Title: Plastic Rain in Protected Areas of the United States

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Abstract:

Eleven billion 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 wet versus dry have unique 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 important sources for wet deposition. In contrast, plastics deposited dry were smaller in size and rates were related to indices that suggest longer range or global transport. Deposition rates averaged 132 plastics m^{-2} day^{-1} amounting to > 1000 tons of plastic deposition to western U.S. protected lands annually.

One Sentence Summary: Plastic spiraling in the Earth system

Main Text:

The world produced 348 million metric tons of plastic in 2017 and this number grows every year by approximately 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 the same characteristics lead to the unrestrained accumulation of synthetic material in nearly every ecosystem on the planet (*3*). Though

atmospheric microfibers have been recently 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 UV light. To determine potential sources of atmospheric microplastics and the rate of 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), and dry deposition, collected at monthly or bi-monthly intervals (n=103). We used relationships between plastic deposition rates and air-mass back-trajectories' intersections with population centers, contemporaneous dust (soil) deposition, global indices of climate, and plastic composition to identify both emission and product sources. Understanding the sources of microplastics to the atmosphere, both in terms of emission points and products will, in turn, allow us to implement scale-relevant solutions to mitigate plastic pollution.

Microplastics were present in 98% of all the wet and dry samples analyzed from US protected areas. Observed microplastic particle sizes were between 4 μm and 188 μm and fibers between 20 μm and ~3 mm with average width and depths of 18 and 6 μm respectively (Fig. S2). Approximately 70% of the particles were within range for long-range and even global transport of dusts (< 25 μm) (*6*, *7*) while the majority of fiber lengths suggested regional transport (10 to 1000 km) (*8*). Because plastic density is lower (0.65-1.8 g cm^{-3}) than 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. The process may be similar to

ballooning in spiders where a combination of electrostatic forces and drag allow spiders attached to silk fibers to travel 1000s of km (*10*).

Daily 48-hour atmospheric back-trajectory analyses were determined using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model (*11*, *12*) and compared to weekly wet plastic deposition rates through 2018. Our analyses suggest that wet deposited microplastics originate from different source regions than dry deposition. Wet plastic deposition rates at half 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 at individual sites. The observation that microplastics deposited wet are larger in size and lower in number (Fig. S2) and correlated to both dust deposition and population metrics reflect the role of regional storms in the entrainment and subsequent rainout of microplastics, often after these storms 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, and may thus reflect 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 textiles used for clothing including cotton, polyester, and nylon. We also observed fibers composed of polyolefin more commonly used for household and vehicle carpeting as well as polytetrafluoroethylene and polyethylene fibers used in a variety of industrial applications (*13*). Industrial coatings on fibers, such as Valbond 6053, were also identified, underscoring the diversity in microfiber sources to US 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 greater than fibers released to wastewater during the washing phase (*15*, *16*) and are then transported to protected areas during favorable wind speeds and trajectory (Fig S5).

The polymer compositions of individual plastic particles below 20 μm were more difficult to identify due to diffraction limitation of mid-infrared light. However, in subsamples, almost all brightly colored particles that fell within our counting criteria were identified as synthetic using FT-IR particle mapping in reflection mode, which allows for the mass identification of particles in the subsamples. Using this reflectance mapping technique on 32 sub-samples, we found that 2.5 - - 5%, average 4%, of the identifiable particles were synthetic polymers. This 4% included particles that, being clear or white, did not meet our visual counting criteria, suggesting our estimates of plastic deposition rates based on counts are conservative (Table 3). Most particle compositions found in our samples can be linked back to 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 and 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 (74-800 μm) (*16*) than what 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 are 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 seawater, entrainment could also occur from the surface of aquatic systems through aerosolization 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 less than 20 μm and therefore also subject to global atmospheric dispersal, indicating that the source of these beads is not necessarily from the continental U.S.

First-order estimates of mass deposition rates to each National Park and Wilderness areas were determined using two independent methods. The first method uses the mean deposition rate based on visual count estimates (Fig. 1, Table 3) and the range of densities observed for the plastics identified (0.92-2.2 g cm^{-3}) to calculate the total annual loading of plastic to each protected area. The second method uses FT-IR based estimates of the polymer proportions within our samples. Method 2 estimates are larger but similar to method 1 (r = 0.89). Estimated, site-specific, annual deposition rates ranged from 48 ± 7 to 435 ± 9 p m^{-2} day^{-1}, or 0.22 to 22 metric tons of plastic per year scaled to the Park/Wilderness area (Table 3). Based on this data, we approximate > 1000 tons of plastic from the atmosphere is delivered to western protected areas in the United States including National Parks and Wilderness Areas each year. This is equivalent to approximately 123 - 300 million plastic water bottles.

The finding that microplastics are ubiquitous in the atmosphere and are transported to distant locations has widespread ecological concerns. Though the literature is still sparse on the effects of microplastics on terrestrial organisms (*18*), accidental ingestion 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 suggests plastics can influence microbial community composition (*20*). This observation leads to key questions on 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, perhaps leading to declines in biodiversity based on 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*), making them particularly sensitive to perturbations and creating a potential 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 to 14x), though a significant proportion of the variation is not explained by these local to regional factors. This result, combined with the size distribution of identified plastics, and the relationship to global-scale climate patterns, suggest that plastic emission sources have extended well beyond our population centers and, through their longevity, spiral 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 vs wet plastic deposition we provide additional detail on the source, transport, and fate of plastics on the Earth's surface. Though regional storms were important in delivering larger plastics to National Parks, dry deposition accounted for more than 75% of the plastic mass deposited. This result, along with the relationship of dry deposition to large scale climate patterns, suggests that while 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. Importantly, 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 microplastic are to be mitigated, both the scale of the solution and the level of cooperation that will be required call upon the engagement of the global community.

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Supplementary Materials:

Materials and Methods

Figure S1 to S5

Table S1

References (*25–30*)

External Databases S1-S4

Fig. 1. Average deposition rates of plastic fibers and particles, wet plus dry, to National Park and Wilderness areas of the United States. The pie chart size reflects plastic fluxes to each site. Protected areas base map from the USGS Protected Areas Database (PAD).

Table 1. Relationships between weekly wet plastic deposition rates, dust, population statistics, and air-mass trajectories. Pearson correlation coefficients and model coefficients of determination between wet plastic deposition rates and potential drivers, $p<0.01$***, $p<0.05$**, $p<0.1$*, $p=0.1^{+}$. Full model selection is based on the Akaike Information Criterion (AIC) and parameters included shown in bold.

	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, 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

Table 2 Comparison of dry and wet plastic deposition rate and their potential drivers. Pearson correlation coefficients between weekly wet and monthly dry deposition rates of plastic fibers/particles and indices of regional and broad-scale climate patterns, $p<0.01$***, $p<0.05$**,

$p<0.1$*, $p=0.1^{+}$. Temperature anomaly for the Western USA is used here as an index of jet stream location (Data: NOAA National Center for Environmental Information https://www.ncdc.noaa.gov/cag/regional/time-series)

	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*	-0.22**
Dry particle depoistion	-0.24**	5.64***	0.29***	-0.36***
Wet deposition (total)	0.37***	3.61**	-0.13*	-0.02
Wet fiber deposition	0.22***	2.91**	-0.12*	-0.03
Wet particle deposition	0.33***	1.71	-0.08	-0.04

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/Wilderness	State	Size km^2	Mean plastic deposition rate (p m^{-2} day^{-1})	Metric Tons of plastic per year (visual counts)	Metric Tons of plastic per year (FT-IR proportion)
Grand Canyon	AZ	4,926	112 +/- 6	10.7 - 11.9	11.0 - 21.3
Wind River Range	WY	7,252	68 +/- 6	9.3 - 11.1	10.9 - 22.3
Craters of the Moon	ID	2,893	139 +/- 10	7.7 - 8.8	11.5 - 19.3
Rocky Mountain	CO	1,047	435 +/- 9	9.4 - 9.8	4.2 - 9.0
Joshua Tree	CA	3,200	54 +/- 2	3.4 - 3.7	3.7 - 9.8
Uinta High Wilderness	UT	1,849	120 +/- 6	4.3 - 4.8	1.6 - 2.8
Canyonlands	UT	1,366	48 +/- 7	1.2 - 1.5	3.0 - 6.1
Indian Peaks	CO	311	148 +/- 9	0.9 - 1.0	0.4 - 1.28
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	134 +/- 8	1360 -2450	1238 - 3880