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Letter

Preferential Emission of Microplastics from Biosolid-Applied Agricultural Soils: Field Evidence and Theoretical Framework

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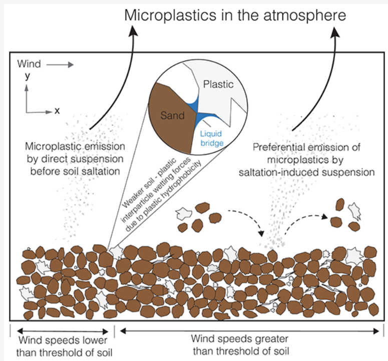

ABSTRACT: Land application of wastewater biosolids on agricultural soils is suggested as a sustainable pathway to support the circular economy; however, this practice often enriches microplastics and associated contaminants in topsoil. Wind could transport these contaminated microplastics, thereby increasing their inhalation health risks. Analyzing wind-borne sediments collected from wind tunnel experiments on biosolid-applied agricultural fields, we show enrichment of microplastics in wind-blown sediments. We explain this preferential transport and enrichment of microplastics by using a theoretical framework. This framework reveals how the combined effects of the low density of microplastics and weakened wet-bonding interparticle forces between microplastics and soil particles lower their threshold velocity, the minimum wind velocity necessary for wind erosion to occur. Our calculations indicate that microplastics could be emitted at wind speeds lower than the characteristic threshold of background soil. Analyzing the windspeed distribution for 3 months of wind events over a bare soil surface, we showed that more than 84% of the wind events exceed the threshold velocity of microplastics of size 150 μm, while only 23% of the wind events exceed the threshold velocity of the background soil. Thus, current models for fugitive dust emissions may underestimate the microplastic emission potential of biosolid-amended soils.

KEYWORDS: *microplastics, agriculture, biosolids, aeolian transport, wind erosion, emission potential*

■ INTRODUCTION

The application of wastewater biosolids on agricultural lands contributes to the circular economy by utilizing waste while reducing greenhouse gas emissions related to fertilizer use.[1,2] The annual production of biosolids is estimated to be 100 million tons globally, with a projected annual increase of 175 million tons by 2050.[3] While this practice provides several benefits,[4,5] it could potentially introduce microplastics and other persistent pollutants adsorbed on microplastics to agricultural lands.[6−8] Biosolids application in the US alone could introduce trillions[9] of microplastics on agricultural fields, resulting in microplastic concentration in the topsoil as high as 10 mg kg^{-1} after just 5 applications.[6] In these biosolids, more than 90% of microplastics are undetectable due to their small size (<10 μm) and difficulty in identification using current methods.[9] Smaller microplastics have a higher potential to carry a wide range of pollutants due to their high surface area[10−13] and therefore pose a greater inhalation risk to humans.[14,15] The inhalation risk is higher in arid and semiarid agricultural lands because of higher susceptibility to wind erosion coupled with the increase in biosolid application to improve soil quality. Wind-driven erosion is expected to accelerate in the future because of recent increases in aridity, recurrent droughts, and disturbances.[16]

While many studies have measured microplastics in agricultural soils,[6,17−19] limited field studies have examined the wind transport of microplastics from biosolid-applied agricultural lands,[20−22] and far fewer studies have examined the conditions that affect the resuspension potential of microplastics using wind tunnel experiments in agricultural fields.[23−25] Some of these studies added microplastics created in the laboratory at a concentration much higher than that expected in agricultural fields, thereby affecting interactions between soil grains and microplastic particles.[21] Additionally, microplastics created in the laboratory by abrading plastic materials could differ significantly in shape, size, and surface properties compared to microplastics present in biosolids. Thus, previous conclusions about the enrichment of microplastics in agricultural dust could be inaccurate, since they do not model environmentally relevant conditions.

The mechanisms of why microplastics may be enriched in wind-blown sediment are unclear. The physics of particle emission by wind is complex, as it involves atmospheric, soil, and land surface processes.[26] Erosion by wind occurs when the shear stress exerted by the wind on the ground surface exceeds

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the shear strength of the soil aggregates and their resistance to detachment. The minimum wind shear stress required to cause erosion, commonly known as the threshold shear velocity, depends on several factors including particle characteristics, size and stability of the soil aggregates, field surface conditions, vegetation cover, and near-surface soil moisture.[16,27] At wind speeds beyond the threshold, saltation-sized particles (70−500 μm) are entrained and carried by wind short distances away as a horizontal flux within the lowest 1 m of the atmosphere.[28] The saltating particles collide with other particles or aggregates on the surface and generate fine particles, which can also be resuspended.[29−31] Most airborne sediments loads to the atmosphere are during the few events when wind speed exceeds the characteristic threshold velocity necessary for soil saltation to occur.[28] Thus, events when the wind speed is below this threshold are rarely considered in estimating the mass transfer of particles from the land surface to the atmosphere, though they could be critical for microplastic emissions for several reasons. Compared to soil particles, microplastics could be preferentially resuspended by wind due to their low density and hydrophobic surface characteristics.[32,33] While low density reduces the downward force exerted on the particle by gravity, higher hydrophobicity can lower the interparticle wet bonding forces (adsorption and liquid bridge bonding) that typically act as a glue against the liftoff force exerted by the wind.[34] Thus, a combination of both factors could make microplastics more likely to be entrained by the wind even at lower wind velocities. The objectives of this study are to estimate the enrichment of microplastics in wind-blown sediments on agricultural soils with historic biosolid applications and to explore the cause of this enrichment using a theoretical framework.

■ MATERIALS AND METHODS

Wind-Blown Sediment Collection during a Wind-Tunnel Experiment. Soil and windborne sediment samples were collected from plots with historic application of biosolids (∼6 tons ha^{-1}) in Lind, Washington (47°00′ N, 118°34′ W) during a wind-tunnel experiment as described in a previous study (Figure S1).[20] Briefly, a portable wind tunnel[35] was set up over two identical biosolid-applied and traditionally tilled (disk tilled method) plots within the same agricultural field, and a wind speed of 16 m s^{-1} was sustained over 10 min to collect around 5−10 g of wind-blown sediments using a vertically integrating isokinetic slot sampler.[35] These wind-blown sediment samples represent active saltation conditions that normally occur in the field during high wind events.[20,36] Applied biosolid samples (n = 4) were taken from the supply stockpile as a reference for expected contamination levels as they are a source of microplastics in soil. To estimate the enrichment of microplastics in suspended sediments by wind, soil (n = 12) and wind-blown sediment samples (n = 8) were collected and analyzed for microplastics.

Microplastic Extraction and Analysis. Microplastics were isolated from all samples using density separation to remove denser soil particles followed by the digestion of natural organic matter, as described in detail in the Supporting Information (Figure S2). Briefly, 1 g of a dry solid sample was mixed thoroughly with 40 mL of a 10 M KI solution (density 1.7 g cm^{-3}) so that microplastics and organic debris with a density lower than 1.7 g cm^{-3} would float. This mixture was centrifuged, vacuum filtered, digested using Fenton's reagent (2:1 ratio of 30% H_2O_2 and iron sulfate),[37,38] and acidified. A second round of density stratification and vacuum filtration followed. Microplastics isolated after the digestion of organics were counted manually using a compound microscope. Additionally, a subset of wind-blown sediment samples was characterized by using Fourier-transform infrared spectroscopy (FTIR, Thermo Scientific NicoletTM iN10) in reflectance mode, using a minimum 60% match criteria across all available commercial libraries to identify the microplastics' compositions and sizes (described in detail in the Supporting Information).

Plastic cross-contamination was eliminated during sampling processing steps by using nonplastic tools and containers wherever possible. All glassware, containers, and filtration devices were rinsed with deionized water three times before use. A Tukey one-way test ($p < 0.05$ indicates significance) was used to compare microplastic concentrations among three sample types: wind-blown sediments, soils, and biosolids.

Theoretical Framework for Entrainment of Microplastics. Under the influence of wind, a soil grain at the surface experiences several forces: aerodynamic drag, aerodynamic lift, and stabilizing forces like gravity and interparticle cohesive forces.[39] The resulting force balance is often used to derive an expression of threshold shear velocity (u_{*t}).[26] Wind speed controls the erosive action of wind, while field surface conditions, soil texture, size and shape of the aggregates, as well as near surface soil water content affect the threshold shear velocity. Several theoretical and empirical equations have been suggested in the past to express the wind threshold shear velocity at which saltation is initiated as a function of these factors.[39] Here, we adopt a semiempirical expression (eq 1) developed by Shao and Lu[26]

$$u_{*t} = A_N \sqrt{\frac{\rho_p - \rho_a}{\rho_a} g D_p + \frac{\gamma}{\rho_a D_p}} \tag{1}$$

where the ρ_a is the air density, ρ_p is the microplastic density, D_p is the microplastic diameter, g is the acceleration due to gravity, $A_N = 0.111$ is a dimensionless parameter,[27] and $\gamma = 2.9 \times 10^{-4}$ N m^{-1} is a parameter that scales the strength of the interparticle forces[40] (Table S1). The interparticle forces in soil include electrostatic forces, van der Waals forces, and capillary forces due to the presence of a liquid bridge between the particle and soil surface.[41−43] Among all forces, the capillary force exerted by the liquid bridge can be orders of magnitude greater than electrostatic and van der Waal forces. This cohesive force exerted by the liquid bridge depends on the contact angle, which in turn depends on the hydrophilic or hydrophobic nature of both surfaces.[44] Thus, the cohesive force from the liquid bridge can be calculated using eq 2[45,46]

$$F_i = 2\pi b T \sin(\theta + \gamma) + \pi b^2 T\left(\frac{1}{c} - \frac{1}{b}\right) \tag{2}$$

where $T = 0.07275$ J m^{-2} is the surface tension of water, γ is the contact angle, $\theta = 40°$ depends both on moisture content (12.5%, expressed as percentage of pore space) and on soil packing[45] (open packing of spherical particles), b is the radius of the fluid neck connecting two spherical grains, c is the radius of the meridian curve, and $(1/c - 1/b)$ represents the total curvature of the surface at this point between the pore air and the water in the liquid bridge. Further methodological details and values for calculated constants (b, c) are summarized in the Supporting Information (Table S2).

Calculation of the Exceedance of the Threshold Wind Velocity of Microplastics in an Agricultural Field. To

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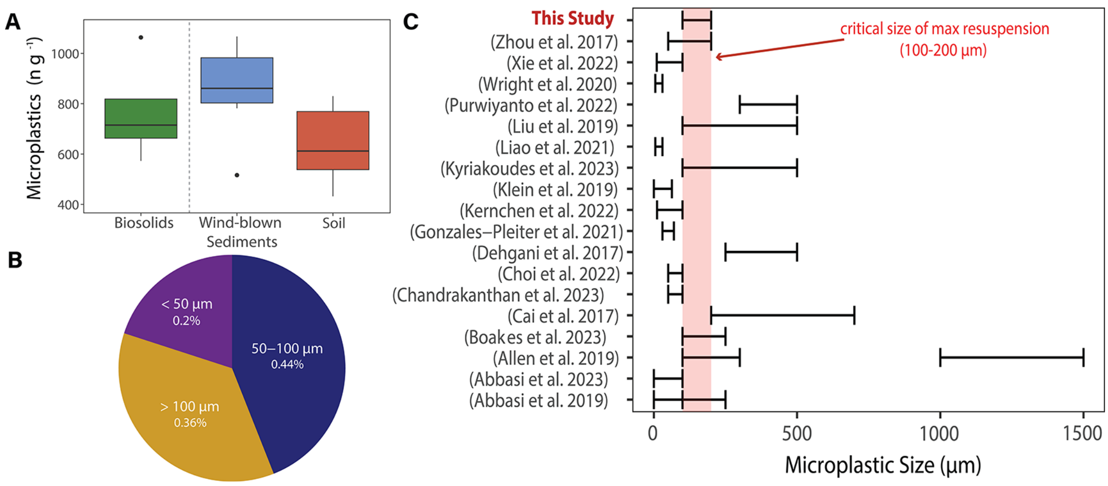

Figure 1. (A) Microplastic concentrations (pieces, n, per gram of solid) for each type of sample processed: biosolids, wind-blown sediments, and soil. Microplastic concentrations in the wind-blown sediments were significantly higher ($p < 0.05$) than those in the soil from where they are emitted. (B) Size distribution of microplastics from wind-blown sediment samples based on the longest side of 35 identified microplastic pieces. (C) Dominant size ranges from 18 global studies[53−70] analyzing microplastics in wind-blown sediments, where the highlighted region is the theoretical estimate of the critical size for maximum resuspension.

assess the frequency of microplastic emissions at typical wind speeds in an arid agricultural field in the US, we plotted a Weibull distribution of shear wind velocity records during a three-month windy season taken from Burger et al.[47] Briefly, wind speed records were collected between March 11, 2016, and June 30, 2016, from a bare soil site at the Sevilleta National Wildlife Refuge (N 34°23.961′ and W 106°55.710′). A 4-m high solar-powered meteorological tower recorded average and maximum wind velocities at 4, 2, 1, and 0.5 m above ground every second and averaged the reading over 1 min intervals.[47] The average wind velocities at all 4 heights were used to fit the wind profile to the Prandtl−von Karman logarithmic law[47] (eq 3)

$$u(z) = \frac{u_*}{0.4} \ln\left(\frac{z - d}{z_0}\right) \tag{3}$$

where $u(z)$ is wind velocity at height z, and u_* is the shear velocity (or friction velocity), d is the zero-plane displacement, z_0 is the aerodynamic roughness length, and 0.4 is the Von Karman constant.[48] The displacement height was assumed to be ∼0 for the surface with no vegetation. The slope and y-intercept of the best-fit line relating $\ln(z)$ with $u(z)$ in Figure S3 were used to calculate $z_0 = 0.049$ and u_*.[48]

This fitted wind profile was used to convert recorded maximum wind velocities at 4 m, $u(z = 4)$, into maximum wind speed at 2 m, $u(z = 2)$, for each minute. Weibull wind distribution was generated to calculate the probability of windspeed exceeding any specific value. Using the theoretical threshold shear velocity (eq 1) for microplastics and experimentally measured threshold shear velocity for the background soil at this site,[49] this distribution of wind speeds was used to estimate the number of 1 min wind events that exceed the threshold velocity of typical microplastics (PP). The threshold shear velocity for soil particles[49] and microplastics (eq 1) was converted to corresponding wind speeds at 2 m (eq 2) (Table S3), and the probability of windspeeds exceeding these critical wind speeds was calculated from the generated wind velocity distribution. Here, we represented microplastics as PP, the lowest density commonly used plastic with a size D_p, 150 μm, which is the critical size range for resuspension estimated in our study.

RESULTS AND DISCUSSION

Enrichment of Microplastics in Wind-Blown Sediments. The wind-blown sediments collected from the wind tunnel experiments contained more microplastics than either the source soils or the biosolids that released microplastics into the soil (Figure 1A). The mean microplastic concentrations in soil, biosolids, and wind-blown sediments were 635 n g^{-1}, 766 n g^{-1}, and 859 n g^{-1}, respectively. We measured microplastic concentrations in wind-blown sediments to be 78−12560 n kg^{-1}, which is comparable to concentration ranges reported in previous studies.[50,51] Furthermore, the result indicates microplastics were enriched in the wind-blown sediments by a factor of 1.35 when compared to traditional soil minerals, such as quartz, aluminosilicate clays, and metal oxides.

Analyzing the size distribution of microplastics, we estimated the dominant microplastic size in the wind-blown sediments was > 100 μm (Figure 1B). This is consistent with the critical size of the resuspension determined theoretically using eq 1 (Figure 2A). This critical size is where microplastics require a minimum fluid threshold and thus are most likely to be resuspended. In our study, FTIR was used to measure characteristic properties of microplastics (Figure S4), but it cannot positively confirm (>60%) the composition of smaller particles due to a detection limit of 20 μm.[52] Thus, smaller microplastic fractions could be underestimated in this study. The dominant size range of microplastics in dust- and wind-blown sediment samples varied in comparable studies (Figure 1C). However, the size reported in these studies could be affected by the microplastic quantification protocol such as minimum detection limit or resolution of microscopy techniques and collection methodology. Much of the data in previous studies were not from controlled wind tunnel experiments. Thus, future wind tunnel and field experiments should measure particle size distribution of emitted microplastics to confirm the preferentially resuspended particle size.

Enhanced Suspension of Microplastic below Threshold Wind Velocity of Background Soil. Using a semi-

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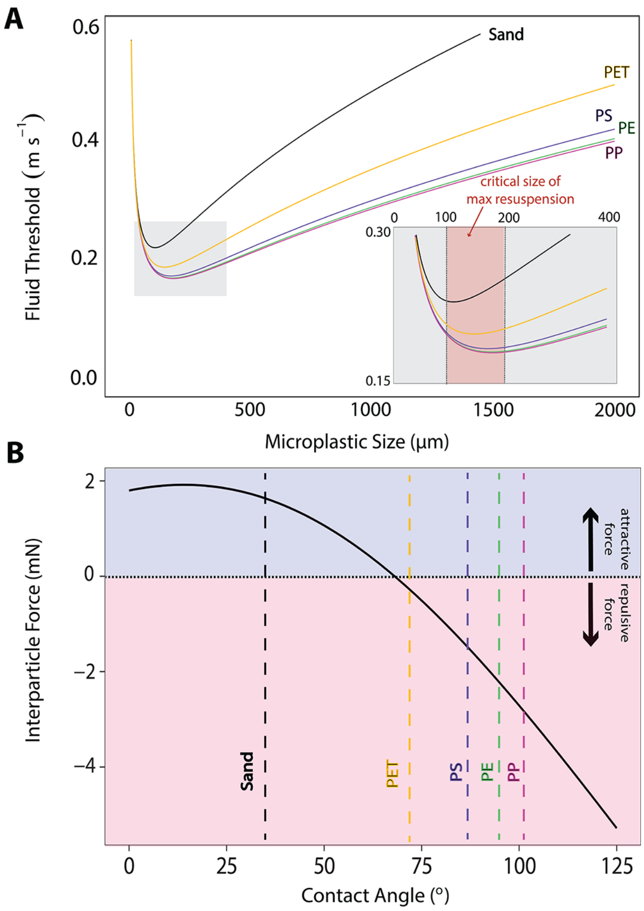

Figure 2. (A) Fluid threshold for initiating saltation (resuspension) for four types of microplastics with different sizes (0−2000 μm) and densities (0.91−1.40 g cm^{-1}). Within the region of interest in the inset map between 100 and 200 μm, microplastics require a minimum fluid threshold as visualized by the curves' minimums. (B) The interactive force from the water film on particles is calculated based on particle size and contact angle for an open packing system with 12.5% moisture content. *Abbreviations are defined as follows: PE: polyethylene; PET: polyethylene terephthalate; PS: polystyrene; PP: polypropylene.

empirical expression for the threshold shear velocity (eq 1), we show that microplastics could be eroded at a lower fluid threshold than quartz sand, the most common mineral found in soil[48] (Figure 2A). The threshold shear velocity depends on particle size: Increasing particle size until the critical size range (∼100−200 μm) decreases the fluid threshold due to a decrease in interparticle force and negligible impact of gravity. Any further increase in particle size beyond the critical size range increases the threshold shear velocity due to the increasing effect of density and the resulting dominance of gravity. Thus, the effect of particle density becomes more apparent when the particles are bigger than 75 μm.[26] For the resuspension of small-sized microplastics (<10 μm)—the relevant size for inhalation risk—interparticle forces could play a greater role than the density of plastic particles. For these small microplastics, a larger wind velocity is required to overcome the interparticle forces, which become increasingly important as the particle size becomes smaller.

The surface adsorption of moisture is generally limited on microplastics, which are generally hydrophobic.[46] Thus, the microplastic particles might be expected to retain less adsorbed water and to be consequently lighter with a lower threshold velocity. When soil moisture (or humidity) increases in pore spaces, condensation starts to occur in the contact points between the particles, adsorbing onto the grain surface and thereby forming a "liquid bridge", which increases the interparticle forces between particles. The hydrophobic surfaces of microplastics can delay the formation of liquid bridges or even prevent their formation in cases of extremely hydrophobic microplastics. Consequently, the interparticle forces associated with the liquid bridges between microplastics and soil particles[71] are expected to be less adhesive than the bridges between soil particles due to increasing degrees of hydrophobicity (i.e., increasing contact angle[72]) (Figure 2B). The high hydrophobicity of plastic surfaces increases their contact angle, which results in a net repulsive interparticle force (Figure 2B). In contrast, a smaller contact angle on quartz particles allows the liquid bridge to act as a glue for quartz particles, resulting in a net attractive force. Thus, hydrophobic microplastics have weaker interparticle forces, or more repulsive force, than that experienced by quartz or other soil minerals with lower hydrophobicity. We conclude that a reduction in both adsorbed and liquid bridge bonding significantly reduces the interparticle forces associated with moisture bonding and makes microplastics more susceptible to erosion by wind than soil particles of the same size.[46]

■ ENVIRONMENTAL IMPLICATIONS

Our analysis indicates that microplastics are more likely to be transported by wind below the characteristic threshold velocity of background soil due to their lower density and weaker liquid bridge bonding potential compared to typical soil particles. To demonstrate the environmental implications of this current finding, we analyzed a distribution for wind events in a typical arid region, where biosolids are typically applied to increase soil fertility. Our analysis shows that microplastic's lower threshold velocity would result in approximately 3-fold increase in the number of 1 min wind events that would resuspend microplastics (Figure 3A). Based on a semiempirical model to account for density effects, we estimated the threshold friction or shear velocity for microplastic (PP) emission is around 0.176 m s^{-1}, resulting in a threshold wind velocity of 1.64 m s^{-1} at 2 m above the ground surface. This is much lower than an experimentally determined wind speed velocity (5.84 m s^{-1}) required to move the soil particles at this site.[49] The current models for fugitive dust emissions assume the soil surface to be stable with negligible dust emissions in the absence of wind speeds exceeding the saltation threshold of the soil or surface disturbances.[73] With that assumption, current emission models may underestimate microplastic emission potential from agricultural soils—missing around 61% of wind events, which are too low to move soil sediments but sufficient to initiate microplastic movement and drive their emission (Figure 3B).[74] The threshold wind velocity for microplastics can become even lower if both low density and low interparticle forces are accounted for. Microplastic particles are readily emitted from soil surfaces under low wind speed scenarios, which are more frequent. The theoretical framework proposed in this study can be used to assess microplastic emissions more accurately by accounting for microplastic fluxes that are typically underrepresented. Thus, these emission events should be incorporated into fugitive dust emission models that inform environmental and human health risk assessments.

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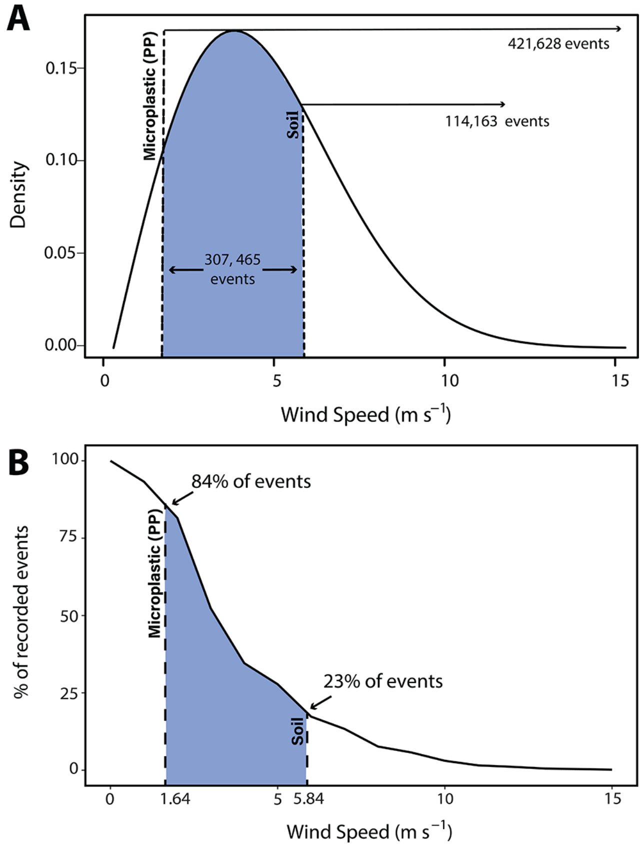

Figure 3. (A) Distribution of wind speeds (at 2 m) for the windy season showing the threshold wind velocity range for microplastics (PP, 150 μm) and background soil at this site. (B) Percentage of wind events for each wind speed between 0 and 15 m s^{-1} for three months of wind data.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.estlett.3c00850.

Experimental setup (Figure S1), microplastic isolation methodology (Figure S2), assumed constant parameters in eq 1 (Table S1) and eq 2 (Table S2), best fit of eq 3 (Figure S3), and FTIR analysis of microplastics (Figure S4) (PDF)

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Notes

Any opinions, findings, conclusions, or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of the US National Science Foundation.

The authors declare no competing financial interest.

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