PLASTIC POLLUTION

Plastic rain in protected areas of the United States

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Eleven billion metric tons of plastic are projected to accumulate in the environment by 2025. Because plastics are persistent, they fragment into pieces that are susceptible to wind entrainment. Using high-resolution spatial and temporal data, we tested whether plastics deposited in wet versus dry conditions have distinct atmospheric life histories. Further, we report on the rates and sources of deposition to remote U.S. conservation areas. We show that urban centers and resuspension from soils or water are principal sources for wet-deposited plastics. By contrast, plastics deposited under dry conditions were smaller in size, and the rates of deposition were related to indices that suggest longer-range or global transport. Deposition rates averaged 132 plastics per square meter per day, which amounts to >1000 metric tons of plastic deposition to western U.S. protected lands annually.

The world produced 348 million metric tons of plastic in 2017, and this number grows every year by ~5% (*1*, *2*). A large proportion of this production accumulates as waste in the environment, and progressive fragmentation leads to the presence of secondary plastics in terrestrial, freshwater, atmospheric, and marine environments (*2*). Extremely high resilience and longevity give plastics their utility, but these same characteristics lead to the unrestrained accumulation of synthetic materials in nearly every ecosystem on the planet (*3*). Though atmospheric microfibers have recently been documented in Europe and the Arctic (*4*, *5*), the route of primary or secondary microplastics (microfibers and particles) to the atmosphere has not been clear. Primary microplastics are defined as plastics that were manufactured in the size range observed (e.g., microbeads), whereas secondary plastics are derived from the fragmentation of larger pieces of plastics through physical abrasion and/or weakening after exposure to ultraviolet light. To determine potential sources of atmospheric microplastics and the rate of their accumulation in conservation areas of the United States, we quantified the fallout of primary and secondary microplastics to 11 remote and protected areas in both wet atmospheric deposition, collected at week-long intervals while precipitation occurred (n = 236 samples), and dry atmospheric deposition, collected at monthly or bimonthly intervals (n = 103). We used relationships between plastic deposition rates and the intersections of air-mass back trajectories with population centers, contemporaneous dust (soil) deposition, global indices of climate, and plastic composition to identify both emission and product commodity sources. Understanding the sources of microplastics to the atmosphere—both in terms of emission points and product commodities—will, in turn, allow us to implement scale-relevant solutions to mitigate plastic pollution.

Microplastics were present in 98% of all of the wet and dry samples analyzed from U.S. protected areas. Observed microplastic particle sizes were between 4 and 188 μm and fiber sizes were between 20 μm and ~3 mm, with average widths and depths of 18 and 6 μm, respectively (fig. S2). Approximately 70% of the particles were within the size range for long-range and even global transport of dust (<25 μm) (*6*, *7*), whereas most fiber lengths suggested regional transport (10 to 1000 km) (*8*). Because plastic density (0.65 to 1.8 g cm^{-3}) is lower than that of soil particles (~2.65 g cm^{-3}) (*9*), microplastics are more transportable. Fibers, in particular, have greater surface area–to–volume ratios, which increase drag forces and reduce settling velocity. This process may be similar to ballooning in spiders, where a combination of electrostatic forces and drag allows spiders attached to silk fibers to travel thousands of kilometers (*10*).

Daily 48-hour atmospheric back-trajectory analyses were determined using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (*11*, *12*) and were compared with weekly wet plastic deposition rates through 2018. Our analyses suggest that wet-deposited microplastics originate from different source regions than those that are dry deposited. Wet plastic deposition rates at half of the sites were significantly correlated to population metrics, as determined by the intersection of the air mass with population centers (Table 1). Distance traveled, mean wind speeds, and contemporaneous dust deposition also described significant portions of the variance noted at individual sites. We observed that microplastics deposited in wet conditions are larger in size and lower in number (fig. S2) and are correlated to both dust deposition and population metrics. This observation reflects the role of regional storms in the entrainment and subsequent rainout of microplastics, as these storms often pass through urban centers or over erodible soils. In contrast, dry deposition shows a negative relationship with regional dust deposition rates and is related instead to indices that represent broad-scale atmospheric patterns, specifically a more southerly jet stream. This suggests that dry-deposited plastics are subject to large-scale, global dispersion (Table 2).

Microfibers made up most of the synthetic material found in both wet (66%) and dry (70%) deposition. Fiber compositions were mainly consistent with those of textiles used for clothing, including cotton, polyester, and nylon. We also observed fibers composed of polyolefin, which is more commonly used for household and vehicle carpeting, as well as polytetrafluoroethylene and polyethylene fibers, which are used in a variety of industrial applications (*13*). Industrial coatings on fibers, such as Valbond 6053, were also identified, which underscores the diversity in microfiber sources to U.S. protected areas. It is worth noting that polypropylene and polytetrafluoroethylene are also commonly used in outdoor gear, including fleece, tents, waterproof clothing, and climbing ropes (*13*). Because microfibers are known to shed from clothing during normal wear (*14*), emissions from park users may contribute to the observed deposition rates, particularly in national parks with high visitation rates. Clothing fibers are also directly released to the atmosphere during laundry drying at rates that are several times the rates at which fibers are released to wastewater during the washing phase (*15*, *16*), and these fibers are then transported to protected areas during times of favorable wind speeds and trajectories (fig. S5).

The polymer compositions of individual plastic particles smaller than 20 μm were more difficult to identify using Fourier transform infrared (FTIR) spectroscopy because of the diffraction limitation of midinfrared light. However, in subsamples, almost all brightly colored particles that fell within our counting criteria were identified as synthetic using FTIR spectroscopy particle mapping in reflection mode, which allows the mass identification of particles in the subsamples. Using this reflectance mapping technique on 32 subsamples, we found that 2.5 to 5% (on average, 4%) of the identifiable dust particles were synthetic polymers. This included particles and fibers that did not meet our visual counting criteria (because they were clear or white), which suggests that our estimates of plastic deposition rates based on counts are conservative (Table 3). Most plastic particle compositions found in our samples can be linked back to

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industrial applications and coatings. Polyethylene, polypropylene, polyvinyl acetate, and ethylene-acrylic copolymer were also identified. Approximately 30% of the particles were primary plastic microbeads ranging in size from 5 to 30 μm in a wide variety of colors (fig. S1). Primary plastics derived from personal care products have received much attention but are generally larger in diameter (74 to 800 μm) (*16*) than those we observed. Manufacturers of brightly colored microbeads cite primary uses in research and medical applications as well as industrial paints. We identified several pink microbeads as poly(methyl methacrylate) (PMMA), which is used in a variety of industrial paint and coating applications. Entrainment to the atmosphere could easily occur for the many industrial coatings and paints that are applied using aerosol sprays, but these may not be the only atmospheric emission sources. Because the density of most microbeads is lower than that of seawater, entrainment could also occur from the surfaces of aquatic systems through aerosolization

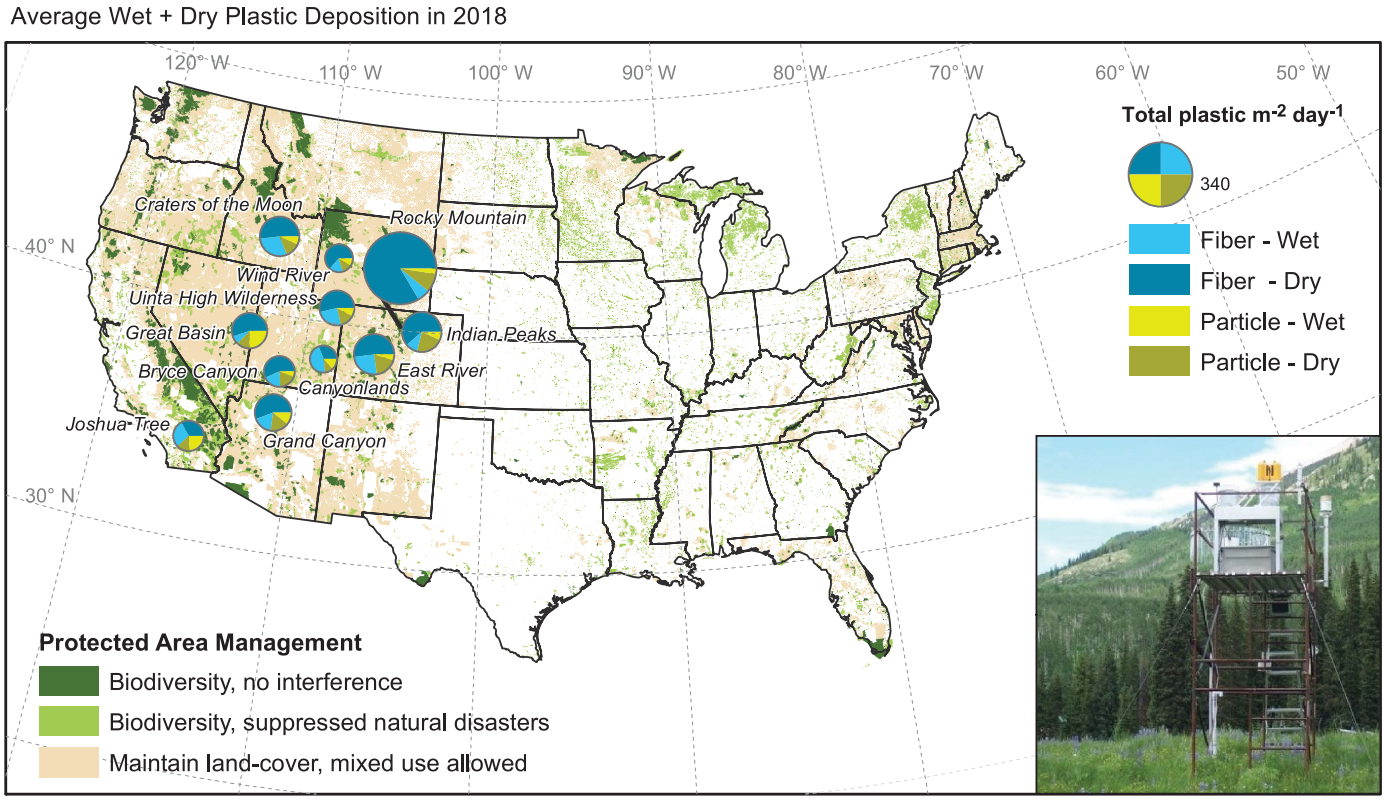

Fig. 1. Average deposition rates of plastic fibers and particles, wet plus dry, to selected national parks and wilderness areas of the United States. The pie chart sizes reflect plastic fluxes to each site. Protected areas base map is from the United States Geological Survey (USGS) Protected Areas Database (PAD). Pictured on the bottom right is a standard National Atmospheric Deposition Program (NADP) Aerochem Metrics wet-dry collector located in East River, Colorado.

Table 1. Relationships between weekly wet plastic deposition rates, dust, population statistics, and air-mass trajectories. Pearson correlation coefficients (r) and model coefficients of determination (r^2) between wet plastic deposition rates and potential drivers. Full model selection is based on the Akaike Information Criterion (AIC), and parameters included are shown in bold. NA, not applicable.

National park or wilderness	Dust (r)	Total population (r)	Total populated area (r)	Distance (r)	Mean wind speed (r)	Full model (r^2)
Grand Canyon, AZ	0.16	0.70***	**0.56****	**0.80*****	0.41	0.69***
Wind River Range, WY	**0.74*****	**0.32**	**0.31**	0.34*	**0.3**	0.77***
Craters of the Moon, ID	−0.11	0.05	**0.43****	**−0.12**	0.01	0.21**
Rocky Mountain, CO	**0.27***	**0.35****	0.12	0.18	0.05	0.20**
Joshua Tree, CA	0.16	**−0.45**	**−0.24**	**0.96****	0.63*	0.71*
Uinta High Wilderness, UT	**0.2**	**−0.48**	**−0.32**	**0.08**	**0.11**	0.86
Canyonlands, UT	**0.44***	0.01	−0.16	0.05	**0.25**	0.19
Indian Peaks, CO	**0.77****	**0.42**	**0.66***	**0.29**	**−0.15**	0.99**
East River, CO	**0.58*****	−0.11	−0.12	−0.05	−0.04	0.34***
Great Basin, NV	**0.41***	**0.51****	**0.17**	**0.48****	0.18	0.59***
Bryce Canyon, UT	−0.13	−0.02	0.001	−0.06	−0.1	NA

*P < 0.1; **P < 0.05; ***P < 0.01.

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under turbulent conditions. An analogous process has been shown to aid in the dispersal of algae and other particles across thousands of kilometers (*17*). The dominant size classes of microbeads observed were <20 μm, therefore also subject to global atmospheric dispersal, which indicates that the source of these beads is not necessarily in the continental United States.

First-order estimates of mass deposition rates to each national park and wilderness area were determined using two independent methods. The first method uses the mean deposition rate based on visual count estimates (Fig. 1 and Table 3) and the range of densities observed for the plastics identified (0.92 to 2.2 g cm^{-3}) to calculate the total annual loading of plastic to each protected area. The second method uses FTIR-based estimates of the polymer proportions within our samples. Method 2 estimates are larger but similar to those of method 1 (r = 0.89, where r is the correlation coefficient). Estimated, site-specific annual deposition rates ranged from 48 ± 7 to 435 ± 9 plastics m^{-2} day^{-1}, or 0.22 to 22 metric tons of plastic per year scaled to each park or wilderness area (Table 3). On the basis of these data, we approximate that >1000 tons of plastic from the atmosphere are delivered to western protected areas in the United States, including national parks and wilderness areas, each year. This is equivalent to ~120 to 300 million plastic water bottles.

The finding that microplastics are ubiquitous in the atmosphere and are transported to distant locations has widespread ecological implications. Though the literature is still sparse on the effects of microplastics on terrestrial organisms (*18*), accidental ingestion of plastics by aquatic organisms has been shown to lead to blockages in the intestinal tract causing internal injury, reduced energy, and behavior modifications (*16*, *18*, *19*). In some cases, ingested plastics have been shown to transfer up the food chain (*16*, *18*, *19*). Less is known about the influence of microplastics on microbes, but recent work has suggested that plastics can influence microbial community composition (*20*). This observation leads to key questions about whether plastic-altered microbial communities in receiving terrestrial ecosystems could lead to changes in biogeochemical processes. As plastics accumulate in pristine wilderness, we may anticipate shifts in community composition, possibly leading to declines in biodiversity on the basis of the different tolerances to the physical and toxicological consequences of consuming microplastics. Further, because plastics can influence thermal and hydrologic properties of soils (*21*), changes in the biogeochemical cycling of nutrients in protected environments may also occur with unforeseen consequences. Many of our study locations are mountain environments that tend to have simple food webs and shallow soils (*22*, *23*), which makes them particularly sensitive to perturbations and might lead to an amplified response to microplastic deposition.

To date, only a handful of studies have quantified atmospheric microplastic loading rates to urban and remote settings (*4*, *5*, *24*), and there is a clear, growing need for these types of studies. We show that the intersection of 48-hour air-mass trajectories with, and their proximity to, population centers are coincident with enhanced rates of plastic deposition (up

Table 2. Comparison of dry and wet plastic deposition rates and their potential drivers. Pearson correlation coefficients (r) between weekly wet and monthly dry deposition rates of plastic fibers and particles and indices of regional and broad-scale climate patterns. The temperature anomaly for the Western United States is used here as an index of jet stream location [data: National Oceanic and Atmospheric Administration (NOAA) National Centers for Environmental Information; www.ncdc.noaa.gov/cag/regional/time-series]. ENSO, El Niño–Southern Oscillation; F-stat, F statistic.

Deposition type	Dust (r)	Season (F-stat)	ENSO (r)	Temperature anomaly (r)
Dry deposition (total)	−0.24**	0.63	0.21**	−0.25***
Dry fiber deposition	−0.22**	0.36	0.19*	−0.23**
Dry particle deposition	−0.24**	5.64***	0.29***	−0.36***
Wet deposition (total)	0.37***	3.61**	−0.13*	−0.02
Wet fiber deposition	0.21***	2.91**	−0.12*	−0.03
Wet particle deposition	0.36***	1.71	−0.08	−0.04

*P < 0.1; **P < 0.05; ***P < 0.01.

Table 3. Annual plastic deposition rates to 11 U.S. protected areas. Estimated annual deposition rates of microfibers and plastic particles to national parks and wilderness areas of the United States. Data are based on observed deposition rates to each site from late 2017 to early 2019.

National park or wilderness	State	Size (km^2)	Mean plastic deposition rate (plastics m^{-2} day^{-1})	Metric tons of plastic per year (visual counts)	Metric tons of plastic per year (FTIR proportions)
Grand Canyon	AZ	4926	112 ± 6	10.7–11.9	11.0–21.3
Wind River Range	WY	7252	68 ± 6	9.3–11.1	10.9–22.3
Craters of the Moon	ID	2893	139 ± 10	7.7–8.8	11.5–19.3
Rocky Mountain	CO	1047	435 ± 8	9.4–9.8	4.2–9.0
Joshua Tree	CA	3200	54 ± 2	3.4–3.7	3.7–9.8
Uinta High Wilderness	UT	1849	120 ± 6	4.3–4.8	1.6–2.8
Canyonlands	UT	1366	48 ± 7	1.2–1.5	3.0–6.1
Indian Peaks	CO	311	148 ± 5	0.9–1.0	0.4–1.3
East River	CO	300	140 ± 9	0.8–0.9	0.4–0.9
Great Basin	NV	312	107 ± 5	0.65–0.72	0.4–1.3
Bryce Canyon	UT	145	80 ± 6	0.22–0.26	0.4–0.8
All western protected areas	USA	496,350	132 ± 6	1012–2419	1185–3773

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to 14-fold), though a large proportion of the variation is not explained by these local-to-regional factors alone. This result, combined with the size distribution of identified plastics and the relationship to global-scale climate patterns, suggests that plastic emission sources have extended well beyond our population centers and, because of the longevity of plastics, have spiraled through the Earth system. The long-range transport of microplastics, reminiscent of the global dust cycle but distinctly human in origin, is indicative of the ubiquity of the human fingerprint on atmospheric composition; microplastics have the potential to be found far from initial production and source areas.

In highlighting independent life histories for dry versus wet plastic deposition, we provide additional details on the source, transport, and fate of plastics on Earth's surface. Though regional storms were important in delivering larger plastics to national parks, dry deposition accounted for >75% of the plastic mass deposited. This result, along with the relationship of dry deposition to large-scale climate patterns, suggests that although urban centers may be the initial source, plastics accumulate in the atmosphere over longer time periods, are transported long distances, and are deposited during favorable conditions, such as slower air-mass velocities or intersections with mountain ranges. In fact, dry plastic deposition rates showed a significant and positive relationship to elevation (r = 0.69, P < 0.05). However, key questions remain on emission mechanisms and the transport physics of low-density polymers, including atmospheric lifetimes and the role of latitudinal atmospheric circulation patterns. Greater spatial resolution, particularly across latitudinal gradients, and perhaps in situ aircraft-based sampling would provide the data needed to model the atmospheric limb of the global plastic cycle. Identifying the key mechanisms underpinning plastic emissions to the atmosphere is the first step in developing scalable solutions. The consequences to ecosystems are not yet well understood but are inescapable in the immediate future. If the potential dangers posed by environmental microplastics are to be mitigated, both the scale of the solution and the level of cooperation that will be required call on the engagement of the global community.

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SUPPLEMENTARY MATERIALS

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Data S1 to S4

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Here, there, and everywhere

No place is safe from plastic pollution. Brahney *et al.* show that even the most isolated areas in the United States —national parks and national wilderness areas—accumulate microplastic particles after they are transported there by wind and rain (see the Perspective by Rochman and Hoellein). They estimate that more than 1000 metric tons per year fall within south and central western U.S. protected areas. Most of these plastic particles are synthetic microfibers used for making clothing. These findings should underline the importance of reducing pollution from such materials.

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