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Constraining the atmospheric limb of the plastic cycle

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Author Contributions

JB conceived of the study with NM and provided the western U.S. dataset. MP incorporated plastics source, transport, and deposition into model (using version originally developed by HM); NM and MP ran the model. MP, NM, JB, ZK, GC, KP developed ideas for sources. NM developed and ran the optimal estimation. JB, NM and MP wrote the original draft of the manuscript, and all the authors edited the manuscript.

This PDF file includes:

Main Text

Figures 1 to 4

Abstract

Plastic pollution is one of the most pressing environmental and social issues of the 21st century. Recent work has highlighted the atmosphere's role in transporting microplastics to remote locations (1, 2). Here we use *in situ* observations of microplastic deposition combined with an atmospheric transport model and optimal estimation techniques to test hypotheses of the most

likely sources of atmospheric plastic. Results suggest that atmospheric microplastics in the western USA are primarily derived from secondary re-emission sources including roads (84%), the ocean (11%) and agricultural soil dust (5%). Using our best estimate of plastic sources and modeled transport pathways, most continents were net importers of plastics from the marine environment, underscoring the cumulative role of legacy pollution in the atmospheric burden of plastic. This effort is the first to use high resolution spatial and temporal deposition data along with several hypothesized emission sources to constrain atmospheric plastic. Akin to global biogeochemical cycles, plastics now spiral around the globe with distinct atmospheric, oceanic, cryospheric, and terrestrial residence times. Though advancements have been made in the manufacture of biodegradable polymers, our data suggest that extant non-biodegradable polymers will continue to cycle through the Earth's systems. Due to limited observations and understanding of the source processes, there remain large uncertainties in the, transport, deposition, and source attribution of microplastics. Thus, we prioritize future research directions for understanding the plastic cycle.

Significance Statement

Microplastic particles and fibers generated from the breakdown of mismanaged waste are now so prevalent that they cycle through the Earth in a manner akin to global biogeochemical cycles. In modeling the atmospheric limb of the plastic cycle, we show that most atmospheric plastics are derived from the legacy production of plastics from waste that has continued to build up in the environment. Roads dominated the sources of microplastics to the western U.S., followed by marine, agriculture, and dust emissions generated downwind of population centers.

74 At the current rate of increase of plastic production (~4% per year), understanding the sources
75 and consequences of microplastics in the atmosphere should be a priority.
76
77 **Main Text**
78 **Introduction**
79 Humans have been generating synthetic polymers or “plastics” since the early 1900s and
80 annual production rates have increased exponentially over the last 70 years. To date, nearly 10
81 billion metric tons (10,000 Mt or 10 Pg) of plastic have been produced globally (3). Though much
82 of this waste is buried in landfills, recycled, or incinerated, an estimated 12-18% of plastic waste
83 ends up in the environment through inadequate management and littering (3–5). Due to their
84 resilience and synthetic nature, plastics do not appreciably decompose; rather, they continually
85 fragment into smaller and smaller pieces. This trait combined with the explosive growth in
86 mismanaged plastics suggests that the mass of accumulated mismanaged plastics may be
87 increasing at a rate of 2- to 10-fold on the decadal time scale (4, 6–8).
88 Though research is still limited, microplastics in the environment influence soil processes
89 and plant production (9–11), alter microbial community composition (12–14), are consumed by
90 biota leading to impaired health and mortality (15), transfer up the food chain (16), and act as
91 vectors for contaminants (17, 18). Microplastics and their associated contaminants are inevitably
92 consumed by humans, which may lead to adverse health effects (19, 20). Given these preliminary
93 findings, the accumulation and transport of microplastics in the natural environment may have
94 negative and as yet unknown consequences for ecosystems and human health. As plastics make
95 up an increasing fraction of our soils, surface waters, biota, and atmosphere, quantifying the
96 environmental transport processes, rates, and residence times in ways that are analogous to global
97 biogeochemical cycles is necessary to constrain the global plastic cycle (21–23).
98 While the role of the ocean and riverine systems in accumulating and transporting
99 microplastics has been recognized (4, 24), recent studies have highlighted the importance of the
100 atmosphere as a transporter and reservoir of plastics (2, 25). Remote deposition rates recorded

from around the world range from 50-700 plastics m^{-2} d^{-1} (1, 2, 25–29). Based on available data, some 22 Gg (22,000 tons) of microplastic are potentially deposited across the contiguous U.S. each year (2). These studies have generated numerous questions regarding the plastic cycle including, how are plastics emitted to the atmosphere? What are the main sources? Where can we expect to find hotspots of microplastic deposition? How long do plastics remain aloft? And, given the current rate of plastic deposition, what can we expect in the future?

At present, it is unclear how plastics are emitted to the atmosphere. Unlike smaller atmospheric particles (<2.5um) that are emitted directly through combustion or formed in the atmosphere, coarse mode particles (>2.5um) are typically entrained into the atmosphere through mechanical processes, such as dust entrainment during strong wind events, or wind or wave breaking of sea surface spray (30). It is reasonable to hypothesize that plastic emissions may occur around population centers, where available data indicate relatively high plastic deposition rates (27–29). However, back-trajectory analyses have shown that only a small portion (10 and 25%) of total plastic deposition to remote locations is attributable to direct emissions from population centers (2). Notably, most of the deposited mass was instead related to large-scale atmospheric patterns. In addition to population centers, other, less intuitive sources of atmospheric microplastics are likely.

We postulate that, similar to other coarse mode aerosols, microplastics (<5 mm) are entrained into the atmosphere through mechanical processes, even if this is not the primary source of the plastics to the environment (Figure 1). For example, concentrated areas of plastics and microplastics in marine environments represent an important potential source of microplastics that can be aerosolized through wind or wave action, similar to sea spray aerosols (24, 31). Insoluble plastic particles tend to be concentrated at the top of the mixed layer due to their low density and upward transport by gas bubbles. Thus, these particles are easier to entrain into wind or bubble-generated sea spray (32). Secondly, vehicle tires, brakes, and road surfaces contain plastic, which can be worn and generate microplastics that are emitted into the environment (33–36). More importantly, the mechanical process of vehicle tire movement, the braking process, and the intense

turbulence in the wakes of vehicles, allow these roadside plastics to gain sufficient mechanical energy to overcome inertial or cohesive forces and be resuspended into the atmosphere. A third potential re-emission source of plastics are dusts produced from agricultural fields during tilling or when fallow. Agricultural fields are likely hotspots of soil plastic concentrations for two primary reasons. Approximately 55% of the biosolids produced in U.S. waste treatment operations are applied as fertilizer around the country. This application of biosolids to agricultural fields is also practiced globally (37, 38). Because ~98% of the microplastics in wastewater are retained in biosolids (39), the application of biosolids to agricultural fields represents a significant pathway for microplastics to enter the environment. In addition, plastic mulch is often added to soils to increase temperatures while retaining moisture (40). Fourth, if atmospheric plastic deposition is ubiquitous, microplastics should be found in the soils of most landscapes. Thus, it stands to reason that microplastics can be re-emitted to the atmosphere from soils undergoing wind deflation, especially close to or downwind of population centers (hereafter referred to as “population dust”).

To what extent each of these sources may contribute to the atmospheric burden of plastics is not yet clear. Our goal in this study is to combine the limited observations of atmospheric microplastics with models to better identify the open research questions. Here we use a detailed deposition dataset available for the remote parts of the western U.S. in combination with a microplastics-enabled version of the Community Atmospheric Model (CAM) (41) to determine for the first time the most likely sources of atmospheric microplastics, their residence time in the atmosphere, and accumulation areas. This very detailed deposition dataset includes temporal and spatial variability, in addition to size-resolved count and volume information about plastics, which allow us to uniquely consider the plastics number and mass (2). Both mass and number are important, but here we focus our results on mass to frame our understanding of plastic movement through the Earth System and because mass will better reflect the ecological and biogeochemical implications.

The aerodynamic size of plastics is very important for atmospheric residence time but is still poorly understood for fibers and other asymmetric shapes (42, 43). Recognizing that models

155 may overestimate dry deposition rates for large asymmetric particles of well-studied aerosols like
156 dust (42, 43), we simulated transport of particles with aerodynamic diameters ranging from 0.3 to
157 70 micrometers and used three different assumptions about the size distribution of the deposited
158 microplastics for our model-data comparison, with our base case being the medium size (Figure
159 2a). Similar to other relatively large insoluble particles like dust, we assume that plastics can be
160 scavenged in precipitation events, as seen in the observations (2, 44). Note that our observations
161 only cover the diameter size range from 4 μm to 250 μm, and that is the size range we consider
162 here. The spatial distribution of the sources is fixed and used as the plastic source for the
163 atmospheric modeling of the 3-dimensional distribution, transport, and deposition, but the strength
164 and size distribution of the different sources is varied to best match the observations (more details
165 in the Methods section). Errors are based on model-data comparisons for the exact same time
166 period as well as field, lab, and process blanks.
167
168 **Results and Discussion**
169 We estimated that the current average total atmospheric burden (content) of microplastics
170 over the land regions of the western U.S. is 1 Gg (0.001 Mt) (Figure 1). The largest contributor to
171 modeled plastic deposition in the western U.S. is from road dust sources (84%), while ocean
172 emissions contributed 11% of the plastic deposition. Agricultural dust represents plastics entrained
173 into the atmosphere from agricultural fields and contributes 5% of the plastic deposition (Figure 1).
174 Interestingly, both sources of plastics from population centers, either from dust generated
175 downwind of population sources (population dust) or directly related to population, represent a
176 much smaller contribution (0.3% and 0%, respectively) (Figure 1).
177 To better understand the relative plastic source contributions at the observational sites, we
178 report the modeled contribution at the sites over the observed time period (Figure 2b). The road
179 source contributed 92.5% [69-100%] of the modeled annual average deposition (the bracketed
180 values represent the 95% confidence limits across the three size cases), while the oceans and
181 agricultural dust sources contributed 4.0% [0-17%] and 3.4% [0-22%], respectively (Figure 2b). The

182 population dust and the population source contributed $2x10^{-7}$% [0-8%] and 0% [0-13%],
183 respectively. Notice that the contribution of plastic sources at our observational sites are slightly
184 different than over the whole western U.S. (in Figure 1). This is, in part, because the ocean source
185 tends to be larger closer to the coasts than at our observational sites. The road source has the
186 smallest uncertainty, with a range spanning 30%, while the other sources suggest an uncertainty
187 of 100%, indicating that this first estimate of the relative contributions of sources should be refined
188 in future studies.

189 A comparison of our model results with the detailed plastic deposition data from the
190 western U.S. suggests that the model is able to simulate the range of plastic deposition seen at the
191 sites, which provides a measure of confidence in the relative source attributions presented for the
192 western U.S (Figure 3b). The model is not able to simulate all the variability in these remote and
193 mountainous regions (Figure 3b; SI Appendix, Fig. S1; Table S1), as it is similarly unable to
194 simulate dust and sea salt aerosols at these sites (SI Appendix, Table S2). While remote sites
195 represent our best opportunity to sample air uncompromised by local sources, and thus understand
196 the long-range transport of plastics, the complex terrain of mountainous regions make it difficult for
197 models to accurately simulate transport and deposition. The model results showing the relative
198 contribution of different sources to the final model result highlight that the ocean source of plastics
199 is not well constrained by these observations. These stations are distant from the shore, which
200 contributes to the large uncertainty in the ocean source (Figure 3c-3e).

201 The uncertainties in the aerodynamic size of the plastics are important for the modeled
202 uncertainties (contrast the square, triangle, and diamond symbols in Figure 2b), and we include all
203 three size distributions in our 95% estimates above. This suggests that characterizing the
204 aerodynamic behavior of these heterogeneous particles is important for understanding their
205 transport pathways. In addition, the sensitivity studies show that having temporally resolved data
206 improves our constraints on the sources (Figure 2b: cyan symbols and lines are constraints using
207 only annually averaged model-data comparisons at each of the 11 sites). Even more important is
208 having 11 different observing stations; excluding individual stations from the analysis enlarges the

95% confidence limits for all the sources (Figure 2b: green symbols and lines). Thus, more detailed spatial and temporally resolved data is vital for improving our understanding of the long-range transport of microplastics.

Importantly, our data do not include plastics smaller than 4 μm as this fraction has yet to quantified (45), but such particles could have longer residence times than the ones included here (weeks instead of hours). It is not known how important plastics smaller than 4 μm are in the atmosphere and their behavior needs to be measured and assessed. The data shown here do, however, indicate that particles and fibers decrease in number as they decrease in size (Figure 2a). This suggests that, similar to other mechanically generated aerosols (like dust), most of the mass is emitted in larger sizes; for dust, less than 10% of the PM10 is emitted in PM1 (46, 47). Smaller particles tend to be more difficult to entrain into the atmosphere as they experience stronger cohesive forces and yet have smaller cross-sectional areas exposed to the winds. While this is not the case for the plastics emitted with sea spray, which could theoretically contain small plastics, the locations of the measurement stations far from shore do not allow us to constrain the ocean source well, especially as these small particles would be embedded in larger hygroscopic sea salt particles that would reduce their atmospheric residence time. More in situ and laboratory studies are required to better understand the number and mass of plastics that are emitted in the submicron size fraction.

Next, we consider whether other observations support the inferred sources. Because of the limited data, we extrapolate our study globally for this comparison. Our emission estimates of long-range transported microplastics from tire wear and braking are on the low end of recent bottom-up estimates (here 96 [63-110] Gg yr^{-1} vs. 284 (102-787) Gg yr^{-1}) (36) (Figure 4a). The range in (36) is associated with assumptions about the fraction of the plastics emitted for long-range transport, which they assume is less than 10µm. In their model, they used a range of values representing their range of observations (29). Here, in contrast, we constrain this source using the remote observations. Our results suggest that the lower end of their assumption is more likely correct. However, the differences between their bottom-up estimates and our top-down results

236 could be due to errors in our modeling or model-data comparisons, or that emissions may be
237 smaller in the western U.S., where our sites are located, compared to Europe, where the bottom-
238 up estimate derives from. For example, in Europe and Asia, there is documented recycling of
239 plastics into the production of road surfaces (35, 48), while this practice is still limited in the United
240 States. The differences could also be due to the small fraction of emitted microplastics that are
241 actually suspended high enough to be entrained in the atmosphere for long-range transport.
242 Concentrations of large particles close to the surface (<2m) can be 1-4 orders of magnitude greater
243 than the concentration in the boundary layer that can be transported long-range, as large particles
244 settle rapidly out of the atmosphere (49, 50).

245 Our modeled spatial distribution of the ocean deposition is driven by the source term for
246 the microplastics, which is driven by a combination of observed microplastic concentrations in
247 gyres, especially the North Pacific and elsewhere that the sea-spray source (driven mostly by
248 winds) is strongest in the model (Figure 4; SI Appendix, Fig. S4; (24), see Methods for more details).
249 Our assumptions of oceanic microplastic concentrations are based on syntheses of ocean
250 microplastic observations (10). Notice that, in the western U.S., the deposition from the ocean
251 source drops off quickly as we move onto land, and, by the time we are at the remote mountain
252 stations used for this study, plastic deposition has dropped substantially (Figure 3d). This is
253 because of the short residence time of the plastics in the size range studied here (SI Appendix,
254 Table S3). In other words, our results hint that there could be significantly large sources of plastics
255 from oceans (globally perhaps 8.6 [0-22] Tg or Mt yr^{-1}, including the 95% confidence limits), but the
256 location of our sites is not ideal to characterize these emissions. Recent studies on the ocean coast
257 in France suggest higher observed marine concentrations, as compared to our modeled
258 concentrations of microplastics (Observed: ~9 microplastics m^{-3} vs. base modeled value of 0.06
259 microplastics m^{-3} (31)). However, the observations in France were taken close to the surface and
260 may not be representative of long-range transported plastics. In addition, the mass of plastics
261 required for our source is lower than recent studies have observed in the ocean for a slightly larger

size fraction (SI Appendix, Fig. S3; (24)), suggesting that our estimates of ocean sources may actually be on the low side.

Our estimates of agricultural microplastic emissions with dust over the western U.S. are ~0.2 Gg per year, and, if we extrapolate globally, 69 [0-450] Gg yr^{-1}; notice there tends to be greater agricultural dust emissions outside the region where we have data, so this extrapolation requires validation through additional sample collection (SI Appendix, Fig. S4b). Using satellite data, global emission estimates of long-range transported agricultural dust suggest a source of ~34 Tg yr over North America (much of this east of our region), or ~170 Tg yr^{-1} globally (51). Dividing our inferred sources for the plastics from agricultural dust in the western U.S. and the world by these published estimates of the agricultural dust production, we infer a concentration of microplastics in agricultural soils of 7-400 mg kg^{-1}. Observations range from 0.2 mg kg^{-1} in areas of the U.S. where biosolids have not been applied to >2000 mg kg^{-1} in areas of China where plastic mulching is commonly used (52–56), indicating our agricultural microplastics sources are within the large range of reported values.

Constraints on the strength of the plastic sources downwind of populations (population dust) or directly from population sources are not available in the literature, even though this is where most plastics are used. It is somewhat surprising that population sources were less important in describing our observed spatial and temporal microplastic data. It may be that the modeled distribution of plastics from population centers looks very similar to the tire wear and braking source, but the latter does a better job of simulating the observed distribution. This suggests that, although population centers may provide the initial source of plastics waste, roads provide the mechanical energy to emit these plastics to atmosphere (33, 34). Plastics emitted directly from population centers could be too large for long-range transport and get deposited nearby, where they can gradually degrade to microscopic sizes due to sunlight exposure, temperature changes, freezing and melting water, and mechanical forces from vehicles.

Hann et al. (2018) (57) analyzed the generation and fate of microplastics in the environment from populated areas of the European Union. Sources related to roads (break and tire

289 wear, road markings) made up most of their reported emissions. In addition, they identified several
290 other relevant sources, the largest being losses of pre-production plastic pellets, clothes washing,
291 building paints, and artificial turfs. Plastic pellets come in sizes too large for long range atmospheric
292 transport. Building paints and artificial turfs could lead to direct atmospheric emissions of
293 microplastics; however, these are likely to only be a small fraction of the emissions as it is hard to
294 get small particles airborne. Emissions of synthetic microfibers to the environment from apparel
295 washing are reported to be up to 350 kt/y globally (58). Emissions from laundry drying have been
296 reported to be several times greater (59) or comparable (60) to the fibers emitted in wastewater.
297 Emissions to the environment from plastic waste recycling may also be similar in magnitude (60).
298 Unfortunately, there is no information available on what fraction of the microplastic from these
299 sources would be small enough for long range atmospheric transport.
300 Reported fluxes of mismanaged macroplastic pollution to the environment are much larger
301 than microplastic fluxes and, according to the model of Kawecki & Nowack (2019) (60), about 2/3
302 of mismanaged macroplastic pollution end up on roadsides where exposures to environmental
303 factors can break it down to microscopic fragments. Thus, by virtue of their proximity to people,
304 roads can effectively accumulate from a variety of sources plastics that are subsequently broken
305 down and emitted high into the atmosphere by traffic. Support for this idea comes from an
306 examination of road dust in the district of Tehran, Iran. The researchers found a diversity in urban
307 microplastic color and character, suggesting most plastics were derived from multiple commodity
308 sources rather than just tire wear (61). Based on this reasoning, it follows that road sources
309 dominated the atmospheric loads to the western U.S., even though tire and road wear plastics were
310 rare in the observed samples (2). It is worth noting that most of the sites in the western US study
311 region are in arid regions (9/11 sites), which may increase the potential or road-based emissions
312 as compared to wet regions where surface runoff may migrate microplastics to soils or waterways.
313 More studies on primary plastic sources, environmental fate, and entrainment are required to better
314 constrain this aspect of the plastic cycle.
315

316 **Implications and Future Work**

317 For this study, we used the most complete observational dataset published of atmospheric
318 plastic deposition, which comes from in the western U.S.(2). We extrapolated our model to the
319 global level (Figure 4a) to see what additional work needs to be done, which regions should be
320 prioritized for more observations (Figure 4b), and to understand the implications of this study on
321 the global plastic cycle. This extrapolation assumes that similar sources of dry and wet deposition
322 occur globally as compared to those in the western U.S., and thus should be considered tentative.
323 Comparisons of our model results to the limited available global data suggest that the model results
324 tuned to one dataset in North America may underestimate deposition in Europe, although most of
325 the compared estimated rates are based on deposition data from a limited time period and may not
326 robustly represent long term microplastic deposition (SI Appendix, Fig. S4; Table S1) (1, 25, 27,
327 28, 62).

328 Our study suggests the strongest sources and greatest deposition rates of plastics occur
329 over the ocean (Figure 4b), especially over the Pacific and Mediterranean, which aligns with recent
330 evidence indicating that these waters have 2 to 3 times the fiber concentrations of other ocean
331 basins (24, 63); however, even these emissions are poorly constrained by our observational sites
332 (see large uncertainty in Figures 2b and Figure 4a, and distribution of modeled deposition from
333 oceans: Figures 3d and SI Appendix, Fig. S2). The U.S., Europe, Middle East, India, and Eastern
334 Asia were hotspots for terrestrial plastic deposition (Figure 4b, SI Appendix, Fig. S2). Ocean
335 sources of terrestrial plastic deposition were important in coastal areas including the west coast of
336 North America, the Mediterranean region, and southern Australia (SI Appendix, Fig. S2). Dust and
337 agricultural sources of plastic deposition were more important in northern Africa and Eurasia, while
338 road sources were more important in heavily populated regions (SI Appendix, Fig. S4). More
339 observations are required in all these locations that are modeled to have high concentrations in
340 order to verify that the sources postulated here are correct.

341 Next, we considered our estimated net import and export of plastics from each ocean and
342 land region (Figure 4c). While the data we used to constrain the sources concentrated on the

western U.S., the largest source of plastics was hypothesized to be from Africa, followed by Asia, suggesting more work in these continents is vital. The total deposition of plastic to ocean surfaces from land sources was 13 Gg yr^{-1} and total terrestrial deposition from ocean sources was 22 Gg yr^{-1} (excluding coastal grid boxes). Because oceans dominated the atmospheric burden of plastics, most continents were net importers of plastic material, except for South America, which was neutral. Antarctica had the greatest imbalance as it has zero emissions of plastics, and yet receives deposition of 3.4e-5 Gg yr^{-1}. Most other regions imported plastics from the ocean at a rate that was 4-9% of their emissions, considering only grid boxes away from the coastal regions.

As plastic aerosols have unusual shapes (e.g. long fibers), the spectrum of residence times of these particles and fibers in the atmosphere is not well known and needs to be studied. Here, we used three different size distributions that span the available sizes within the data (Figure 2a). We estimated that atmospheric residence times ranged from 0.04 days (~1 hour) to 6.5 days for the different sizes of plastic particles simulated here (SI Appendix, Table S4). The largest atmospheric source, the ocean, had the shortest residence time ranging from 0.1 to 1.7 days, with the mass-weighted mean lifetime being 0.10 days. Road sources ranged from 1 hour to 2.9 days (mean: 0.62 days), while agriculture or population dust plastics had the longest atmospheric residence times ranging from 0.06 - 5.8 days and 0.07 to 6.5 days respectively (mean: 0.92 and 0.91 days, respectively). Since fine aerosols can travel between continents in just a few days (64–66), these data suggest that, under the right conditions, plastics can be transported across the major oceans and between continents, either in one trip or by resuspension over the oceans. More data on how these plastics will behave in the atmosphere in terms of their dry and wet deposition rates is needed to better constrain the residence times. Note that here we do not have data for microplastics smaller than 4um, and these difficult to measure particles need to be quantified as well.

We determined that, at present, microplastics make up less than 1% of the anthropogenic aerosol deposition over terrestrial environments, but alarmingly already may make up greater than 50% [0-90%, 95% confidence limits] of the net anthropogenic atmospheric aerosol deposition over

parts of the oceans downwind of the major ocean plastic source, making up more than black carbon, organic carbon, sulfate, and agricultural dusts combined (SI Appendix Fig. S5). This is not entirely surprising, given that the observed percentage of plastics in all aerosol deposition in remote mountains sites ranged from 2-6% (average 4%) (2). A recent examination of plastic concentrations in agricultural soils found microplastics even in fields where biosolid and plastic mulch applications had not occurred (52). In addition, despite the large size of the microplastics and their short residence times, contemporary atmospheric surface concentrations over the ocean source regions may be as large as 5% of the mass of anthropogenic aerosols (SI Appendix, Fig. S5). While these concentrations remain low, they are unlikely to impact climate or radiative forcing, these findings underscore the role of atmospheric deposition in contributing plastics broadly across landscape types and their potential for re-emission to the atmosphere. Due to the ubiquity of microplastic deposition, it seems plausible that any and all dusts should contain microplastic pollution. In fact, Brahney et al. (2020)(2) showed strong positive relationships to contemporaneous dust deposition in wet deposition. Given this understanding, the global production of dust and the land-use activities that contribute to the destabilization of soils (67–70) also contribute to the global dispersion of plastic. Although smaller than the current total anthropogenic fine particle emissions (30 Tg yr^{-1}), the current atmospheric source of microplastics is 8.6 [0-22] Tg yr^{-1}, which is of the same order of magnitude as the current anthropogenic and biomass burning sources of black carbon aerosols to the atmosphere (10 Tg yr^{-1}) (71–73). Industrial aerosol emissions should be decreasing in the next few decades (74), while microplastics could be increasing if unmitigated, suggesting that plastic aerosols could become more important in the coming years. These numbers are heavily dependent on the very uncertain ocean source, so more investigation of the ocean source and its potential growth is vital as the direct and indirect effect of plastic aerosols on the climate is not known.

One of the most compelling results to emerge from our synthesis of model and data is that re-emission sources dominate the atmospheric burden of plastics. This implies that the historical production of plastics is important for determining atmospheric plastic concentration and deposition. This result aligns with global plastic production and the fact that most polymer types

397 can take decades or centuries to decompose to base elements (75, 76). In the meantime, they
398 fragment into smaller and smaller pieces and become available for wind transport. Extrapolating
399 from Geyer et al (2017)(3), indicates that, in 2019, plastic production would have represented only
400 4% of total plastic production since 1950. Thus, the amount of microplastics in the environment
401 available for atmospheric transport has grown to the point that it dwarfs primary annual emissions
402 from urban centers. The fact that most continents were net importers of atmospheric plastics from
403 the marine environment highlights the role of legacy plastics in contributing to the atmospheric
404 burden of plastics and its eventual fallout. Removing plastics from the oceans might not only
405 improve marine water quality but also significantly reduce the atmospheric redistribution of the
406 microplastics (7).

407 Current deposition rates in terrestrial environments peak at above 10 mg m^{-2} day^{-1} (Figure
408 3a). Though future estimaes are uncertain, using the high growth rate estimates from (4) suggest
409 that, plastic deposition rates may increase to 100 mg m^{-2} day^{-1} by 2050, which raises questions on
410 the impact of accumulating plastics in the atmosphere on human health as well as wildland soils
411 and waters. The inhalation of particles can be irritating to lung tissue and lead to serious diseases
412 (45), but whether plastics are more or less toxic than other aerosols is not yet well understood (77).
413 At present we know very little about the effect of plastics concentrations in soils or waters and what
414 threshold concentrations start to incur negative abiotic or biotic effects on ecosystem functioning.
415 Although the addition of plastic mulch has shown short-term benefits for plant production, recent
416 studies showed that plastic accumulation in soils ultimately has a negative effect on plant yield and
417 nutrient availably (9, 10). Microplastics may even have the potential to accumulate in plants (78,
418 79). Our relative ignorance of the consequences despite rapidly rising plastic concentrations in our
419 environment highlights the importance of improving plastic waste management (6) or, indeed,
420 capturing ocean plastics and removing them from the system (80).

421 To better constrain the global plastic cycle, in particular the atmospheric limb, several
422 knowledge and data gaps need to be filled. These include size-resolved temporal deposition data
423 across diverse landscapes and specifically on continents where limited to no data exist (e.g. S.

America, Africa), more studies on prospective emission sources including marine, agricultural, and road emissions as they may differ by region, emissions from households and industries, and additional studies on the aerodynamics of plastic fibers, films, and particles. In situ atmospheric concentrations of microplastics and paleo studies including ice core data can improve our understanding of the temporal changes in microplastic transport, transport distances, and deposition rates.

Conclusion

In modeling the sources of plastic to the atmosphere, we show how the global plastic cycle is influenced more by historical plastic sources from mismanaged waste than emissions in the current year. In the western U.S., roads and ocean sources contributed 84% and 11% of the modeled plastic deposition, while agricultural dusts and population sources contributed 5% and 0.4%, respectively. Oceans dominated plastic sources at the global scale accounting for 99% of the deposition to oceans and 7% of the deposition to land surfaces away from coastal regions. Roads, agricultural dust, and dust sources near population centers were also important sources of deposition to terrestrial environments. Because marine sources dominated atmospheric loads, most terrestrial environments were net importers of plastics.

Though our modeling efforts have advanced our understanding of plastic movement through atmospheric reservoirs and provided key insights into the major sources of atmospheric plastics, our first study on the relative importance of different sources leads to more questions than it definitively answers. Specifically, how do rates of emissions from different sources vary by land-use or technology? For example, do European roads emit more plastic than U.S. roads due to polymer additions to asphalt binding agents? Or is population density a better predictor? How do agricultural soil microplastic concentrations vary between countries that use different practices? How do coastal sea spray emissions vary? Do changes in ocean circulation matter? In addition, key questions remain on the dominant size fractions found within the atmosphere, particularly those in the nanoplastic range. With respect to future emissions, changes in global waste management of plastics will influence emission rates from the key oceanic and terrestrial sources, but more

information is needed on oceanic and terrestrial reservoir residence times. Finally, important questions remain on the potential impacts to the atmosphere and climate. Similar to other insoluble particles such as desert dust, do atmospheric microplastics act as cloud condensation or more likely ice nuclei? Additional data on in situ atmospheric microplastic concentrations as well as contemporary and historical deposition rates in space and time will further improve our understanding of the atmospheric limb of the plastic cycle.

Materials and Methods

Deposition data:

Deposition data from the western U.S. were collected at National Atmospheric Deposition Network (NADP) stations over a 14 month period using Aerochem Metrics model 31 wet/dry collectors (ACMs) fitted with Dry Sampling Units (81). Dry deposition data were collected at monthly intervals while wet deposition was collected at weekly intervals at 11 stations (listed in SI Appendix, Table S1). A total of 313 samples were counted for total microplastic abundance. The size and length of each particle and fiber were determined. The minimum visible size was approximately 4 μm, and size distribution was skewed unimodal tapering towards the smaller size classes, suggesting the data captured the bulk of the plastic mass by size class. Mass deposition rates were determined based on the mean density and detailed analysis of the distribution of polymer sizes and volumes. See (2) for more details.

Atmospheric Microplastics Transport model:

The Community Atmosphere Model (CAM) version 5 in Community Earth System Model (CESM) version 1.2.2 (82) was applied for computing the atmospheric transport of the microplastics. The model was run with 1-degree horizontal resolution for years 2015-2019, with the first year discarded for spin-up, forced with meteorological data from the Modern Era Retrospective Reanalysis for Research and Analysis (MERRA-2), which represents a combination of observations and models for each 6 hour time period (83). The default model includes aerosol representations of anthropogenic and natural aerosols, which interact through radiative and cloud processes with the physical climate (84). The default model includes sea spray and dust sources, which are

478 prognostically calculated based wind strength as well as soil moisture for dust sources (84, 85).
479 The model output consisted of monthly and daily maps of source, concentrations, and wet and dry
480 deposition of microplastic tracers, windblown dust, sea spray aerosols, and monthly average fields
481 of other aerosols and meteorological quantities.

482 The microplastics were added to the model as insoluble aerosol tracer species with 6
483 different aerodynamic diameters in different bins (0.3, 2.5, 7, 15, 35 and 70 micrometers) using a
484 separate aerosol framework (86), as these size bins are likely to be able to resolve the evolution of
485 the size distribution due to differences in lifetimes of particles (e.g.(87)). These do not impact
486 radiation or cloud formation but are subject to wet and dry deposition removal processes. Since the
487 aerodynamic size of the plastics is very important for the atmospheric residence time, but poorly
488 understood for fibers and other asymmetric shapes (42, 43), and recognizing that there are
489 suggestions that models may overestimate dry deposition rates for large asymmetric particles even
490 for well-studied aerosols like dust (88), we use three different size assumptions for our model-data
491 comparison, with our base case being the medium size (Figure 2a). We assumed a density of 1 g
492 cm^{-3} for all particles. For each source, the source size distribution is tuned so that the average size
493 distribution of deposition at the sites is matched for each of the three different sizes, using the
494 information from the observations for both the number and volume of the plastic particles and fibers.

495 We considered a number of possible microplastic emission sources including marine, road,
496 dust, and population centers. In order to simulate the marine emission of microplastics with sea
497 spray, we first estimated the distribution of microplastics in the ocean surface layer using the
498 following method to reproduce a smooth field that reproduced the observations (10). As the
499 microplastic particles in the ocean surface layer are lighter than the water, they accumulate in
500 convergence zones with downwards currents and slow velocities. We used the horizontal water
501 velocity fields of the upmost ocean layer from the CESM Large Ensemble (89) transient simulations
502 of the 20^{th} century and computed a proxy for the plastic concentrations proportional to the flow
503 convergence and inversely proportional to its velocity. This proxy was normalized to one and taken
504 to power x=10, which was determined by fitting to reproduce the dynamic variability of published

505 measurements and modeling studies (5, 80, 90–94). The different ocean basins were further
506 calibrated so that the cumulative microplastics for each basin matched the mean of the three
507 estimates for each basin (24). The spatial distribution of microplastic concentrations in the upper
508 ocean was held fixed, while the emissions were prognostically calculated at every time step, as a
509 function of this spatial distribution times the sea-spray source and multiplied by a globally constant
510 factor deduced from the observations, as described later.

511 Road tire and braking emissions come from an inventory (36), representing microplastics
512 in fine, coarse, and larger aerosol size ranges, based on road-miles driven and braking estimates
513 using the GAINS (Greenhouse gas– Air pollution Interactions and Synergies;
514 http://gains.iiasa.ac.at) model (73). The spatial distribution of this source was fixed to the sum of
515 tire and braking emissions and held constant in time and multiplied by a global constant deduced
516 from the observations.

517 Simulations of agricultural sources of microplastics assumed that all crop areas have the
518 same soil fraction of microplastics and simply apply the fraction of the grid box that is a crop for
519 2005 based on land use land cover datasets for the Climate Model Intercomparison Project
520 (CMIP5) for the CESM (95). This crop area factor is multiplied by the prognostic dust generation
521 within the model, which depends on low values of leaf area index (<0.3 m^2 m^{-2}) and soil moisture,
522 as well as strong winds, to generate dust and thus microplastics in dust (85).

523 Population density map from Gridded Population of the World (GPW), v4 (96) for 2015 at
524 15 arc minutes resolution was used as a proxy for direct emissions form households (e.g. dryer
525 vents), businesses, construction work, and waste management. This source was assumed to be
526 constant in time (referred to as population source). To estimate the plastics generated downwind
527 from population sources near arid regions (referred to as population-dust source), the wet and dry
528 deposition maps from this source were added together and overlain with the natural weather and
529 land cover dependent windblown dust emissions from the model to simulate the re-emission of
530 the deposited particles from dry erodible landscapes.

Modeling is done for 2015-2019, and comparisons are made for the exact time period of the observations for the optimal estimation. For the mean distribution, to account for interannual variability, 3 years of simulations are averaged and shown.

Optimal estimation of atmospheric microplastics source

The atmospheric plastic module produces the spatial and temporal variability in the deposition to be compared against observations for the exact same time period. For each source, one global tuning number is estimated using optimal estimation methods to best fit the observations of deposition. For each model source (ocean, road tire and braking (referred to as roads), agricultural dust, population dust, and population) the deposition (dry or wet) is calculated at each site over the observation time period separately for the weekly to monthly dry (n=103) and approximately weekly wet (n=213) observations at each of 11 sites described in (2) and listed in SI Appendix, Table S1. The source strength for each source (S_i) is calculated to minimize the following cost function, which also represents a χ^2 goodness of fit (97).

$$\chi^2 = \sum_j \left[\frac{(y_{model,j} - y_{obs,j})}{\sigma_j}\right]^2 + P \qquad \text{(eq. 1)}$$

Where $y_{obs,j}$ are the 316 observations, and $y_{model,j}$ is found using the following equation:

$$y_{model,j} = \sum_i S_i * R_{i,j}$$

Where $R_{i,j}$ is the modeled relationship between each source (i) and the deposition at each site (j). P represents a penalty which is assessed if any of the sources become negative. The magnitude of this is set to force all the estimations to result in positive values. The σ_j is the model-observational error or uncertainty in each observation, when compared against the model. This will be the sum of direct observational error, in addition to the error in the model's ability to represent accurately the observation. The observational error had a mean of 1.3 fibers in the process blanks for the wet deposition. This translates to a mean error 20% for wet deposition and 1.6% for dry deposition. In addition, the model is not able to perfectly represent the deposition to a particular

557 location, especially in complex terrain. Previous estimates have suggested for natural coarse mode
558 aerosols such as dust that model-data deposition comparisons may be off by 1 order of magnitude
559 (98). Here we estimate our errors using model-data comparisons of dust and sodium (in wet
560 deposition), which indicate an error of ~50% (SI Appendix, Table S2). We also have a constraint
561 that the goodness of fit (χ^2) should be about the same value as our number of observations (n=313)
562 (97), and thus for the comparison using all values, we needed to increase our errors by 50% to
563 meet this criteria (therefore 75%+(30% or 2.4% depending on whether wet or dry deposition)).
564 The minimum in the cost function (χ^2) was found using a global search across the viable
565 values of the source strengths (between 0 and a value that would provide all the required plastics
566 at the deposition sites) combined with the fminsearch in matlab. This was done sequentially across
567 all 5 sources with 30 values for each source (thus 30^5 calculations to initialize the global search).
568 In addition, some of the values were small, so this was repeated using a smaller areal range (0 to
569 0.1 for the small sources) to better define the confidence levels. The 95% confidence levels for
570 each of the source strengths can be estimated by the values of the source strength which produce
571 χ^2 values equal to the minimum value plus 4 (97), if the errors are gaussian. Although our errors
572 are not strictly gaussian, we report these values as an estimate of the source strengths that are
573 within our 95% confidence intervals. We obtain the following cost function (χ^2) for the 3 sizes: 376.7,
574 375.4, and 432.5 for the large, medium, and small particles (shown in Figure 2a). The strength of
575 each source for the base case, using the optimal estimation, by bin is shown in SI Appendix, Table
576 S3.
577 We conduct two additional sensitivity studies using different subsets of our data to
578 understand the importance of the temporal and spatial resolution of our deposition data. First,
579 instead of using 313 data points across 11 sites, we average the dry and wet deposition at each
580 site and use that value to conduct the optimization in equation 1, using only 11 values instead of
581 313. For this sensitivity study, we used the original 50% error estimate, as the (χ^2) were of the same
582 order as the number of sites in that analysis. This study shows that having temporal resolution
583 reduces the range of confidence limits (cyan colors in Figure 2b). Our second sensitivity study

584 explores the value of each additional site, and tests whether one site is overly driving the analysis.
585 Here, each of the 11 sites is excluded from the analysis, resulting in a large range in the confidence
586 intervals (green colors in Figure 2b).

587 In addition, we conduct an additional sensitivity study where we include the a priori
588 information about the source strength of the road and braking source, using a Bayesian approach
589 (described in more detail in the SI Appendex). We also conduct a sensitivity study where we use
590 a different estimate of the spatial distribution of the ocean source, based on a different interpolation
591 of the observations (24) with more details (results described in more detail in the SI Appendix).
592 Neither of these sensitivity studies were as important as which data were included (described
593 above).

594

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608
609 **References**
610
611 1. M. Bergmann, *et al.*, White and wonderful? Microplastics prevail in snow from the Alps to
612 the Arctic. *Sci. Adv.* **5**, eaax1157 (2019).

2. J. Brahney, M. Hallerud, E. Heim, M. Hahnenberger, S. Sukumaran, Plastic rain in protected areas of the United States. *Science (80-.).* **368**, 1257 LP – 1260 (2020).

3. R. Geyer, J. R. Jambeck, K. L. Law, Production, use, and fate of all plastics ever made. *Sci. Adv.* **3**, e1700782 (2017).

4. J. R. Jambeck, *et al.*, Plastic waste inputs from land into the ocean. *Science (80-.).* **347**, 768–771 (2015).

5. M. Eriksen, *et al.*, Plastic Pollution in the World's Oceans: More than 5 Trillion Plastic Pieces Weighing over 250,000 Tons Afloat at Sea. *PLoS One* **9**, e111913 (2014).

6. L. Lebreton, A. Andrady, Future scenarios of global plastic waste generation and disposal. *Palgrave Commun.* **5**, 1–11 (2019).

7. W. W. Y. Lau, *et al.*, Evaluating scenarios toward zero plastic pollution. *Science (80-.).* **369**, 1455 LP – 1461 (2020).

8. S. B. Borrelle, *et al.*, Predicted growth in plastic waste exceeds efforts to mitigate plastic pollution. *Science (80-.).* **369**, 1515–1518 (2020).

9. D. Zhang, *et al.*, Plastic pollution in croplands threatens long-term food security. *Glob. Chang. Biol.* **26**, 3356–3367 (2020).

10. J. Wang, *et al.*, Effects of plastic film residues on occurrence of phthalates and microbial activity in soils. *Chemosphere* **151**, 171–177 (2016).

11. Y. Huang, *et al.*, LDPE microplastic films alter microbial community composition and enzymatic activities in soil. *Environ. Pollut.* **254**, 112983 (2019).

12. E. L. Ng, *et al.*, Microplastic pollution alters forest soil microbiome. *J. Hazard. Mater.*, 124606 (2020).

13. M. T. Kettner, K. Rojas-Jimenez, S. Oberbeckmann, M. Labrenz, H. Grossart, Microplastics alter composition of fungal communities in aquatic ecosystems. *Environ. Microbiol.* **19**, 4447–4459 (2017).

14. S. Oberbeckmann, M. G. J. Loeder, G. Gerdts, A. M. Osborn, Spatial and seasonal variation in diversity and structure of microbial biofilms on marine plastics in Northern European waters. *FEMS Microbiol. Ecol.* **90**, 478–492 (2014).

15. W. Wang, H. Gao, S. Jin, R. Li, G. Na, The ecotoxicological effects of microplastics on aquatic food web, from primary producer to human: A review. *Ecotoxicol. Environ. Saf.* **173**, 110–117 (2019).

16. Y. Chae, Y.-J. An, Nanoplastic ingestion induces behavioral disorders in terrestrial snails: trophic transfer effects via vascular plants. *Environ. Sci. Nano* **7**, 975–983 (2020).

17. C. M. Rochman, E. Hoh, T. Kurobe, S. J. Teh, Ingested plastic transfers hazardous chemicals to fish and induces hepatic stress. *Sci. Rep.* **3**, 3263 (2013).

18. L. M. Ziccardi, A. Edgington, K. Hentz, K. J. Kulacki, S. K. Driscoll, Microplastics as vectors for bioaccumulation of hydrophobic organic chemicals in the marine environment:

A state-of-the-science review. *Environ. Toxicol. Chem.* **35**, 1667–1676 (2016).

19. N. Hirt, M. Body-Malapel, Immunotoxicity and intestinal effects of nano-and microplastics: a review of the literature. *Part. Fibre Toxicol.* **17**, 1–22 (2020).

20. L. Rubio, R. Marcos, A. Hernández, Potential adverse health effects of ingested micro-and nanoplastics on humans. Lessons learned from in vivo and in vitro mammalian models. *J. Toxicol. Environ. Heal. Part B* **23**, 51–68 (2020).

21. C. M. Rochman, T. Hoellein, The global odyssey of plastic pollution. *Science (80-.).* **368**, 1184–1185 (2020).

22. M. S. Bank, S. V Hansson, The plastic cycle: A novel and holistic paradigm for the Anthropocene (2019).

23. A. A. Horton, S. J. Dixon, Microplastics: An introduction to environmental transport processes. *Wiley Interdiscip. Rev. Water* **5**, e1268 (2018).

24. E. Van Sebille, C. Spathi, A. Gilbert, The ocean plastic pollution challenge: towards solutions in the UK. *Grant. Brief. Pap* **19**, 1–16 (2016).

25. S. Allen, *et al.*, Atmospheric transport and deposition of microplastics in a remote mountain catchment. *Nat. Geosci.* **12**, 339 (2019).

26. R. Dris, *et al.*, A first overview of textile fibers, including microplastics, in indoor and outdoor environments. *Environ. Pollut.* **221**, 453–458 (2017).

27. S. L. Wright, J. Ulke, A. Font, K. L. A. Chan, F. J. Kelly, Atmospheric microplastic deposition in an urban environment and an evaluation of transport. *Environ. Int.* **136**, 105411 (2020).

28. L. Cai, *et al.*, Characteristic of microplastics in the atmospheric fallout from Dongguan city, China: preliminary research and first evidence. *Environ. Sci. Pollut. Res.* **24**, 24928–24935 (2017).

29. R. Dris, *et al.*, Microplastic contamination in an urban area: a case study in Greater Paris. *Environ. Chem.* **12**, 592–599 (2015).

30. J. H. Seinfeld, S. N. Pandis, *Atmospheric chemistry and physics: from air pollution to climate change* (John Wiley & Sons, 2016).

31. S. Allen, *et al.*, Examination of the ocean as a source for atmospheric microplastics. *PLoS One* **15**, e0232746 (2020).

32. G. C. Cornwell, *et al.*, Ejection of dust from the ocean as a potential source of marine ice nucleating particles. *J. Geophys. Res. Submitt.* (2020).

33. S. Wagner, *et al.*, Tire wear particles in the aquatic environment-a review on generation, analysis, occurrence, fate and effects. *Water Res.* **139**, 83–100 (2018).

34. F. Sommer, *et al.*, Tire abrasion as a major source of microplastics in the environment. *Aerosol Air Qual. Res.* **18**, 2014–2028 (2018).

35. C. Vogelsang, *et al.*, “Microplastics in road dust–characteristics, pathways and measures” (Norsk institutt for vannforskning, 2019).

36. N. Evangeliou, *et al.*, Atmospheric Transport, a Major Pathway of Microplastics to Remote Regions. *Nat. Geosci.* (2020).

37. S. I. Torri, R. S. Corrêa, G. Renella, L. Perelomov, A. Valdecantos, Biosolids soil application: agronomic and environmental implications 2013. *Appl. Environ. Soil Sci.* **2014** (2014).

38. L. Nizzetto, M. Futter, S. Langaas, Are agricultural soils dumps for microplastics of urban origin? (2016).

39. F. Murphy, C. Ewins, F. Carbonnier, B. Quinn, Wastewater treatment works (WwTW) as a source of microplastics in the aquatic environment. *Environ. Sci. Technol.* **50**, 5800–5808 (2016).

40. M. Lalitha, V. K. Thilagam, N. Balakrishnan, M. Mansour, Effect of plastic mulch on soil properties and crop growth-A review. *Agric. Rev.* **31**, 145–149 (2010).

41. R. B. Neale, *et al.*, The mean climate of the Community Atmosphere Model (CAM4) in forced SST and fully coupled experiments. *J. Clim.* **26**, 5150–5168 (2013).

42. A. Heymsfield, Ice crystal terminal velocities. *J. Atmos. Sci.* **29**, 1348–1357 (1972).

43. P. Ginoux, Effects of nonsphericity on mineral dust modeling. *J. Geophys. Res. Atmos.* **108** (2003).

44. N. Mahowald, *et al.*, Aerosol impacts on climate and biogeochemistry. *Annu. Rev. Environ. Resour.* **36** (2011).

45. A. Churg, M. Brauer, Human lung parenchyma retains PM2. 5. *Am. J. Respir. Crit. Care Med.* **155**, 2109–2111 (1997).

46. J. F. Kok, *et al.*, Smaller desert dust cooling effect estimated from analysis of dust size and abundance. *Nat. Geosci.* **10**, 274–278 (2017).

47. J. Neff, M. P. Reynolds, S. Munson, D. Fernandez, J. Belnap, The role of dust storms in total amtospheric particle concentration at two sites in the western U.S. *J. Geophys. Res.* **118**, 1–12 (2013).

48. K. Magnusson, *et al.*, Swedish sources and pathways for microplastics to the marine environment. *A Rev. Exist. data. IVL, C* **183** (2016).

49. Y. Shao, *et al.*, Dust cycle: An emerging core theme in Earth system science. *Aeolian Res.* **2**, 181–204 (2011).

50. E. D. Nilsson, *et al.*, Turbulent aerosol fluxes over the Arctic Ocean: 2. Wind-driven sources from the sea. *J. Geophys. Res. Atmos.* **106**, 32139–32154 (2001).

51. P. Ginoux, J. M. Prospero, T. E. Gill, N. C. Hsu, M. Zhao, Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products. *Rev. Geophys.* **50**, RG3005 (2012).

52. J. Crossman, R. R. Hurley, M. Futter, L. Nizzetto, Transfer and transport of microplastics from biosolids to agricultural soils and the wider environment. *Sci. Total Environ.*, 138334 (2020).

53. G. S. Zhang, Y. F. Liu, The distribution of microplastics in soil aggregate fractions in southwestern China. *Sci. Total Environ.* **642**, 12–20 (2018).

54. E. K. Liu, W. Q. He, C. R. Yan, 'White revolution' to 'white pollution'—agricultural plastic film mulch in China. *Environ. Res. Lett.* **9**, 91001 (2014).

55. F. Corradini, *et al.*, Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal. *Sci. Total Environ.* **671**, 411–420 (2019).

56. M. Rezaei, M. J. P. M. Riksen, E. Sirjani, A. Sameni, V. Geissen, Wind erosion as a driver for transport of light density microplastics. *Sci. Total Environ.* **669**, 273–281 (2019).

57. S. Hann, *et al.*, Investigating options for reducing releases in the aquatic environment of microplastics emitted by (but not intentionally added in) products. *Rep. DG ENV EC* (2018).

58. J. Gavigan, T. Kefela, I. Macadam-Somer, S. Suh, R. Geyer, Synthetic microfiber emissions to land rival those to waterbodies and are growing. *PLoS One* **15**, e0237839 (2020).

59. U. Pirc, M. Vidmar, A. Mozer, A. Kržan, Emissions of microplastic fibers from microfiber fleece during domestic washing. *Environ. Sci. Pollut. Res.* **23**, 22206–22211 (2016).

60. D. Kawecki, B. Nowack, Polymer-specific modeling of the environmental emissions of seven commodity plastics as macro-and microplastics. *Environ. Sci. Technol.* **53**, 9664–9676 (2019).

61. S. Dehghani, F. Moore, R. Akhbarizadeh, Microplastic pollution in deposited urban dust, Tehran metropolis, Iran. *Environ. Sci. Pollut. Res.* **24**, 20360–20371 (2017).

62. M. Klein, E. K. Fischer, Microplastic abundance in atmospheric deposition within the Metropolitan area of Hamburg, Germany. *Sci. Total Environ.* **685**, 96–103 (2019).

63. G. Suaria, *et al.*, Microfibers in oceanic surface waters: A global characterization. *Sci. Adv.* **6**, eaay8493 (2020).

64. A. Stohl, S. Eckhardt, C. Forster, P. James, N. Spichtinger, On the pathways and timescales of intercontinental air pollution transport. *J. Geophys. Res. Atmos.* **107**, ACH-6 (2002).

65. M. Prank, S. C. Kenaley, G. C. Bergstrom, M. Acevedo, N. M. Mahowald, Climate change impacts the spread potential of wheat stem rust, a significant crop disease. *Environ. Res. Lett.* **14**, 124053 (2019).

66. D. Griffin, C. Kellogg, E. Shinn, Dust in the Wind: Long Range Transport of Dust in the Atmosphere and Its Implications for Global Public and Ecosystem Health. *Glob. Chang. Hum. Heal.* **2**, 20–33 (2001).

67. J. C. Neff, *et al.*, Increasing eolian dust deposition in the western United States linked to

761 human activity. *Nat. Geosci.* **1**, 189–195 (2008).

762 68. I. Tegen, M. Werner, S. P. Harrison, K. E. Kohfeld, Relative importance of climate and
763 land use in determining present and future global soil dust emission. *Geophys. Res. Lett.*
764 **31** (2004).

765 69. N. M. Mahowald, G. D. R. Rivera, C. Luo, Comment on “Relative importance of climate
766 and land use in determining present and future global soil dust emission” by I. Tegen et al.
767 *Geophys. Res. Lett.* **31** (2004).

768 70. J. P. Field, *et al.*, The Ecology of Dust. *Front. Ecol. Environ.* **8**, 423–430 (2009).

769 71. M. Gidden, *et al.*, Global emissions pathways under different socioeconomic scenarios for
770 use in CMIP6: a dataset of harmonized emissions trajectories through the end of the
771 century. *Geosci. Model Dev. Discuss.* **12**, 1443–1475 (2019).

772 72. R. M. Hoesly, *et al.*, Historical (1750-2014) anthropogenic emissions of reactive gases and
773 aerosols from the Community Emission Data System (CEDS). *Geosci. Model Dev.* **11**,
774 369–408 (2018).

775 73. Z. Klimont, *et al.*, Global anthropogenic emissions of particulate matter including black
776 carbon. *Atmos. Chem. Phys. Discuss.* **17**, 8681–8723 (2017).

777 74. K. Riahi, *et al.*, The shared socioeconomic pathways and their energy, land use, and
778 greenhouse gas emissions implications: an overview. *Glob. Environ. Chang.* **42**, 153–168
779 (2017).

780 75. H. K. Webb, J. Arnott, R. J. Crawford, E. P. Ivanova, Plastic degradation and its
781 environmental implications with special reference to poly (ethylene terephthalate).
782 *Polymers (Basel).* **5**, 1–18 (2013).

783 76. R.-J. Müller, I. Kleeberg, W.-D. Deckwer, Biodegradation of polyesters containing aromatic
784 constituents. *J. Biotechnol.* **86**, 87–95 (2001).

785 77. S. L. Wright, F. J. Kelly, Plastic and human health: a micro issue? *Environ. Sci. Technol.*
786 **51**, 6634–6647 (2017).

787 78. M. C. Rillig, Plastic and plants. *Nat. Sustain.*, 1–2 (2020).

788 79. L. Li, *et al.*, Effective uptake of submicrometre plastics by crop plants via a crack-entry
789 mode. *Nat. Sustain.*, 1–9 (2020).

790 80. P. Sherman, E. Van Sebille, Modeling marine surface microplastic transport to assess
791 optimal removal locations. *Environ. Res. Lett.* **11**, 14006 (2016).

792 81. J. Brahney, *et al.*, A new sampler for the collection and retrieval of dry dust deposition.
793 *Aeolian Res.* **45**, 100600 (2020).

794 82. J. W. Hurrell, *et al.*, The community earth system model: a framework for collaborative
795 research. *Bull. Am. Meteorol. Soc.* **94**, 1339–1360 (2013).

796 83. R. Gelaro, *et al.*, The modern-era retrospective analysis for research and applications,
797 version 2 (MERRA-2). *J. Clim.* **30**, 5419–5454 (2017).

84. X. Liu, *et al.*, Towards a minimal representation of aerosol direct and indirect effects: Model description and evaluation. *Geosci. Model Dev* **5**, 709–735 (2012).

85. S. Albani, *et al.*, Improved dust representation in the Community Atmosphere Model. *J. Adv. Model. Earth Syst.* **6**, 541–570 (2014).

86. H. Matsui, N. Mahowald, Development of a global aerosol model using a two-dimensional sectional method: 2. Evaluation and sensitivity simulations. *J. Adv. Model. Earth Syst.* **9**, 1887–1920 (2017).

87. N. Mahowald, *et al.*, The size distribution of desert dust aerosols and its impact on the Earth system. *Aeolian Res.* **15**, 53–71 (2014).

88. C. L. Ryder, *et al.*, Coarse and giant particles are ubiquitous in saharan dust export regions and are radiatively significant over the sahara. *Atmos. Chem. Phys.* **19**, 15353–15376 (2019).

89. J. E. Kay, *et al.*, The Community Earth System Model (CESM) large ensemble project: A community resource for studying climate change in the presence of internal climate variability. *Bull. Am. Meteorol. Soc.* **96**, 1333–1349 (2015).

90. A. Cózar, *et al.*, Plastic debris in the open ocean. *Proc. Natl. Acad. Sci.* **111**, 10239–10244 (2014).

91. E. Van Sebille, *et al.*, A global inventory of small floating plastic debris. *Environ. Res. Lett.* **10** (2015).

92. E. Van Sebille, M. H. England, G. Froyland, Origin, dynamics and evolution of ocean garbage patches from observed surface drifters. *Environ. Res. Lett.* **7** (2012).

93. L. C. M. Lebreton, S. D. Greer, J. C. Borrero, Numerical modelling of floating debris in the world's oceans. *Mar. Pollut. Bull.* **64**, 653–661 (2012).

94. N. Maximenko, J. Hafner, P. Niiler, Pathways of marine debris derived from trajectories of Lagrangian drifters. *Mar. Pollut. Bull.* **65**, 51–62 (2012).

95. P. J. Lawrence, *et al.*, Simulating the biogeochemical and biogeophysical impacts of transient land cover change and wood harvest in the Community Climate System Model (CCSM4) from 1850 to 2100. *J. Clim.* **25**, 3071–3095 (2012).

96. L. Warszawski, *et al.*, Center for International Earth Science Information Network—CIESIN—Columbia University.(2016). Gridded population of the World, Version 4 (GPWv4): Population density. Palisades. NY: NASA Socioeconomic Data and Applications Center (SEDAC). doi: 10. 7927/H4N. *Atlas Environ. Risks Facing China Under Clim. Chang.*, 228 (2017).

97. W. H. Press, S. A. Teukolsky, W. T. Vetterling, B. P. Flannery, *Numerical recipes 3rd edition: The art of scientific computing* (Cambridge university press, 2007).

98. N. M. Mahowald, *et al.*, Atmospheric global dust cycle and iron inputs to the ocean. *Global Biogeochem. Cycles* **19** (2005).

99. C. and I. S. Laboratory, Cheyenne: HPE/SGI ICE XA System (University Community

836 Computing) (2017).
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839 **Figure Legends**

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841 **Figure 1.** Representation of the major sources of microplastics to the atmosphere and their
842 relative contributions to deposition to the terrestrial environment over the western U.S. (30°-50°N,
843 120 to 100°W). Over this region the deposition of microplastics is 84% from roads, 11% from sea
844 spray, 5% from agricultural dust, and 0.4% from dust near population centers. The atmospheric
845 burden above this region is 1Gg (0.001Mt).

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847

848 **Figure 2. (**a) The fractional mass by size of the particles (green x's) and fibers in the observations
849 (black triangles). In the model the particles are assumed to be transported as observed, but
850 because the fibers are mostly 1 μm in diameter but much longer, the aerodynamic size that is
851 appropriate is not well established, and thus are simulated in three sizes: small (red), medium
852 (cyan) and big (dark blue), with medium being the base case. (b) Estimates of the contribution at
853 the observing sites from different sources as inferred using the method described in the Methods
854 section for the three sizes: big (diamond), medium (triangle), and small (square). For all sources
855 and cases, the 95% confidence limits are shown as vertical lines. Multiple sensitivity studies were
856 conducted (see Methods for details), with time averaging (cyan), and with each site withheld
857 (green), showing the ranges of values that can be obtained for each relative source strength.

858

859 **Figure 3:** Model estimates of microplastic deposition using the best estimate ($\mu g/m^2$ /day[1]). (a)
860 Total plastic deposition from all the different model sources and the annual average of the
861 observations at the sites (filled circles). (b) Scatterplot of the model versus observations for wet
862 (triangles) and dry (squares) deposition (r=0.11, n=313). Station names are abbreviated from the
863 NADP network, following SI Appendix, Table S1, and the latitude and longitude are as follows:
864 AZ03 (36.1°N 247.8°E), CA67 (34.1°N 243.6°E), CO02 (40.1°N 254.4°E), CO10 (39.0°N
865 253.0°E), CO98 (40.3°N 254.4°E), ID03 (43.5°N 246.5°E), NV05 (39.0°N 245.8°E), UT09
866 (38.46°N 250.2°E), UT95 (40.8°N 250.5°E), UT99 (37.6°N 247.8°E), WY06 (42.9°N 250.2°E).
867 The total microplastic deposition (a) is the sum of the (c) road tire and braking, (d) ocean, (e)
868 agricultural dust and (f) population dust, which is the dust generated downstream from population
869 centers. Notice that the population source is not plotted as it is zero in the best estimate.
870

Figure 4. Globally averaged sources of microplastics (a) as inferred using the methods described in the Methods section for the three size ranges: big (diamond), medium (triangle), and small (square). For all sources, the 95% confidence limits are shown as vertical lines. Multiple sensitivity studies were conducted (see Methods for details), with time averaging (cyan), and with each site withheld (green), showing the ranges of values that can be obtained for each source strength extrapolated globally. (b) Total microplastic deposition estimated in the model ($\mu g/m^2/day$). (c) Modeled budgets for different continents in terms of sources and deposition (excluding coastal grid boxes). Roads (brown), Ocean (blue), Agricultural Dust (white) and Population Dust (yellow) contributions in Gg/yr are shown for each continent using the medium size (base case).