

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

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

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33 **Potential sources and occurrence of macro-plastic and microplastics pollution in farmland**
34 **soils: A typical case of China**

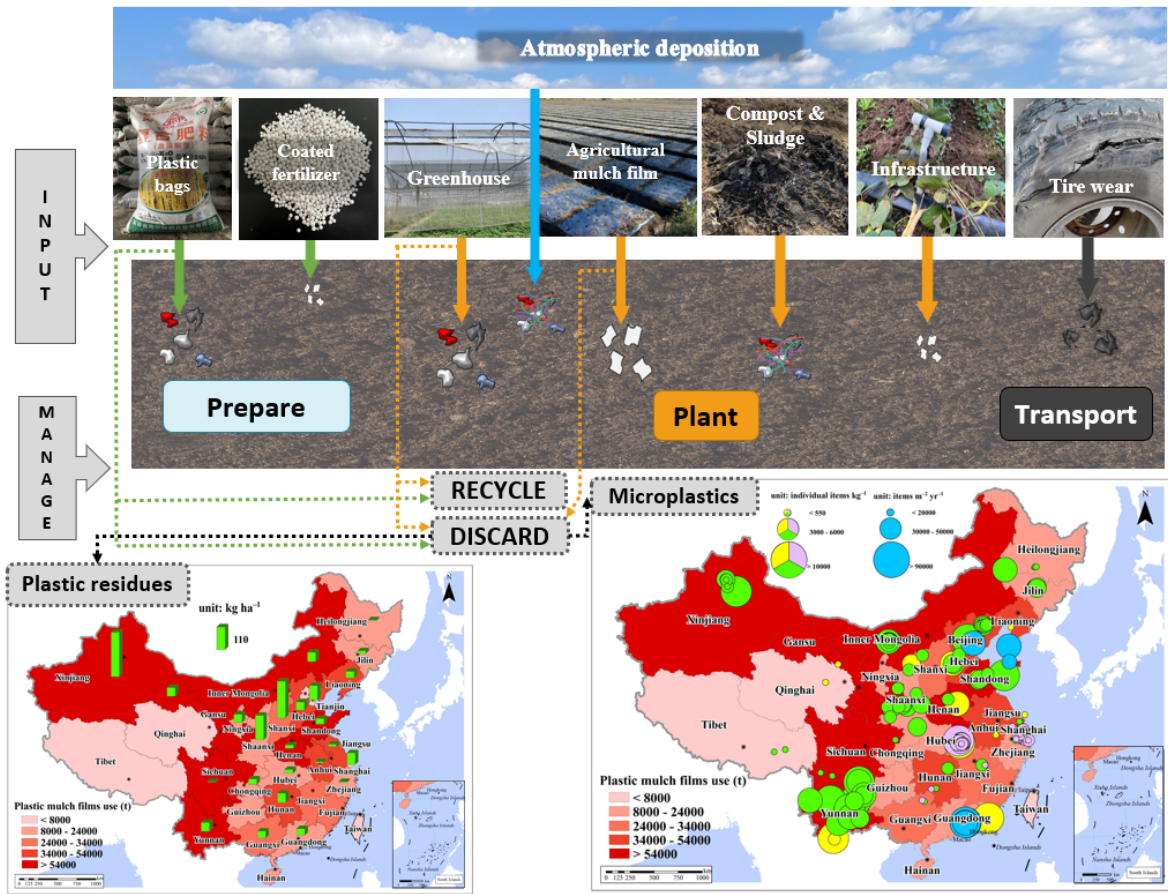
35 **Abstract**

36 Plastic debris (including macro-plastics, microplastics (MPs), and nanoplastics), defined
37 as an emerging contaminant, has been proven to significantly affect soil ecosystem
38 functioning. Accordingly, there is an urgent need to robustly quantify the pollution
39 situation and potential sources of plastics in soils. China as the leading producer and user
40 of agricultural plastics is analysed as a typical case study to highlight the current situation
41 of farmland macro-plastics and MPs. Our study summarized information on the
42 occurrence and abundance of macro-plastics and MPs in Chinese farmland soils for the
43 first time based on 163 publications with 728 sample sites. The results showed that the
44 average concentration of macro-plastics, and the abundance of MPs in Chinese farmlands
45 were 103 kg ha⁻¹ and 4537 items kg⁻¹ (dry soil), respectively. In addition, this study
46 synthesized the latest scientific evidence on sources of macro-plastics and MPs in
47 farmland soils. Agricultural plastic films and organic wastes are the most reported sources,
48 indicating that they contribute significantly to plastic debris in agricultural soils.
49 Furthermore, the modelling methods for quantifying macro-plastics and MPs in soils and
50 estimating the stock and flow of plastic materials within agricultural systems were also
51 summarized.

52

53 **Keywords:** macro-plastics, microplastics, abundance, source apportionment, quantitative
54 **method, farmland soils**

55 Graphical abstract



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93 **1. Introduction**

94 Plastics are widely used across almost all sectors of society due to their versatility relatively cheap
95 cost, light weight and durability (Jambeck et al., 2015; Plastics Europe-The Facts 2022). The
96 production of plastics is increasing with global cumulative production predicted to reach up to 33
97 billion tons by 2050 (Sharma et al., 2020). As most plastics have a relatively short functional
98 lifespan, the disposal of plastics represents a major global problem with a large proportion of
99 plastics not being recycled (Luan et al., 2021; Wang et al., 2021). Currently, it is estimated that
100 79% of plastic waste enters either landfills or the natural environment where it represents a threat
101 to terrestrial, freshwater, and marine ecosystems (Geyer et al., 2017). Although macro-plastics
102 (particles size > 5 mm) represent the primary type of waste entering the environment, they
103 gradually degrade into smaller fragments in response to ultraviolet (UV) irradiation, mechanical
104 abrasion, and biodegradation (Barnes et al., 2009; Yang et al., 2022). Microplastics (MPs) are
105 defined as particles < 5 mm and > 1 μm , including fragments, fibers, particles, foams, and films,
106 while plastic particles with the size between 1 nm and 1 μm are defined as nanoplastics (NPs)
107 (Frias & Nash, 2019; Thompson et al., 2004). Since MPs and NPs are small in size, and present
108 in large quantities and degrade slowly, they are easily absorbed, inhaled, or ingested by organisms,
109 leading to bioaccumulation (Barnes et al., 2009; Leslie et al., 2022; Wu et al., 2022). Studies have
110 found the presence of MPs and NPs in plants, soil fauna, human feces, and blood (Leslie et al.,
111 2022; Li et al., 2020; Lwanga et al., 2017; Zhang et al., 2021). Current evidence indicates that
112 MPs can be transferred to the human body through the food chain as well as via inhalation and
113 are likely to give rise to a range of cytotoxic effects that are now becoming evident, albeit still
114 incomplete (Wu et al., 2022; Brachner et al., 2020; Hua et al., 2022).

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

137 through the terrestrial food chain, causing a potential threat to livestock and human health, and
138 natural ecosystem food webs (Lwanga et al., 2017; Zhou et al., 2021).

139 MPs can also be vectors for the attachment and transmission of other contaminants (e.g.
140 hydrophobic organic contaminants, heavy metals, harmful microorganisms), posing a potential
141 threat to (human, livestock, soil fauna) organismal health and the wider environment
142 (Brennecke et al., 2016; Wang et al., 2019; Zettler et al., 2013). For example, the heavy metals
143 cadmium (Cd) and lead (Pb) were detected in MPs samples (n = 924) from two beaches in
144 Southwest England with maximum concentrations of 3.4 and 5.3 mg g⁻¹, respectively (Massos
145 & Turner, 2017). In addition, they evaluated the maximum bio-accessible concentrations of Cd
146 and Pb in the proventriculus-gizzard of seabirds (*Fulmarus glacialis*) and found concentrations
147 exceeded the safe dietary intake limit by a factor of about 50 and 4, respectively (Massos &
148 Turner, 2017). Furthermore, high concentrations of zinc (Zn, 9407 mg kg⁻¹) and polycyclic
149 aromatic hydrocarbons (PAH, 47 mg kg⁻¹) have been detected in MPs samples recovered from
150 earthworms and the surrounding soil, and MPs exposure resulted in the steep rise of the
151 abundance of pathogenic microorganisms in the worm intestinal tract (Ding et al., 2020).

152 Many studies have indicated that the additives contained in MPs may represent a greater
153 threat to terrestrial ecosystems than the plastic polymer itself (Hahladakis et al., 2018; Halden,
154 2010). There have been very few studies on the effects of MPs on human health, however, some
155 of the additives used in plastics manufacturing, e.g. plasticizers and heavy metals, have been
156 shown to interfere with gene expression, cell metabolism (e.g. signal transduction, enzyme
157 function), and animals and humans development as well as reproduction (e.g. endocrine
158 disrupting properties) (Rist et al., 2018). For instance, the common plasticizer of bisphenol A

159 (BPA), as an endocrine disruptor, could disrupt the endocrine system and various functions of
160 organisms, including the thyroid, reproductive system, and metabolism (Halden, 2010).

161 As plastic waste (including macro-plastics and MPs) in terrestrial environments poses a
162 potential threat to food security, human health, and the health of our natural environment, it is
163 important to control plastic input and manage legacy plastic in soils. Greater effort is needed to
164 quantify the sources of macro-plastics and MPs and the fate of different plastic fragments. A
165 study by Jambeck et al. (2015) reported that 80% of marine plastic residuals arise from land,
166 suggesting that soil is not only an important sink of MPs but also an important source (Rachman,
167 2018). The accumulation of macro-plastics and MPs waste in soils is the result of various human
168 activities and environmental origins, such as discarded plastic litter (Rillig, 2012), plasticulture
169 practices (e.g. plastic mulch films, greenhouse films, irrigation pipes, and associated
170 infrastructure) (Bläsing & Amelung, 2018, Gündoğdu et al., 2022; Huang et al., 2020; Wang et
171 al., 2022), sewage sludge application (Long et al., 2019), coated fertilizers (Katsumi et al.,
172 2021), organic fertilizer and agricultural compost (Weithmann et al., 2018), atmospheric
173 deposition (Allen et al., 2019), digested food waste (Porterfield et al., 2023), and rubber tire wear
174 (Evangelidou et al., 2020). Of these, plastic films represents an important source of macro-
175 plastics and MPs in agricultural soils and has attracted extensive research and discussion (Qi et
176 al., 2020). Plastic films are used extensively throughout the world in both horticulture and
177 arable cropping (e.g. mulch films and polytunnels) as well as within livestock production (e.g.
178 silage wrapping). Typically, these plastics are not recycled efficiently for several reasons
179 including (i) difficulty in removing them from the soil after use, (ii) contamination by soil and
180 vegetation residues, (iii) loss from the field due to wind erosion, (iv) lack of recycling

181 infrastructure, and (v) poor financial incentives (Li et al., 2021; Mekonnen et al., 2016). These
182 barriers to recycling have led to the accumulation of legacy plastic in soils.

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

197

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

199 China is suffering from serious plastic pollution, particularly within agricultural environments
200 where recycling is problematic (Plastics Europe-The Facts 2022; Qi et al., 2020). In 2021, the
201 use of plastic mulch film (PMFs) in China was 1.3 million tons, representing 75% of global plastic
202 agri-use (NBSC, 2019; Yan, 2022). Further, the area covered by plastic mulch film in China in

203 2021 was 17.3 million hectares, equivalent to 70% of the area of the United Kingdom (NBSC,
204 2022). However, the recovery rate of plastic mulch film from the field at the end of cropping
205 was < 60% (Zhao et al., 2017). Because of the low plastic film thickness of 6–8 μm commonly
206 used in China, it is difficult to completely retrieve it from soil (Yan et al., 2006), and what is
207 retrieved is contaminated with soil, limiting opportunities for recycling. Based on previous
208 studies, summary information on the occurrence and potential sources of macro-plastics and MPs
209 is important to control them in agricultural soils.

210

211 ***2.1 Literature search and data collection***

212 To understand the distribution of soil macro-plastics and MPs in China's agricultural soils, this
213 review searched the literature from three scientific databases (i.e. Web of Science, EI Compendex,
214 and China Knowledge Resource Integrated Database) for "search terms" including (plastic
215 residue or plastic debris) and (macro-plastic or microplastic or nanoplastic) and (soil or terrestrial)
216 (specific search strings in Table S1). The literature search was limited to papers published before
217 the 31st of January 2022. In summary, these papers were chosen according to the following
218 selection criteria: (a) the study must be practical measurement data, without extra addition of non-
219 agricultural plastics and MPs; (b) these sampling data must have a specific location name (to
220 cities or counties) or longitude and latitude; (c) the samples must be collected in the farmland soil
221 of China; (d) must be collected in bare soil, rather than in the greenhouse. Finally, 163 articles
222 (including 123 studies for macro-plastics and 40 studies for MPs) were selected from more than
223 4,800 publications using strict inclusion criteria (Table S2 and S3). Details of search strings and
224 the number of publications were presented in Table S1 and S4 in Supporting Information. The

225 database covered 30 provinces with 728 data points to show the situation of macro-plastics and
226 MPs in Chinese agricultural soils (Figure 1 and 2). However, there is a limitation that due to
227 the longitude and latitude of some points being located close together, they overlapped and
228 appeared to only be one location presented in Figure 2. This situation was shown in Jiangsu
229 (data num = 5), Guangdong (13), Jilin (7), Hebei (16), Heilongjiang (4) and Inner Mongolia
230 (13) Province.

231

232 ***2.2 Distribution and occurrence of macro-plastics and MPs in Chinese farmlands soil***

233 *2.2.1 The occurrence of macro-plastics in Chinese farmlands soil*

234 The concentration of macro-plastics was investigated in 24 provinces of China collected from
235 123 articles (see the list of articles in Table S2) as shown in Figure 1. The concentration of
236 macro-plastics found in soil (0 – 80 cm depth) varied from 0.2 to 421.6 kg ha⁻¹ with an average
237 value of 103.3 kg ha⁻¹, with the median of 54.7 kg ha⁻¹. The province's highest concentration
238 (421.6 kg ha⁻¹) was found in Xinjiang province (Northwestern China). The highest average
239 concentration of macro-plastics (142.7 kg ha⁻¹) was found in Northwestern China (e.g. Xinjiang,
240 Ningxia, and Gansu province), followed by 37.4 kg ha⁻¹ in Northern China (e.g. Hebei,
241 Shandong, and Shanxi province), and 30.7 kg ha⁻¹ in the soils of Southern China (e.g. Hubei,
242 Yunnan province and Shanghai) (Table S2). This result was consistent with previous results that
243 investigated 384 soil samples collected from 19 provinces, and the macro-plastics concentrations
244 in the typical areas covered with mulch films soil samples ranged from 0.1 to 324.5 kg ha⁻¹ and
245 the highest concentration was observed in Northwestern China (i.e. Xinjiang, Gansu province and
246 Inner Mongolia) (Huang et al., 2020). More than 40% of soil samples (n = 51) were from the

247 plough layer, i.e. a depth of 0 – 30 cm. Of these samples, 49% (n = 25) were divided into three
248 soil layers: 0 – 10, 10 – 20, and 20 – 30 cm. Based on an assumed film thickness of 6 – 8 μm
249 and density of 0.910 – 0.925 g cm^{-3} (low-density polyethylene, LDPE), a weight of 103 kg ha^{-1}
250 ¹ macro-plastics corresponded to an area of $1.4 - 1.9 \times 10^4 \text{ m}^2 \text{ ha}^{-1}$ (Yan et al., 2006). The region
251 in Northwestern China is a typical area where plastic films are used widely, e.g. Shaanxi
252 province (21 thousand tons of plastic mulch films used in 2021) and Gansu province (122
253 thousand tons of plastic mulch films used in 2021) (NBSC, 2022). The preference for using
254 plastic mulch films in these regions is to conserve soil water and increase soil temperature in
255 maize, cotton, peanut, potato, and other vegetable cropping (Dong et al., 2015; Gao et al., 2019).
256 As the most important cotton-producing area in China, Xinjiang province uses the largest
257 amount of plastic mulch film (240 thousand tons per year in 2021, NBSC, 2022). The region of
258 Southern China has adequate water and mild temperatures for crop growth without the need for
259 plastic mulch films. Nevertheless, there are some exceptions. For example, Yunnan province,
260 which is located in the Southwestern of China with the largest flower and tobacco production,
261 used 90 thousand tons of plastic mulch films for the cultivation of cash crops cultivation in
262 2021, exceedingly more than 80% Chinese provinces (NBSC, 2022). The average amount of
263 macro-plastics in this area was relatively high at 44 kg ha^{-1} . It is noted that the value of median
264 of macroplastics was consistent with previous results of plastic residues (55 kg ha^{-1}) from the
265 second national pollution source census of China, but the average number slightly exceeded the
266 value (Atlas of the Second National Agricultural Pollution Source Census, 2022). This may be
267 due to differences of study area and data sources. This study included 24 province with the data
268 from 1991 to 2021, nevertheless the result of the second national pollution source census

269 included 31 provinces and based on the samples collected in 2017.

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

274 275 *2.2.2 The occurrence of MPs in Chinese farmlands soil*

276 The abundance of MPs was investigated in 28 provinces of Chinese farmland through the data
277 collected from 40 articles (see the list of articles in Table S3) shown in Figure 2. The abundance
278 of MPs varied from 1.6 to 6.2×10^5 items kg^{-1} (dry soil) with an average abundance of 4536.6
279 items kg^{-1} , with the median of 1640.0 items kg^{-1} . The highest average MPs abundance (4,817.9
280 items kg^{-1}) was found in Southern China, followed by 4,156.1 items kg^{-1} in Northern China,
281 3,602.7 items kg^{-1} in Northwestern China, and 82.3 items kg^{-1} in the Qinghai–Tibet Plateau
282 (Table S3). Interestingly, Northwestern China with a higher concentration of macro-plastics was
283 not the highest abundance of MPs. This might be due to the lower temperatures and plough
284 frequency in Northwestern China, causing the lower generation rate from macro-plastics to MPs.
285 The most commonly researched regions for MPs were Hubei province (abundance of MPs from
286 327.5 to 6.2×10^5 items kg^{-1}), and Shanghai city (abundance of MPs from 1.6 to 2153.0 items
287 kg^{-1}) (see more information in Table S3). The number of sampling points in these two regions
288 was the highest, with 59 (Hubei province) and 123 (Shanghai city), indicating that more
289 research of MPs has been carried out in Hubei province (Zhou et al., 2019; Wang et al., 2021)
290 and Shanghai City (Lv et al., 2019; Zhou et al., 2020) compared to other Chinese regions.

291 However, there were only a few studies of MPs in Northwestern China (such as Xinjiang and
292 Gansu provinces), where the concentration of macro-plastics was relatively high. Therefore,
293 further studies are recommended that focus on quantifying terrestrial MPs pollution in
294 Northwestern China.

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

307

308 ***2.3 The potential sources of MPs in Chinese farmlands soil***

309 Most of the macro-plastics in farmland soils come from the damage of agricultural plastic
310 products. However, the sources of MPs in soils are much more various. In the last 10 years, the
311 agricultural sector has become increasingly important as a source of MPs in soils. The most
312 potential agricultural sources (i.e. agricultural mulch films, compost and sewage sludge, and

313 atmospheric deposition) have been focused and mapped the distribution and abundance of MPs
314 from these sources in China (Figure 2 and Table S3). It should be noted that the data points of
315 MPs caused by other sources, such as coated fertilizer and food waste (which are not well
316 investigated in previous literature but may contribute to MPs in soils) are presented in yellow
317 circles in the revised Figure 2.

318 This research explored the distribution and abundance of MPs from the agricultural plastic
319 films with 146 sampling points (Figure 2(green)). It highlighted that most of the research on
320 agricultural plastic films has focused on the regions of Northern and Northwest China (Zhang et
321 al., 2021; Wang et al., 2021). In contrast, most of the research on compost and sewage sludge was
322 concentrated in Southern China as shown in Figure. 2(purple) with 57 soil samplings, where there
323 are more wastewater treatment plants and larger quantities of sludge production (Yang et al.,
324 2021). Only a few cities have measured MPs in the atmosphere, including Shanghai, Dongguan,
325 Dalian, Tianjin, Wenzhou, and Yantai, with a total of 15 samples as shown in Figure. 2(blue). The
326 average abundance of MPs from agricultural plastic films, composts and sludge, and atmospheric
327 deposition were 4,231.1 items kg⁻¹, 1,002.3 items kg⁻¹, and 7.9 × 10⁴ items m⁻² yr⁻¹. Taking
328 into account all the potential sources on MPs, plastic mulch films represent the most important
329 source in Chinese farmland soils.

330

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

332 The sources of macro-plastics mainly consist of input from improper disposal of agricultural
333 plastic practices and solid waste (e.g., domestic waste) from the surroundings of farmland soil
334 (Hurley & Nizzetto, 2018; Qi et al., 2020). In contrast, the sources of MPs are more complex.

335 MPs are generally categorized into two types: primary and secondary MPs. Primary MPs mainly
336 refer to micro-size plastic particles that are manufactured intentionally for commercial uses, and
337 they often act as raw materials for industrial production (Bläsing & Amelung, 2018; Yang et al.,
338 2021). The rapid growth in the fibers from man-made textiles, microbeads from personal care
339 products, and fragments produced during the plastic manufacturing process mean that they are
340 significant sources of primary MPs (Fu & Wang, 2019). Furthermore, increasing quantities of
341 primary MPs are being introduced to agricultural soils via organic wastes and wastewater residue
342 input (Yang et al., 2021). However, the most common MPs in the environment are secondary MPs
343 (Qi et al., 2020; Cole et al., 2011). Secondary MPs are generated from the degradation and
344 decomposition of larger macro-plastics products and debris into micro-nanoplastics by abiotic
345 (e.g. high temperature, wind-blown and ultraviolet radiation, (Horton et al., 2017; Rezaei et al.,
346 2019) and biotic factors (e.g. microbial decomposition, Yuan et al., 2020). Compared to primary
347 MPs, secondary MPs are more difficult to determine the source and rates of generation. The
348 following sections of this study provide a summary of the potential sources and quantitative
349 estimate modelling methods for macro-plastics and MPs.

350

351 ***3.1 Plasticulture practices***

352 ***3.1.1 Plastic mulch films***

353 Plastic mulch films are used for several reasons that improve crop yields, e.g. for increasing the
354 soil temperature and water use efficiency, promoting seed germination, inhibiting weed growth,
355 and reducing soil erosion (Gao et al., 2019). However, with the increasing use of agricultural
356 plastics film, especially in several developing countries (e.g. China, India, Egypt, and Vietnam),

357 there is an increasing legacy of plastic mulch film residue accumulation, including macro-
358 plastics and MPs (Plastics Europe-The Facts 2022; Maraveas, 2020).

359 In Asia, the largest usage of agricultural films is China with consumption of 2.5 million tons,
360 accounting for > 70% of Asia's, and almost 50% of the worldwide in 2018 (NBSC, 2019; Le
361 Moine, 2018; FAOUN, 2021). The use of plastic mulch film in China has increased nearly three
362 times from 375 thousand tons in 1993 to 1320 thousand tons in 2021, however, the recovery
363 rate after crop harvest is under 60% (Zhao et al., 2017). This equates to around 1.18 million
364 tons of legacy plastic (equivalent to the area of 0.16–0.22 million km²) that has been left in the
365 soil over this period ([Atlas of the Second National Agricultural Pollution Source Census, 2022](#)).

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

376 Biodegradable plastic mulches (BDMs) have been developed as substitutes to conventional
377 PE mulch films and are formulated to reduce the persistence of residues in soil (Yang et al., 2022).
378 Because of the growing awareness of the persistence of synthetic plastic mulch films in the

379 environment, BDMs have gradually entered the mulch film market in China (Plastics Europe-
380 The Facts 2022; He et al., 2018). However, studies have reported that MPs formation is more
381 rapid from biodegradable mulch than from traditional non-degradable mulch films. For example,
382 plastic films were exposed to UV irradiation of 2.1 MJ m^{-2} in a lab experiment by Yang et al.
383 (2022). This level of UV exposure simulated the cumulative irradiance level of 70 days of
384 natural summer solar light in Northern China. The average quantity of MPs released from
385 biodegradable, and non-degradable mulch films were 475, and 155 particles cm^{-2} , respectively.

386 In summary, the mulching duration, amounts of mulch films and plastic material are
387 important factors that affect the plastic fragmentation. As an important source of macro-plastics
388 and MPs in agricultural soil, the level of plastic accumulation by agricultural plastic mulch films,
389 especially BDMs, is alarming but has received relatively little attention to date. A few studies
390 have shown that BDMs can contribute more MPs to soil compared with conventional PE mulch
391 films at the same time period (Yang et al., 2022; Zhou et al., 2023). Therefore, further research
392 is needed to investigate the fate and generation process of MPs from mulch films, including
393 BDMs.

394

395 *3.1.2 Greenhouse films*

396 Greenhouses represent the largest proportion of agricultural plastic films used in plant production
397 worldwide (Le Moine, 2018). It was estimated that the global average quantities of greenhouse
398 films used 3500 kg ha^{-1} in 2019, which represented 47% of agricultural film demand (Le Moine,
399 2018; FAOUN, 2021). Greenhouses are used to prolong the growing season in temperate regions
400 of the world, and most plastic greenhouses are concentrated around Asia (FAOUN, 2021). Since

401 there is no direct amount of greenhouse films used in China and agricultural films is mainly used
402 as mulch films and greenhouse films in China, the amount of greenhouse films is represented by
403 the difference between the amount of agricultural films and mulch film (Zhang et al., 2021). In
404 2021, China's use of greenhouse films reached 1.04 million tons, accounting for 44% of the total
405 amount of agricultural film (NBSC. 2022).

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

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

423

424 *3.1.3 Irrigation pipes and associated infrastructure*

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

428 A recent study in Turkey showed the concentration of MPs from disposable greenhouse
429 plastic films and irrigation pipes in agricultural soil ranged from 0.3 to 32 particles kg⁻¹ with an
430 average of 11.1 particles kg⁻¹ (Gündoğdu et al., 2022). Furthermore, MPs with additives could
431 act as vectors for pollutants, e.g. dibutyl phthalate which has been shown to be released from PVC
432 pipe fragments in water, representing an added risk to both organisms and the environment (Ye
433 et al., 2020). In addition, the pollution level is highly correlated with the amount of disposable
434 drip irrigation pipes and greenhouses in the contaminated sites (Katsumi et al., 2021; Ye et al.,
435 2020). Results showed that irrigation pipes and associated infrastructure could be potential
436 sources of MPs in irrigated farmland soils, which should be taken into account when estimating
437 the concentration of MPs in soil (Gündoğdu et al., 2022; Pérez-Reverón et al., 2022). Furthermore,
438 the topics of lifespan and waste management of agricultural plastic infrastructure and source
439 identification of MPs in soil based on their physical properties (e.g. size, shape, and the type) are
440 also worthy of further study.

441

442 *3.1.4 Coated fertilizer use*

443 Polymer-coated fertilizer (PCF) comprises a nutrient core wrapped by a polymer coating and is
444 designed to release nutrients to plants at a gradual and controlled rate (Du et al., 2006). PCF is

445 composed of microcapsules with a thickness of 10 – 80 μm and a diameter of 2 – 5 mm, which
446 are not recovered after use. These microcapsules are primary MPs and can further degrade into
447 NPs (Katsumi et al., 2021; Bian et al., 2022; Trenkel 2010). The use of PCF in China is increasing
448 at a rate of 10% – 15% per year (Li et al., 2022). It is expected that the output of the Chinese PCF
449 will reach 7.6×10^6 – 11.3×10^6 tons by 2025, with the microcapsules input to soils potentially
450 amounting to 0.4×10^6 – 0.6×10^6 tons (Yang et al., 2009).

451 Although PCF can reduce nutrient leaching loss and ammonia emissions, the fate and impact
452 of the residual polymer coating are attracting the attention of fertilizer companies and
453 environmental researchers (Lian et al., 2021). For example, Katsumi et al. (2021) investigated the
454 accumulation of microcapsules derived from coated fertilizer in 19 paddy fields in Japan with
455 concentrations found to range from 6 – 369 mg kg^{-1} (mean 144 mg kg^{-1}). The result showed that
456 legacy plastics from microcapsules will continue to accumulate in farmland soil as long as
457 conventional PCF is used. The spatial distribution of MPs from PCF is also strongly affected by
458 irrigation, and the soil around drainage outlets has been found to be a hot spot (Katsumi et al.,
459 2021). Several studies have also measured the release of macro-plastics in the environment by
460 PCF made from different co-polyesters (Lubkowski et al., 2016). For example, experiments have
461 shown that the residual amount of PCF film shells left in the soil was 50 kg ha^{-1} every year, which
462 accounting for 50% of the average annual nutrient consumption input in the European Union (100
463 kg ha^{-1}) (Lubkowski et al., 2016). Furthermore, it is estimated that the concentration of macro-
464 plastics from PCF in the soil can reach 500 kg ha^{-1} after continuous application of PCF for 10
465 years (Li et al., 2022). Whilst previous research has focused on the benefits of PCF on plant
466 growth, soil properties, soil microbial communities, and reduced risk of nutrient losses to water

467 and air, further attention is needed to assess the contribution of this source to plastic pollution in
468 China (Bian et al., 2022; Lian et al., 2021). The release of MPs may become a potential food
469 safety problem for the long-term application of PCF in farmland.

470

471 ***3.2 Organic wastes and wastewater residue input***

472 *3.2.1 Sewage sludge*

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

481 In Europe and North America, about 50% of sewage sludge is processed for agricultural use,
482 and it is estimated that 125 – 850 tons of MPs per million peoples are added annually to European
483 agricultural soils either through direct application of sewage sludge or as processed biosolid
484 (Nizzetto et al., 2016). A recent study by Lofty et al. (2022) estimated a maximum application
485 rate of 4.8 g of MPs m⁻² yr⁻¹ or 1.15 × 10⁴ MPs particles m⁻² yr⁻¹ in Europe from sewage sludge
486 applied to agricultural soil by measuring the MPs content of sewage sludge at wastewater
487 treatment plants (WWTPs). These studies strongly suggest that the practice of spreading sludge
488 on agricultural land could potentially make them one of the largest global reservoirs of primary

489 MPs pollution (Lofty et al., 2022).

490 In China, the situation is similar to other regions of the world. It was estimated that more
491 than seven million tons of dry sludge were generated from wastewater treatment in China in 2020
492 (MEPC, 2017). However, > 80% of this sludge is disposed of improperly (i.e. dumped) with only
493 2.4% of the sludge applied to land (Yang et al., 2015). Li et al. (2018) investigated the occurrence
494 of MPs in sludge by analyzing 79 sewage sludge samples collected from 28 WWTPs in 11
495 Chinese provinces. The results showed that on average, the concentration of sludge-based MPs
496 entering the soil and the wider environment in China was estimated to be 1.56×10^{14} particles per
497 year, which is the same order of magnitude of MPs released into European farmland soils (i.e. 8.6
498 $\times 10^{13} - 7.1 \times 10^{14}$ particles per year) (Li et al., 2018; Lofty et al., 2022). Yang et al. (2021)
499 investigated the contributions of three types of sludge (i.e. fresh municipal sludge, mainly
500 industrial sludge, and dry heat-treated municipal sludge), which were repeatedly applied to
501 farmland soil for nine years in Jiangsu province, Southwest China. The results showed that the
502 input of sludge led to an accumulation of MPs in the soil, as high as 149.2 particles kg^{-1}
503 (compared with the control treatment). These findings confirm that sewage sludge recycling to
504 land represents an important source of plastic pollution in the environment.

505 At present, the reported treatment methods of MPs in sewage sludge are generally divided
506 into two types: i) physical and chemical methods, and ii) anaerobic digestion methods (Wu et
507 al., 2022). Of these, physical and chemical methods often cause MPs to break into smaller
508 plastic fragments. Whilst there is evidence that some MPs (such as PET and polyurethane
509 reactive, PUR) can be partially degraded under anaerobic digestion (Mahon et al., 2017), most
510 MPs are not degraded, mainly depending on their chemical structure, molecular weight as well

511 as the type of plastic additives in MPs (Moharir & Kumar, 2019). In the future, the technique
512 of efficiently removing MPs from sewage sludge should be developed to reduce the pollution
513 caused by MPs.

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

519

520 *3.2.2 Compost*

521 Organic resources such as compost are rich in plant nutrients and organic carbon and are hence
522 widely used as soil amendments to improve soil properties and soil nutrient content (Cherif et al.,
523 2009). However, there is increasing evidence that soils receive plastic input through the
524 application of compost (Bläsing & Amelung, 2018). Because of improper disposal and
525 insufficient waste separation of plastic from organic matter, macro-plastics in compost can
526 accumulate in the soil and risk entering the food chain via crop plants. In China, Zhang et al.
527 (2022) investigated the abundance, shape, composition, and size of MPs from organic fertilizers
528 using attenuated total Fourier transformed infrared spectroscopy. The results showed that mature
529 compost application to agricultural fields goes along with MPs load of $3.5 \times 10^{12} - 6.6 \times 10^{12}$
530 items per year. Another study in Germany showed that compost application led to an annual input
531 of > 1mm plastic plastics to arable fields that reached up to 35 billion – 2.2 trillion (Weithmann
532 et al., 2018). In recent years, several countries have strongly encouraged farmers to use organic

533 fertilizers and successively formulated subsidy policies for composts. For instance, the Chinese
534 Ministry of Agriculture provides a subsidy for households of 1,500 RMB ha⁻¹ (equal to 215 US
535 dollars ha⁻¹) to use >3,750 kg ha⁻¹ commercial composts in some pilot areas (Ministry of
536 Agriculture and Rural Affairs of the People 's Republic of China. 2018). However, more attention
537 needs to be paid to the potential contribution of compost to macro-plastics and MPs in agricultural
538 soils.

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

543

544 3.2.3 Food waste digestate

545 Diverting food waste from landfills to anaerobic digestion can facilitate the conversion of energy
546 into usable forms and produce nutrient-rich soil improvers (Cheong et al., 2020; Xu et al., 2018).
547 However, concerns arise due to the presence of plastic packaging in many food waste streams,
548 which may inadvertently introduce macro- and micro-plastics into agricultural soils (Porterfield
549 et al., 2023). For example, the abundance of MPs was 3.0×10^5 pieces kg⁻¹ in food waste collected
550 from grocery stores in the USA (Golwala et al., 2021), and it was 4.1×10^3 particles kg⁻¹ in the
551 food compost sample in Lithuania (Sholokhova et al., 2021). In addition, some biodegradable
552 plastic packages (e.g. Polylactic acid, PLA) are widely used in food packaging and disposable
553 tableware and the usage of biodegradable plastic packages are increasing (Lu et al., 2022).
554 However, the aging and fragmentation process of PLA also could be enhanced within

555 thermophilic anaerobic digestion with kitchen waste, generating large amounts of macro-
556 plastics and MPs. Research on the occurrence and relative importance of MPs from food waste
557 is in an early stage and this potential pathway of macro-plastics and MPs to agricultural soils
558 needs further clarification (Porterfield et al., 2023).

559

560 **3.3 Other sources of plastic contamination**

561 *3.3.1 Atmospheric deposition*

562 Atmospheric transport and deposition of MPs is one of the major pathways for plastic fragments
563 entering the soil environment (Allen et al., 2019; Brahney et al., 2020). It is estimated that
564 atmospheric deposition rates of MPs range from 1.1×10^4 to 4.1×10^5 items $\text{m}^{-2} \text{yr}^{-1}$ globally
565 (Allen et al., 2019; Brahney et al., 2020; Bergmann et al., 2019). These particles typically enter
566 the atmosphere through mechanical processes, such as dust entrainment during strong wind
567 events or wave breaking of sea surface spray (Seinfeld & Pandis, 2008). Brahney et al. (2021)
568 created a model to calculate the atmospheric component of the plastic cycle, estimating the current
569 average daily total atmospheric burden (content) of MPs over the land regions of the western
570 United States to be ca. 100 tons. The largest contributor to modeled plastic deposition (84%) in
571 the western United States is road dust. In comparison, agriculturally derived plastics in dust
572 entrained into the atmosphere from agricultural fields are thought to contribute 5% to annual total
573 deposition in the same region (Brahney et al., 2021). In China, Liu et al. (2019) measured indoor
574 and outdoor dust samples collected from 39 major cities of China. The mass concentrations of
575 PET and polycarbonate (PC) MPs were determined, and the concentrations of PET and PC MPs
576 in dust were $1.6 \times 10^3 - 1.2 \times 10^5 \text{ mg kg}^{-1}$ and 4.6 mg kg^{-1} (indoors), $212 - 9,020 \text{ mg kg}^{-1}$ and

577 2.0 mg kg⁻¹ (outdoors), respectively (Liu et al., 2019). Although it is difficult for MPs > 50 µm
578 to enter the respiratory tract, these particles can enter the gastrointestinal tract where adsorbed
579 contaminants may be released, posing a potential threat to human health (Brahney et al., 2020;
580 Bergmann et al., 2019; Liu et al., 2019).

581

582 *3.3.2 Rubber tire wear*

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

593

594 *3.3.3 Water-flow and irrigation*

595 Many studies have indicated that large quantities of MPs are present in irrigation source (Chen
596 et al., 2022; Pérez-Reverón et al., 2022). Research showed that the MPs concentration in
597 irrigation water was significantly correlated with the abundance of MPs in agricultural soil, and
598 the MPs concentration of soils in direct contact with irrigation water was significantly higher

599 than that in deeper soils (Katsumi et al., 2021). A study in Spain showed that the shape, color,
600 size and type of MPs in soil samples collected from cropland were similar to those in the
601 irrigation water used on the crops (Pérez-Reverón et al., 2022). This evidence indicates that
602 irrigation water is an important source of MPs in farmland soils. Moreover, comparing MPs
603 abundance in the different source of irrigation water, the concentration of MPs in recycled
604 wastewater (159 items kg⁻¹) was around three times higher than that in the desalinated brackish
605 water (46 items kg⁻¹) (Pérez-Reverón et al., 2022). In addition, the MPs abundance in
606 agricultural soil irrigated by underground water and rainwater is significantly lower than
607 irrigated with surface water (Chen et al., 2022). Meanwhile, the soil is also a potential MPs sink
608 and MPs in soils could be transported by water off into surface water and ocean (Ren et al.,
609 2021). It is important to explore the natural and anthropogenic processes affecting the fate of
610 MPs in irrigation water.

611

612 **4. Modelling method for quantifying the source of macro-plastics and MPs**

613 ***4.1 Material Flow Analysis (MFA)***

614 MFA is a useful tool applied to better understand pathways of substance. It is an analytical method
615 to quantify flows and stocks of materials or substances in a well-defined system (Bornhöft et al.,
616 2016). Several studies have quantified the possible flows of plastic into the soil from different
617 sources by using this method (Kawecki & Nowack, 2020; Liu et al., 2020; Sieber et al., 2020).

618 MFA has been employed to analyze the flows of plastics in China (Luan et al., 2021; Liu et
619 al., 2020; Luan et al., 2022). However, few studies have focused on plastic emissions and flows
620 in an agricultural context. Of the relevant studies, Zhou et al. (2013) analyzed the emission and

621 accumulation of PVC waste in the environment, and Bai et al. (2018) estimated and predicted the
622 annual input of plastic waste from China into the ocean. Nevertheless, most of these studies have
623 used emission factors to estimate the losses of plastics directly or estimated the amounts of plastic
624 waste through municipal solid waste indirectly, rarely covering all life cycle processes and not
625 distinguishing between plastic types.

626 Luan et al. (2021) conducted a study to assess MPs and macro-plastics losses throughout the
627 plastic life cycle by using a dynamic MFA approach. The losses were analyzed based on different
628 polymers (including PE, PP, PS), PVC, acrylonitrile-butadiene-styrene (ABS), and PET, sources
629 (including personal care products, laundry process, indoor dust, fishery waste), environmental
630 media (ocean and soil) and lifecycle processes (i.e. production, use, recycling, and end-of-life
631 treatment). Based on field research and published literature, localized emission factors were
632 obtained to systematically and comprehensively estimate the plastic losses to the environment in
633 China. The results showed that MPs and macro-plastics losses entering the environment were
634 352.1 kt yr^{-1} and 12.7 Mt yr^{-1} . Of these losses, PET accounted for the highest proportion (29.1%
635 and 32.2%), and the net loss to the ocean and soil were 4.0 Mt and 173.7 Mt, respectively in 2020
636 (Luan et al., 2022).

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

643 rates, they are discussed in more details below.

644

645 *4.1.1. Generation rate.*

646 Generation rate is an essential part of MFA, which refers to the mass ratio of MPs generated from
647 macro-plastics. This is a necessary index in the calculation of the flows and stocks of materials in
648 different environments. The methods for calculating generation rates of MPs from macro-plastics
649 vary (Ren et al., 2020; Sieber et al., 2020). Plastic fragmentation may be caused by solar
650 ultraviolet (UV) irradiation, physical abrasion (abiotic), or biological attack (Cole et al., 2011;
651 Kershaw, 2015). Therefore, it is important to provide an improved estimation of MPs generation
652 in farmlands according to the generation rate of MPs.

653

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

662 Some studies have demonstrated the impact of polymer type on MP generation rate,
663 especially following UV exposure. Song et al. (2017) calculated MPs fragmentation rates via the
664 combined effects of MA (using a mechanical roller) and UV exposure for LDPE, PP, and

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

675

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

687 Qin et al., 2021).

688

689 *4.2 The other quantitation methods of macro-plastics and MPs sources*

690 Several studies have listed the possible flows of plastic into soils based on an analysis of plastic
691 use in agriculture (Brandes et al., 2021; Ren et al., 2021; Wang et al., 2021). The empirical
692 formulas for estimating MPS generation from agricultural plastics are summarized in Table 1.
693 Brandes et al. (2021) used data-driven models alongside data on MPs composition from the
694 literature in combination with national statistics on sewage sludge, compost, and plastic waste
695 production, as well as specialty cropping areas, to estimate the spatial distributions of cumulative
696 MPs mass inputs into agricultural soils in Germany. Based on the Nomenclature of territorial units
697 for statistics (NUTS3) scale, the results showed that MPs input range for soils was 0 – 15.7 kg
698 ha⁻¹, 0 – 3.79 kg ha⁻¹, and 0 – 5.18 kg ha⁻¹ from sludge, compost, and plasticulture used in
699 agriculture, respectively. Of these, the contribution of MPs followed the series sewage sludge >
700 compost > plasticulture in Germany (Brandes et al., 2021). Ren et al. (2021) found PMFs
701 contributed 10 – 30% of the total MPs from all sources in farmland soils according to a Monte
702 Carlo simulation. In addition, Wang et al. (2021) developed a novelty model to calculate the
703 distribution of MPs in the soil environment based on the aging process of different types of MPs
704 (fibers, films, fragments and granules). Based on this model, a distinct downsizing phenomenon
705 from fibers, films, and fragments to granules was observed, and human interference accelerated
706 the fragmentation of MPs. However, the quantitative contribution of different plastic sources to
707 Chinese farmland soil MPs, specifically the rate and kinetics of MPs formation, is still in the
708 primary stage.

709

710 ***4.3 Uncertainty statistics***

711 Quantifying MPs in soil mainly relies on the use of multiple statistical sources of data, which may
712 lead to uncertainty of model outputs. Other sources of uncertainty include soil analytical data, as
713 different studies have used different MPs extraction and detection methods (Bläsing & Amelung,
714 2018; Li et al., 2020). Accordingly, systematic uncertainty analyses are necessary to confirm the
715 reliability of the method. A unified approach developed by Laner et al. (2016) could be used for
716 characterizing the uncertainty of data based on this method, where the quantitative calculation of
717 the data uncertainties used coefficients of variation (CV) (Laner et al., 2016).

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

724

725 **5. Gaps in knowledge and priorities for future research**

726 Taking China as an example, this research reviewed the source, location, and occurrence of
727 macro-plastics and MPs pollution in soils. It needs to be noted that a limitation of this research
728 is the uncertainty introduced by the plastic data from various studies. In these studies, different
729 MPs extraction and detection methods are employed, which makes the data less comparable.
730 Therefore, we recommend that future studies could adopt a standardized extraction and

731 detection protocols to reduce this uncertainty. This research also concludes that there are many
732 ways that plastic fragments can enter the soil, including plasticulture practices, irrigation pipes
733 and associated infrastructure, organic wastes and wastewater residues, atmospheric deposition,
734 rubber tire wear and irrigation water. Of these, agricultural plastic mulch films are the most
735 important source of macro-plastics and MPs in Chinese farmland soils, while wastewater-derived
736 sludge may be more important in European countries. Furthermore, a MFA modeling approach
737 can be used to quantify the flows and stocks of macro-plastics and MPs. In terrestrial systems,
738 research on MPs is growing rapidly. However, more real-world studies are needed to narrow the
739 knowledge gaps in the following aspects:

- 740 1. Tracking the transport and fate of MPs from different sources in soil. At present, most of the
741 quantitative studies are estimated by models, with few actual field measurements. The use of
742 isotopically labeled plastics would be useful to track their fate, stocks, and their
743 biodegradation rates. In addition, current research is also restricted by the limited availability
744 of published data. Consequently, more samples to be required from soil, irrigation water,
745 compost and sludge, and other potential sources in the typical regions using plastic films.
746 This will help better parameterize models and reflect regional conditions.
- 747 2. There is still a lack of information on the impacts of MPs on soil health, especially regarding
748 non-point sources. Furthermore, to compare soil MPs concentrations and forms of MPs,
749 sampling, extraction, and detection methods should be standardized. In addition, most soil
750 MPs contents are expressed as the number of MPs per unit weight of soil (e.g. items kg^{-1}),
751 rather than the mass of MPs per unit weight of soil (e.g. mg kg^{-1}). Future research should
752 consider the use of mass units more often, to allow a more accurate description of the mass

753 transfer of MPs through the environment and organisms (i.e. humans, livestock and soil
754 fauna).

755 3. Establishing spatially explicit models to predict the transfer and fate of MPs entering soils,
756 linking specific source inputs to movement within the landscape and soil profile. These
757 models need to account for different soils, climates, and management practices and describe
758 MPs migration, generation rates, and the ultimate fate. Such models could then be used to
759 underpin practical guidance to farmers and regulators to promote more sustainable use of
760 plastics, and their alternatives, in agriculture.

761 4. Strengthening research on the influence of residual plastic film or MPs on crop growth, yield
762 and crop quality. The mechanistic basis of how residual plastic film or MPs affect nutrient
763 cycling in the soil also needs to be explored, to determine the optimal use of plastic mulch
764 films for different crops. Further efforts are also needed to improve the efficacy of the
765 retrieval of used plastic mulch films after crop harvest.

766 5. Strengthen the research on the impact of agricultural plastic use and recycle policies. The
767 formulation of policies can greatly improve the standard of farmers' use of agricultural
768 plastics. In China, for example, the governments have introduced various regulations and
769 incentive mechanisms following research on the hazards of plastic film residues in recent
770 years. In February 2022, the National Development and Reform Commission and the
771 Ministry of Ecology and Environment issued the "*Key Points for the Treatment of Plastic*
772 *Pollution*", which called for standardize use and recycling of plastic film, focusing on the
773 promotion and application of biodegradable plastic film and thickened high-strength plastic
774 film in key film areas. Through improvements to the recycling network system and

775 effectively controlling agricultural film pollution in farmland, the recovery rate of
776 agricultural film should be stabilized at more than 80%. In addition, the “*Opinions on Further*
777 *Strengthening the Treatment of Plastic Pollution*” issued by the Central Office of the Reform
778 and Reform prohibited the production, sale and use of several specific plastic products (e.g.
779 plastic shopping bags with a thickness of less than 25 μm , PE agricultural mulch film with a
780 thickness of less than 10 μm) in 2019, and the “*Notice on Solidly Promoting the Treatment*
781 *of Plastic Pollution*” issued in 2020 by the National Development and Reform Commission,
782 have clearly required the strengthening of the plastic prohibition and restriction policies in
783 the fields of agriculture, retail and catering. These show that China is making efforts to reduce
784 the environmental pollution caused by plastic waste from the aspect of policy control.

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

797

798 **6. Conclusions**

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

810

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819

820 **Disclosure statement**

821 The authors report there are no competing interests to declare.

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825 **References**

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