil environment (Allen et al., 2019; Brahney et al., 2020). It is estimated that atmospheric deposition rates of MPs range from 1.1×10^4 to 4.1×10^5 items m⁻²yr⁻¹ globally (Allen et al., 2019; Bergmann et al., 2019; Brahney et al., 2020). These particles typically enter the atmosphere through mechanical processes, such as dust entrainment during strong wind events or wave breaking of sea surface spray (Seinfeld et al., 1998). Brahney et al. (2021) created a model to calculate the atmospheric component of the plastic cycle, estimating the current average daily total atmospheric burden (content) of MPs over the land regions of the western United States to be ca. 100 tons. The largest contributor to modeled plastic deposition (84%) in the western United States is road dust. In comparison, agriculturally derived plastics in dust entrained into the atmosphere from agricultural fields are thought to contribute 5% to annual total deposition in the same region (Brahney et al., 2021). In China, Liu et al. (2019) measured indoor and outdoor dust samples collected from 39 major cities of China. The mass concentrations of PET and polycarbonate (PC) MPs were determined, and the concentrations of PET and PC MPs in dust were $1.6 \times 10^{3} - 1.2 \times 10^{5} \,\mathrm{mg \ kg^{-1}}$ and $4.6 \,\mathrm{mg \ kg^{-1}}$ (indoors), $212 - 9,020 \,\mathrm{mg \ kg^{-1}}$ and 2.0 mg kg⁻¹ (outdoors), respectively (Liu et al., 2019). Although it is difficult for MPs > 50 μm to enter the respiratory tract, these particles can enter the gastrointestinal tract where adsorbed contaminants may be released, posing a potential threat to human health (Bergmann et al., 2019; Brahney et al., 2020; Liu et al., 2019).

3.3.2. Rubber tire wear

Rubber is also considered a class of plastic, and physical abrasion of tires significantly contributes to the release of MPs into the environment (Lassen et al., 2015). In addition, tire residues are also present in sewage sludge where road run-off enters the wastewater network (Essel et al., 2015). Rubber MPs size and generation rate from tires depends on their composition (Kole et al., 2017). Evangeliou et al. (2020) found high transport efficiencies of rubber MPs to remote regions worldwide. Their results showed that about 34% of the emitted coarse tire wear particles (TWPs) and 30% of the emitted coarse brake wear particles (BWPs) (100 kt yr⁻¹ and 40 kt yr⁻¹, respectively) were subsequently deposited in the world's oceans. However, knowledge about the fate of tire-derived MPs entering Chinese farmland, especially those from farm machinery, is currently lacking (Evangeliou et al., 2020).



3.3.3. Water-flow and irrigation

Many studies have indicated that large quantities of MPs are present in irrigation source (Chen et al., 2022; Pérez-Reverón et al., 2022). Research showed that the MPs concentration in irrigation water was significantly correlated with the abundance of MPs in agricultural soil, and the MPs concentration of soils in direct contact with irrigation water was significantly higher than that in deeper soils (Katsumi et al., 2021). A study in Spain showed that the shape, color, size and type of MPs in soil samples collected from cropland were similar to those in the irrigation water used on the crops (Pérez-Reverón et al., 2022). This evidence indicates that irrigation water is an important source of MPs in farmland soils. Moreover, comparing MPs abundance in the different source of irrigation water, the concentration of MPs in recycled wastewater (159 items kg⁻¹) was around three times higher than that in the desalinated brackish water (46 items kg⁻¹) (Pérez-Reverón et al., 2022). In addition, the MPs abundance in agricultural soil irrigated by underground water and rainwater is significantly lower than irrigated with surface water (Chen et al., 2022). Meanwhile, the soil is also a potential MPs sink and MPs in soils could be transported by water off into surface water and ocean (Ren et al., 2021). It is important to explore the natural and anthropogenic processes affecting the fate of MPs in irrigation water.

4. Modeling method for quantifying the source of macro-plastics and MPs

4.1. Material flow analysis (MFA)

MFA is a useful tool applied to better understand pathways of substance. It is an analytical method to quantify flows and stocks of materials or substances in a well-defined system (Bornhöft et al., 2016). Several studies have quantified the possible flows of plastic into the soil from different sources by using this method (Kawecki & Nowack, 2020; Liu et al., 2020; Sieber et al., 2020).

MFA has been employed to analyze the flows of plastics in China (Liu et al., 2020; Luan et al., 2021; 2022). However, few studies have focused on plastic emissions and flows in an agricultural context. Of the relevant studies, Zhou et al. (2013) analyzed the emission and accumulation of PVC waste in the environment, and Bai et al. (2018) estimated and predicted the annual input of plastic waste from China into the ocean. Nevertheless, most of these studies have used emission factors to estimate the losses of plastics directly or estimated the amounts of plastic waste through municipal solid waste indirectly, rarely covering all life cycle processes and not distinguishing between plastic types.

Luan et al. (2021) conducted a study to assess MPs and macro-plastics losses throughout the plastic life cycle by using a dynamic MFA approach. The losses were analyzed based on different polymers (including PE, PP, PS), PVC, acrylonitrile-butadiene-styrene (ABS), and PET, sources (including personal care products, laundry process, indoor dust, fishery waste), environmental media (ocean and soil) and lifecycle processes (i.e., production, use, recycling, and end-of-life treatment). Based on field research and published literature, localized emission factors were obtained to systematically and comprehensively estimate the plastic losses to the environment in China. The results showed that MPs and macro-plastics losses entering the environment were 352.1 kt y⁻¹ and 12.7 Mt yr⁻¹. Of these losses, PET accounted for the highest proportion (29.1% and 32.2%), and the net loss to the ocean and soil were 4.0 Mt and 173.7 Mt, respectively in 2020 (Luan et al., 2022).

Based on the research mentioned above, our study summarized the necessary steps of this evaluation method (i.e., MFA) to calculate the main stocks and flows of soil macro-plastics and MPs derived from different sources of agricultural activities. There is an essential process in the MFA where the input of secondary MPs is calculated, i.e., generation rate. According to this rate, the concentration of MPs converted from plastic products can be calculated. As mechanical abrasion (MA) and ultraviolet (UV) irradiation are key factors controlling plastic degradation rates, they are discussed in more details below.

4.1.1. Generation rate

Generation rate is an essential part of MFA, which refers to the mass ratio of MPs generated from macro-plastics. This is a necessary index in the calculation of the flows and stocks of materials in different environments. The methods for calculating generation rates of MPs from macro-plastics vary (Ren et al., 2020; Sieber et al., 2020). Plastic fragmentation may be caused by solar ultraviolet (UV) irradiation, physical abrasion (abiotic), or biological attack (Cole et al., 2011; Barnes et al., 2009). Therefore, it is important to provide an improved estimation of MPs generation in farmlands according to the generation rate of MPs.

Mechanical abrasion (MA). MA of plastics is likely to be common in many cropping environments. For example, plastic packaging of seeds, crops, and fertilizers is abraded by external forces during transportation and use, resulting in plastic debris left in the environment. Further, as mentioned previously, a large amount of plastic debris is left in the soil after the crop harvest due to the fragile nature of the mulch films (Rezaei et al., 2019; Zhao et al., 2017). Farmland may be the most favorable environment for plastic weathering and fragmentation because of photodegradation and MA of plastics by soil (Ren et al., 2020). Studies to measure MPs generation via MA are limited.

Some studies have demonstrated the impact of polymer type on MP generation rate, especially following UV exposure. Song et al. (2017) calculated MPs fragmentation rates *via* the combined effects of MA (using a mechanical roller) and UV exposure for LDPE, PP, and expanded polystyrene (EPS). Their results showed that there was a minimal effect of MA (crushing) on MPs generation from LDPE and PP when the plastic was not exposed to UV (fragmented particle generation was 8.7 and 10.7 particles, respectively), but PP generated more MPs following UV radiation. However, MPs generation from EPS was mainly *via* MA, which resulted in 4220 MPs particles after only 2 months of mechanical friction by a roller without any UV irradiation (Song et al., 2017). Ren et al. (2020) utilized a range of sizes of sandpaper to abrade different components of plastic film to simulate MA and demonstrated a positive relationship between fragmentation rates of different plastic mulch films and relative light transmittance (RLT, %). These studies provide new insight for future calculation of MA rates and MPs production.

Ultraviolet (UV) irradiation. One of the most important degradation factors of plastic polymers fracture is UV radiation, which induces oxidation and molecular chain scission for plastics breakdown in the environment (Laycock et al., 2017; Uheida et al., 2021). The random chain scission and cross-linking of plastic polymers results in the progressive formation of micro-, and nano-size plastic particles from macro-plastics fragments (Qi et al., 2020). Yang et al. (2022) found that when different types of macro-plastics were incubated in soil exposed to the same cumulative UV irradiation (2.1 MJ m⁻²), the average quantity of MPs generated from biodegradable and oxo-degradable plastic mulch films was greater than from conventional non-degradable mulch films. These results help us to understand the kinetics and mechanisms of different types of mulch films (Yang et al., 2022). It is worth noting that the generation of MPs from biodegradable plastics in the terrestrial environment is more significant (Bao et al., 2022; Qin et al., 2021).

4.2. The other quantitation methods of macro-plastics and MPs sources

Several studies have listed the possible flows of plastic into soils based on an analysis of plastic use in agriculture (Brandes et al., 2021; Ren et al., 2021; Wang et al., 2021). The empirical formulas for estimating MPS generation from agricultural plastics are summarized in Table 1. Brandes et al. (2021) used data-driven models alongside data on MPs composition from the literature in combination with national statistics on sewage sludge, compost, and plastic waste production, as well as specialty cropping areas, to estimate the spatial distributions of cumulative MPs mass inputs into agricultural soils in Germany. Based on the Nomenclature of territorial units for statistics (NUTS3) scale, the results showed that MPs input range for soils was 0–15.7kg

Table 1	Empirical	formulae f	or actimating	MPs generation	from	agricultural	plactic films	
Table 1.	Embiricai	Tormulas I	or esumauno	i ivies deneration	irom	adricultural	Diastic IIIms.	

Factors	Formula	References
Wind erosion	$WE_{s} = \sum (I \times T)$	Rezaei et al., 2019
Mechanical abrasion (MA)	u · ·	Ren et al., 2020
Mechanical abrasion (MA)	$EF_{MA} = \frac{RLT - a}{b}$ $M_{MP} = EF_{MA} \times U_{AMF}$	Ren et al., 2021
UV irradiation	$\log I = constant - a \log I$	Yang et al., 2022
/	$MP_{12012}^{SC} = A_{12012}^{SC} \times AP_{2012}^{SC} \times L^{SC} \times FM^{SC}$	Brandes et al., 2021
1	$F(x) = \lambda \alpha x^{\alpha - 1} e^{-\lambda e^{\alpha}}$	Wang et al., 2021

Notes: (1) WE_{g} : the annual wind erosion potential (kg ha⁻¹year⁻¹), I: the soil erodibility potential given a specific wind velocity during 1 h as measured in a wind tunnel (kg ha⁻¹ h⁻¹); T: the wind continuity within a specific velocity (hr year⁻¹); (2) EF: the emission factor of MPs by mechanical abrasion (MA) (mg g⁻¹); RLT: the relative light transmittance of plastic films; a: the parameter of film thickness; b: the parameters of film abrasion degree; (3) M_{MP} : the weight of MPs produced by PMFs (g); U_{AMF}: usage of PMFs (ton/year); (4) I: the irradiance intensity; t: the exposure time; constant: dose-dependent damage degree; a: the constant for specific photographic emulsions; (5) $MP_{i,201}^{SC}$: the amount of MP that remained in the soil per NUTS3 region i in 2012; $A_{i,201}^{SC}$: the area of the respective speciality crop category grown in NUTS3 region i; $A_{i,201}^{SC}$: the fraction of $A_{i,201}^{SC}$ on which plastic mulch film or cover tarp was actually applied in 2012; L^{SC} : the crop specific loss factor; FM^{SC} : the mass of foil per ha (depending on plastic material and foil thickness); (6) F(x): the derivative of the cumulative distribution function (CDF); x: the number of MPs λ : the parament of MPs' relative location; α : parament of MPs' shape.

ha-1, 0-3.79 kg ha-1, and 0-5.18 kg ha-1 from sludge, compost, and plasticulture used in agriculture, respectively. Of these, the contribution of MPs followed the series sewage sludge > compost > plasticulture in Germany (Brandes et al., 2021). Ren et al. (2021) found PMFs contributed 10-30% of the total MPs from all sources in farmland soils according to a Monte Carlo simulation. In addition, Wang et al. (2021) developed a novelty model to calculate the distribution of MPs in the soil environment based on the aging process of different types of MPs (fibers, films, fragments and granules). Based on this model, a distinct downsizing phenomenon from fibers, films, and fragments to granules was observed, and human interference accelerated the fragmentation of MPs. However, the quantitative contribution of different plastic sources to Chinese farmland soil MPs, specifically the rate and kinetics of MPs formation, is still in the primary stage.

4.3. Uncertainty statistics

Quantifying MPs in soil mainly relies on the use of multiple statistical sources of data, which may lead to uncertainty of model outputs. Other sources of uncertainty include soil analytical data, as different studies have used different MPs extraction and detection methods (Bläsing & Amelung, 2018; Li et al., 2020). Accordingly, systematic uncertainty analyses are necessary to confirm the reliability of the method. A unified approach developed by Laner et al. (2016) could be used for characterizing the uncertainty of data based on this method, where the quantitative calculation of the data uncertainties used coefficients of variation (CV) (Laner et al., 2016).

Monte Carlo (MC) simulation can be used to investigate the effects of parameters and data changes, combined with CV (Wang & Ma, 2018). The MC method employs observed data to simulate the distribution, and then the uncertainty range is calculated. This is common in MFA research when there are sufficient data (Luan et al., 2022; Tsai & Krogmann, 2013). It is noteworthy that the MC can be best used if the dataset contains more than 30 records (Montangero & Belevi, 2008).

5. Gaps in knowledge and priorities for future research

Taking China as an example, this research reviewed the source, location, and occurrence of macro-plastics and MPs pollution in soils. It needs to be noted that a limitation of this research is the uncertainty introduced by the plastic data from various studies. In these studies, different

MPs extraction and detection methods are employed, which makes the data less comparable. Therefore, we recommend that future studies could adopt a standardized extraction and detection protocols to reduce this uncertainty. This research also concludes that there are many ways that plastic fragments can enter the soil, including plasticulture practices, irrigation pipes and associated infrastructure, organic wastes and wastewater residues, atmospheric deposition, rubber tire wear and irrigation water. Of these, agricultural plastic mulch films are the most important source of macro-plastics and MPs in Chinese farmland soils, while wastewater-derived sludge may be more important in European countries. Furthermore, a MFA modeling approach can be used to quantify the flows and stocks of macro-plastics and MPs. In terrestrial systems, research on MPs is growing rapidly. However, more real-world studies are needed to narrow the knowledge gaps in the following aspects:

- 1. Tracking the transport and fate of MPs from different sources in soil. At present, most of the quantitative studies are estimated by models, with few actual field measurements. The use of isotopically labeled plastics would be useful to track their fate, stocks, and their biodegradation rates. In addition, current research is also restricted by the limited availability of published data. Consequently, more samples to be required from soil, irrigation water, compost and sludge, and other potential sources in the typical regions using plastic films. This will help better parameterize models and reflect regional conditions.
- 2. There is still a lack of information on the impacts of MPs on soil health, especially regarding non-point sources. Furthermore, to compare soil MPs concentrations and forms of MPs, sampling, extraction, and detection methods should be standardized. In addition, most soil MPs contents are expressed as the number of MPs per unit weight of soil (e.g. items kg⁻¹), rather than the mass of MPs per unit weight of soil (e.g. mg kg⁻¹). Future research should consider the use of mass units more often, to allow a more accurate description of the mass transfer of MPs through the environment and organisms (i.e. humans, livestock and soil fauna).
- 3. Establishing spatially explicit models to predict the transfer and fate of MPs entering soils, linking specific source inputs to movement within the landscape and soil profile. These models need to account for different soils, climates, and management practices and describe MPs migration, generation rates, and the ultimate fate. Such models could then be used to underpin practical guidance to farmers and regulators to promote more sustainable use of plastics, and their alternatives, in agriculture.
- 4. Strengthening research on the influence of residual plastic film or MPs on crop growth, yield and crop quality. The mechanistic basis of how residual plastic film or MPs affect nutrient cycling in the soil also needs to be explored, to determine the optimal use of plastic mulch films for different crops. Further efforts are also needed to improve the efficacy of the retrieval of used plastic mulch films after crop harvest.
- 5. Strengthen the research on the impact of agricultural plastic use and recycle policies. The formulation of policies can greatly improve the standard of farmers' use of agricultural plastics. In China, for example, the governments have introduced various regulations and incentive mechanisms following research on the hazards of plastic film residues in recent years. In February 2022, the National Development and Reform Commission and the Ministry of Ecology and Environment issued the "Key Points for the Treatment of Plastic Pollution", which called for standardize use and recycling of plastic film, focusing on the promotion and application of biodegradable plastic film and thickened high-strength plastic film in key film areas. Through improvements to the recycling network system and effectively controlling agricultural film pollution in farmland, the recovery rate of agricultural film should be stabilized at more than 80%. In addition, the "Opinions on Further Strengthening the Treatment of Plastic Pollution" issued by the Central Office of the Reform and Reform prohibited the production, sale and use of several specific plastic products



(e.g. plastic shopping bags with a thickness of less than 25 µm, PE agricultural mulch film with a thickness of less than 10 µm) in 2019, and the "Notice on Solidly Promoting the Treatment of Plastic Pollution" issued in 2020 by the National Development and Reform Commission, have clearly required the strengthening of the plastic prohibition and restriction policies in the fields of agriculture, retail and catering. These show that China is making efforts to reduce the environmental pollution caused by plastic waste from the aspect of policy control.

Discussion the economic evidence of plastic pollution. Based on the survey results of 1067 cotton farmers in Xinjiang Province, China (Liu et al., 2020), most farmers are willing to recycling plastic film rather than incineration or landfill with the supports, such as reducing input cost of agricultural plastic film recycling by government subsidies, improving the mechanization of agricultural film recycling and setting up lectures and training about use and recycling of agricultural plastic film. In addition, there is already evidence suggesting extensive negative impacts of plastic waste on ecosystem services (Beaumont et al., 2019). Researchers have estimated the loss of 1 - 5% in marine ecosystem services due to plastic pollution. This decrease equates to about \$0.5 to \$2.5 trillion per year. In other words, each metric ton of plastic waste costs about $$3.3 \times 10^4$ (Beaumont et al., 2019). This illustrates that plastic pollution is not just damaging the environment, but that the economic losses caused by plastic pollution can also be substantial.

6. Conclusions

This review summarized the potential sources, current concentration and abundance, and modeling methods of quantification and flows of macro-plastics and MPs pollution in soils. Plastic pollution appears to be ubiquitous in soil and evidence suggests that it may represent an environmental risk to soil quality and soil functioning. Furthermore, as China is the biggest agricultural plastic film manufacturer and user it will face more serious pollution risks from legacy plastic in the farmland soil. Therefore, there is an urgent need to quantify the emission of plastic debris from different agricultural activities to the environment and to control the input of plastic from different sources into farmland soil. In addition, tracking the fate of MPs and NPs in soils, analyzing the impact of plastic waste on soil ecological health based on realistic concentration of MPs and NPs in the field, and studying the influence of policy and economic evidence on plastic pollution also deserve of future research.

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