

# Effect of Dry Soil Aggregate Size on Microplastic Distribution and Its Implications for Microplastic Emissions Induced by Wind Erosion

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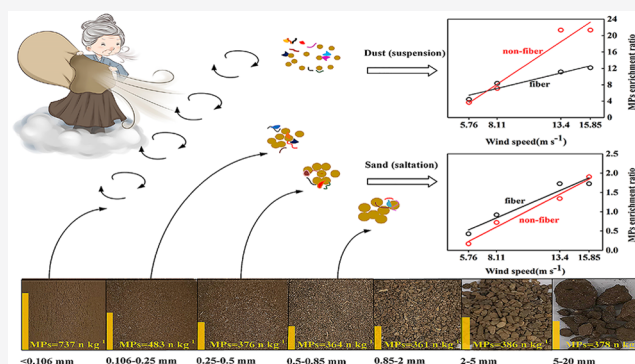
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Supporting Information

**ABSTRACT:** Microplastics (MPs) have become a problematic pollutant in different environments. Dry soil aggregates may have a remarkable influence on the emissions of MPs from surface soil due to wind erosion. Here, we sampled surface soils and monitored wind erosion events to investigate the number of MPs distributed in different dry soil aggregate sizes and the implications for MP emissions induced by wind erosion. Of the MPs in soils, 35% ( $453.49 \pm 187.62 \text{ kg}^{-1}$ ) were associated with soil aggregates and 65% ( $848.69 \pm 412.04 \text{ kg}^{-1}$ ) were dispersed. Only 38% of all fiber and 27% of all nonfiber MPs were associated with soil aggregates. The abundances of <2.5 mm fibers and <0.5 mm nonfibers decreased exponentially with an increase in aggregate size. With an increase in the abundance of microfibers associated with soil aggregates, the total organic matter and nitrogen contents increased while the mean soil particle size decreased. The MP size distributions for different soil aggregate size fractions showed sigmoid trends similar to those described by logistic models. The aggregate stability and wind speed were inversely and positively correlated with microfiber enrichment, respectively, in wind-blown sand and dust. This study provides the first insights into the number distribution of MPs in different dry soil aggregate fractions.

**KEYWORDS:** microplastic, dry soil aggregate size distribution, wind erosion, atmosphere, enrichment



## INTRODUCTION

Mineral fertilizers, pesticides, and plastic films, such as those used as mulching films or greenhouse covers, are typically considered to be the primary contributors for improving land productivity. Conversely, the excessive use of these land management practices can result in severe soil pollution that poses further threats to environmental safety and human health. Approximately 41% of global land is covered by arid and semiarid regions.<sup>1</sup> To support a growing population, plastic film mulch and greenhouses have been utilized for decades in these vast arid and semiarid areas. Nonbiodegradable plastic residues have been observed in the surface soils of farmlands in different management systems and different climatic regions.<sup>2,3</sup> Through long-term physical, chemical, and biological weathering in the environment, plastic debris can gradually degrade into small plastic fragments, of which <5 mm pieces are defined as microplastics (MPs).<sup>4</sup>

Abundant MPs have accumulated in soils. These MPs can effectively aggregate with soil particles,<sup>5,6</sup> thus affecting basic soil physicochemical properties (such as soil aggregate size, bulk density, water-holding capacity, and organic matter), which in turn may affect water and wind erosion processes.<sup>7–9</sup> Dry soils with aggregates of more erodible sizes (<0.85 mm) are more susceptible to wind erosion and therefore have higher

potential for the translocation of MPs present in the soil.<sup>10–14</sup> Studies have further suggested that wind erosion-induced MP emissions may be an important airborne source of MPs.<sup>15,16</sup>

Recently, wind tunnel experiments were conducted to explore the characteristics of MPs in wind-blown sediments in different environments.<sup>17,18</sup> Field studies have been performed to investigate how MPs are enriched in wind-blown sand and dust.<sup>15,16,19</sup> The possible dispersive routes of wind-driven atmospheric MPs were also evaluated using a forward (backward) trajectory model (the HYSPLIT model).<sup>20,21</sup> Fundamentally, MP translocation by wind erosion is affected by processes such as advection, dispersion, diffusion, degradation, settling, adsorption, and aggregation.<sup>15,18,22</sup> These studies have suggested that developing a framework for modeling MP transport by wind erosion at different locations based on local conditions is critical.<sup>22</sup>

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In aerodynamics, wind erosivity (wind speed and turbulence) and soil erodibility (dry soil aggregate size or soil aggregate stability) can be summarized as the basic factors that influence wind erosion-induced MP transport in different environments.<sup>15,18,23</sup> The erodible-sized aggregates and aggregate stability are important parameters that are required to characterize soil erodibility and further model MP emissions due to wind erosion.<sup>15</sup> The formation of aggregates is influenced by the MP shape, including MP fibers, films, or fragments.<sup>9,24,25</sup> Some studies further revealed that various concentrations and abundances of MPs were observed for different wet soil aggregate size fractions.<sup>26,27</sup> However, no attempts have been made to determine the MP number distribution in different size fractions of dry soil aggregates or its implications for MP emissions due to wind erosion. This study used in situ soil sampling and wind erosion observations to explore the distribution of MPs in different aggregate size fractions and the potential impact on MP emissions on the atmosphere.

## MATERIALS AND METHODS

**Field Work.** In Kangbao county (41°25′–42°08′ N, 114°11′–114°56′ E) of Hebei province, China, during the spring of 2020 and 2021, surface soil samples were collected and wind erosion events were monitored. In May 2020, 23 surface soil samples were collected from farmlands with plastic film mulch (Figure S1). Approximately 2 kg of soil for each sample was placed into a glass container and transported to the laboratory for analysis. In the spring of 2020 and 2021, four significant wind erosion events were observed (Table S2). Wind-blown sand (saltation) and dust (suspension) samples were collected only during the four observed wind erosion events using new flat opening collectors (NFOCs) composed of stainless steel<sup>28</sup> and glass dust-collecting cylinders (Figure S1).<sup>10</sup> All of the sand and dust samples for each erosion event were analyzed separately.

**Soil Analysis.** The 23 air-dried farmland soil samples were subjected to dry sieving and determination of the basic physical and chemical soil properties. Approximately 1000 g of bulk soil for each soil sample was separated into different aggregate size fractions through stacked sieving (<0.106, 0.106–0.25, 0.25–0.5, 0.5–0.85, 0.85–2, 2–5, and 5–20 mm). These size boundaries are often used in soil wind erodibility analysis (Figure S1).<sup>29</sup> A flat sieve with a horizontal motor shaker was set to oscillate horizontally 120 times per minute with a sieving duration of 10 min.<sup>29</sup> Prior to sieving, approximately 20 g of air-dried soil was separated to determine the soil properties.

**Microplastic Extraction and Identification.** Most plastic film materials used for farmland soils are low-density plastic materials.<sup>30,31</sup> In addition, the MP polymers in northern Chinese farmlands primarily consist of low-density components such as low-density polyethylene, polypropylene, and polyester.<sup>32,33</sup> The low-density MPs were extracted from the surface soil and wind-blown sand and dust samples using the density separation method with a saturated sodium chloride (1.19 g cm<sup>-3</sup>) solution (Text S1).<sup>34</sup> MPs were collected on a filter film and transferred to glass Petri dishes. Then, the samples in the glass Petri dish were observed using a Nikon stereomicroscope (Nikon SMZ18, Nikon Corp., Tokyo, Japan) and visually identified, counted, and characterized at 40× magnification (Figure S2). The MP types were primarily divided into fibers, films, and fragments.<sup>35,36</sup> The compositions

of the MPs were identified with a micro-Fourier transform instrument ( $\mu$ -FTIR, Nicolet iN 10, Thermo) using infrared spectroscopy.<sup>37</sup> The results regarding MP abundance were presented as the number of MP particles per mass of soil (in kilograms). The content of the dispersed MPs and the concentration of MPs incorporated or associated with aggregates of different size fractions were determined using the method proposed by Zhang and Liu:

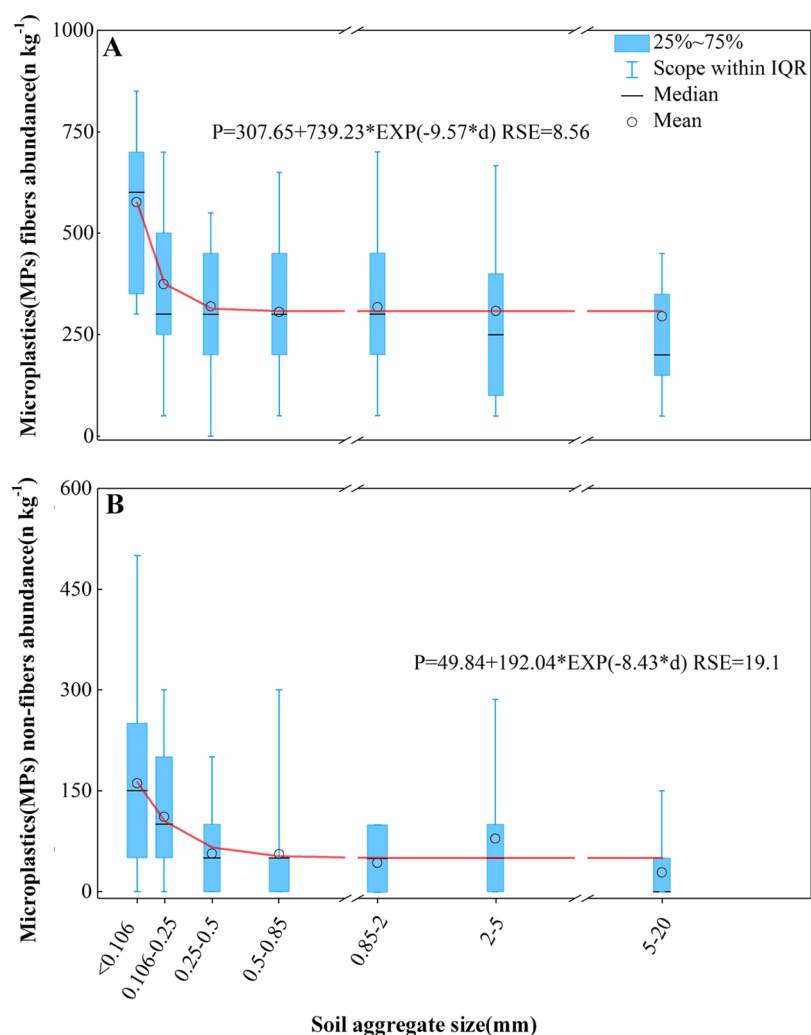
$$C_d = C_b - \sum (c_i p_i) \quad (1)$$

where  $C_d$  is the content of the dispersed plastic particles,  $C_b$  is the concentration of the total plastic particles in a bulk soil sample,  $\sum (c_i p_i)$  is the concentration of plastic particles associated with soil aggregates,  $c_i$  is the concentration of plastic particles in an aggregate fraction, and  $p_i$  is the proportion of the aggregate fractions mentioned above.<sup>26</sup>

**Quality Assurance and Quality Control.** All of the sampling treatment experiments were completed on an ultraclean platform (Figure S1). Cotton clothing and nitrile gloves were worn during experiments to avoid the introduction of MP contamination. Furthermore, five groups of blank experiments were designed to assess the atmospheric contamination during processing and analyzing. The blank experiments included three blank air samples and three samples of a saturated sodium chloride solution for each sample group. After a group of experiments, the blank experiment samples were observed (Table S3). On average,  $0.8 \pm 0.96$  fibers were observed in the five blank air groups (Table S3). This level of pollution was generally consistent with that reported by previous studies.<sup>15,38,39</sup>

## RESULTS AND DISCUSSION

**Microplastic Characteristics in the Different Aggregate Size Fractions.** The MPs were primarily identified by color as transparent, black, blue, and other (Figure S3A). Generally, fibers are preferentially transported by wind compared to fragments and films.<sup>15,18,40</sup> Fibers were the predominant shape of MPs found in most samples (Figure S4). MPs with fiber shapes were found in all different soil aggregate fractions, and the percentages of fibers for the <0.106, 0.106–0.25, 0.25–0.5, 0.5–0.85, 0.85–2, 2–5, and 5–20 mm aggregate fractions were 78%, 77%, 85%, 85%, 88%, 80%, and 91%, respectively (Figure S3B). The MPs were further categorized as fiber and nonfiber MPs on the basis of their shape, as in previous studies.<sup>41,42</sup> In total, 35% ( $453.49 \pm 187.62$  kg<sup>-1</sup>) of the sampled MPs were associated with soil aggregates, where the MPs were fully or partially inside the aggregates, and 65% ( $848.69 \pm 412.04$  kg<sup>-1</sup>) of the sampled MPs were dispersed (Figure S5). Of all of the MPs found in the soil, 28% were fiber MPs associated with soil aggregates and 46% were dispersed MP fibers (Figure S5). In contrast, only 7% of MPs were nonfiber MPs associated with aggregates and 19% were dispersed nonfibers (Figure S5). This finding further indicated that 38% of all fiber MPs were associated with aggregates, whereas only 27% of all nonfibers were associated with aggregates. Compared with soil aggregates, all types of MPs have a larger contact angle with strong hydrophobic interactions between dry plastic pieces and soil particles, which can weaken their aggregation and attachment to soil aggregates.<sup>22</sup> Interestingly, with an increase in the abundance of fibers associated with aggregates, the total organic matter and nitrogen contents increased while the mean soil particle



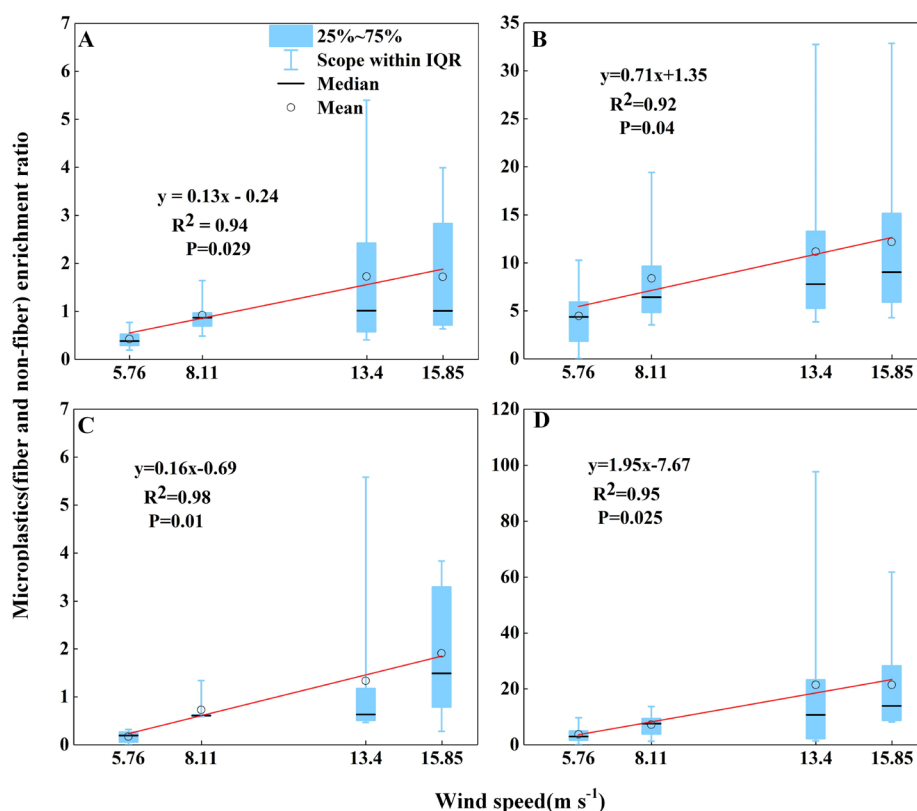
**Figure 1.** Abundances of (A) MP fibers and (B) nonfibers of the different soil aggregate size fractions. The red line represents exponential fitting according to the variation in the MP abundances in different soil aggregate sizes.

size decreased (Figure S6). The finer-textured soils with higher organic matter and nitrogen contents generally facilitated soil aggregation.<sup>43</sup> Thus, soils with greater aggregation tended to incorporate more MPs into soil aggregates.

As shown in Figure 1, the mean of MP (fiber and nonfiber) abundance approximately decreased exponentially with an increase in aggregate size. The <0.106 mm soil aggregate fraction contained the highest MP concentration of  $736.96 \pm 295.7 \text{ kg}^{-1}$ . The MPs generally adhered to, were incorporated into, or were found in the interspaces of the soil aggregates.<sup>15,18,44</sup> Tillage and cultivation can disrupt macroaggregates, decreasing the level of physical protection of MPs. Thus, the macroaggregates with incorporated MPs can be crushed into erodible microaggregates and MPs can be dispersed.<sup>45</sup> Accordingly, more dispersed MPs tended to accumulate in the erodible smaller aggregates (<0.25 mm). This further causes MP enrichment in wind-blown sand and dust during wind erosion.<sup>15</sup> More importantly, the abundance of  $\lesssim 2.5$  mm fibers and  $\lesssim 0.5$  mm nonfibers decreased exponentially with an increase in dry aggregate size, but the abundances of >2.5 mm fibers and >0.5 mm nonfibers were independent of the dry aggregate size (Figure S3C,D). The amount of MPs retained in soils was dependent on the MP size and shape. This indicated that larger nonfibers more easily

incorporated into aggregates compared with smaller fibers. The shape- and size-dependent trends of MPs in soil aggregates were also investigated for wet soil aggregates.<sup>9</sup>

The detailed size distributions of the MPs (fiber and nonfiber) are shown in Figure S7 (see more details in Figure S8). Continuous probability distributions can capture the essential continuous nature of environmental MPs.<sup>46</sup> Interestingly, the MP size distributions of bulk soil were very consistent with that of each aggregate fraction. However, the fiber size distributions were considerably different from the nonfiber size distributions (Figure S7). It was assumed that nonfibers could be more easily mechanically fragmented into smaller pieces by tillage, cultivation, and dry sieving than could fibers. The changing processes of the MP size distributions for each aggregate fraction showed similar sigmoid curves. Several fitting models for characterizing the particle size distribution, including log-normal, bimodal, logistic, and power law distributions, have been proposed.<sup>15,46–48</sup> In this study, the logistic model best depicted the MP size distribution. This indicated that the MP abundances generally increased slowly with small sizes ( $\lesssim 0.1$  mm) and then increased dramatically with median sizes (from  $\sim 0.1$  to  $\sim 2$  mm), which was different from the MP size distribution determined from other environmental media (water and air).<sup>3,48</sup> The sigmoid curve



**Figure 2.** Relationship between wind speed and (A) fiber enrichment ratios in wind-blown sand, (B) fiber enrichment ratios in wind-blown dust, (C) nonfiber enrichment ratios in wind-blown sand, and (D) nonfiber enrichment ratios in wind-blown dust. The blue boxes indicate fibers and nonfibers. The red fitting lines are derived from the mean fiber and nonfiber enrichment ratios in wind-blown sand and dust.

of the MP size distribution may suggest that the natural accretion or fragmentation processes of MPs typically include various deterministic (weathering and sieving) or stochastic (erosion and cultivation) elements.<sup>3</sup>

**Implications for MP Emissions Induced by Wind Erosion.** MPs (approximately  $0.9\text{--}2.2\text{ g cm}^{-3}$ ) are 30–60% lighter than natural soil minerals (approximately  $2.6\text{--}2.8\text{ g cm}^{-3}$ ); thus, they are more likely to be entrained by wind. The response of MPs to wind entrainment was determined by calculating the enrichment ratio of MP items in wind-blown sand (dust) to that in the corresponding surface soil.<sup>15,17,18</sup> It was initially found that the AS, a vital and sensitive indicator of a dry soil aggregate, increased when the fiber enrichment ratios decreased (Figure S9). Theoretically, the higher enrichment ratio indicated that more MPs were enriched in wind-blown sand and dust. A lower AS value generally represents a high breakdown susceptibility of the soil aggregate.<sup>49</sup> This indicates that soil aggregates containing MPs are more easily mechanically fragmented into smaller aggregates depending on the wind strength. During wind erosion, MPs change from a “bound state” (incorporated into soil aggregates) to a “free state” (not incorporated into soil aggregates) within erodible soil, with a correspondingly higher probability of entrainment by wind.<sup>15</sup> However, in this study, the nonfiber enrichment ratios in the wind-blown sand and dust were independent of the AS. This suggests that fibers generally negatively affect aggregate formation. Previous studies have also concluded that fibers and nonfibers exert different effects on the interparticle forces between MPs and soil grains.<sup>9</sup> To reveal the mechanism of MP enrichment in wind-blown sediments, more studies are

required to investigate the influence of the dry aggregate size on MP detachment by wind erosion.

Compared with soil AS, wind speed had a significant influence on the MP enrichment ratios in wind-blown sand and dust. As shown in Figure 2, the enrichment ratios of fibers and nonfibers in wind-blown sand and dust were proportional to the average wind speed. Dispersed MPs within surface bulk soils are more easily eroded, and higher wind speeds can enhance the ability of the wind to detach MPs from surface soils.<sup>15,17,18</sup> Therefore, MPs emitted from dry farmlands (MP sources) can be transported to remote downwind areas (MP sinks).<sup>12,20</sup> Accordingly, the relationship between the wind speed and the MP deposition flux has been studied.<sup>19,50</sup>

Notably, the changes in the fiber and nonfiber enrichment ratios with size in wind-blown sand were well characterized by the logistic model (Figure S10A,C). In contrast, the changing patterns of the fiber or nonfiber enrichment ratios with size in wind-blown dust varied during different storms. The fiber enrichment ratios approximately exponentially decreased with an increase in MP size (Figure S10B). In contrast, the nonfiber enrichment ratios were independent of MP size (Figure S10D). Scientists often portray environmental MPs as diverse and complex materials with multidimensionality that includes polymer composition and density, size, shape, and longevity.<sup>3,48</sup> The actual three-dimensional shape and composition of MPs are critical indicators that influence the ability of an aggregate to cement MPs; they are also key indices that affect the ability of wind to detach MPs from surface soil.<sup>9,17,18</sup> Further control experiments examining how the multidimensionality of MPs affects MP transport by wind and the

subsequent enrichment of MPs in wind-blown sediments are required.

In summary, the quantitative relationships between the MP enrichment ratios in wind-blown sand or dust and aggregate stability, wind speed, and MP size were observed. These results provided valuable information for improving our understanding of the entrainment, transport, and deposition of MPs by wind erosion. Field-scale MP emission experiments are the cornerstone for diffusion route modeling<sup>12,19</sup> and quantitative MP emission evaluations at a regional scale.<sup>16,17</sup> Therefore, it is critical to perform sophisticated experiments to examine the relationship between the primary factors (wind speed, soil aggregates, and MP characteristics) and MP enrichment in wind-blown sediments in different locations with various geographic conditions. It is also necessary to develop a unified, efficient, and stable protocol that includes wind erosion monitoring and MP extraction from soils and airborne sediments. More importantly, these measures can help in the development of a universal model of wind-driven MP transport and further facilitate collaboration between the atmospheric MP research community and environmental managers to share and improve pollution control technology.

## ■ ASSOCIATED CONTENT

### SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acs.estlett.2c00338>.

Details of sampling (S1), details of MP extraction (S2), MP particle size classification method (S3), details of data analysis (S4), sampling site characteristics (Table S1), wind speed during wind erosion events (Table S2), number of MPs in the blank samples (Table S3), soil sampling field campaign, wind erosion observations, and laboratory equipment (Figure S1), images of MPs observed under a stereomicroscope (Figure S2), abundance of MP colors, types, and sizes (Figure S3), abundances of MPs (fiber and nonfiber) in the surface soil and wind-blown sand and dust collected from wind erosion observation sites in 2020 and 2021 (Figure S4), abundances of MP fibers and nonfibers associated with or dispersed from soil aggregates in bulk soil (Figure S5), relationships between the fiber abundance associated with soil aggregates and the organic matter content, total nitrogen content, and mean soil particle size (Figure S6), size distribution of MP fibers and size distribution of MP nonfibers in bulk soil and soil aggregate size fractions (Figure S7), size distribution and percentage of MP particles in different aggregate size fractions (Figure S8), relationships between aggregate stability and the fiber enrichment ratios in wind-blown sand and dust (Figure S9), and fiber and nonfiber size enrichment ratios in wind-blown sand and dust during four wind storms (Figure S10) (PDF)

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### Notes

The authors declare no competing financial interest.

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## ABBREVIATIONS

MPs, microplastics; NFOCs, new flat open collectors; EF, erodible fraction; AS, dry soil aggregate stability;  $\mu$ -FTIR, micro-Fourier transform infrared spectroscopy; ER, enrichment ratio; HYSPLIT, hybrid single-particle Lagrangian integrated trajectory; RSE, residual standard error

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