1 Modeling present and future plastics dispersal in the global environment and recommendations for 2 cleanup scenarios

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1718 List of Abbreviations

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- 20 Tg, teragrams
- 21 P, macroplastics
- 22 LMP, large microplastics
- 23 SMP, small microplastics
- 24 y, year
- 25 F, flux (in Tg y^{-1})
- 26 M, mass (in Tg)
- 27 1σ , one (sigma) standard deviation
- 28 BAU, business as usual
- 29 SCS, systems change scenario

31 Abstract

Since 1950 humans have introduced 8300 teragrams (Tg, 10¹² grams, millions of metric tons) of plastic 32 polymers into the Earth's surface environment. Accounting for the dispersal and fate of produced plastics and 33 fragmented microplastics in the environment has been challenging. Recent studies have fueled debate of the 34 global river budget for plastic transport to oceans, the sinking and beaching of marine plastics and the emission 35 and deposition of atmospheric microplastics. Here we define a global plastics cycle and budget, and develop 36 a box model of plastic cycling, including the fragmentation and transport of large and small microplastics 37 (LMP, SMP) within coupled terrestrial, oceanic and atmospheric reservoirs. We force the model with 38 historical plastics production and waste data, and explore how macroplastics, LMP and SMP propagate 39 through the reservoirs from 1950 to 2015 and beyond. We find that considerable amounts of plastics reside 40 most likely in the deep ocean (82 Tg), in shelf sediments (116 Tg), on beaches (1.8 Tg) and, as a result of 41 marine emissions, in the remote terrestrial surface pool (28 Tg). Business as usual or maximum feasible 42 reduction and discard scenarios show similar, 4-fold increases in atmospheric and aquatic ecosystem SMP 43 exposure by 2050, because future plastics mobilization is controlled by releases from the large terrestrial 44 discarded plastics reservoir (3500 Tg). Zero-release from 2025 onwards illustrates recovery of P and LMP 45 46 reservoirs on centennial time scales, while SMP continue to cycle in air, soil, and surface ocean for millennia. Limiting dramatic future dispersal of plastics requires, in addition to reducing use and waste, remediation of 47 48 the large terrestrial legacy plastics pool.

50 Introduction

- 51 A characteristic feature of the Anthropocene is the widespread dispersal of plastic polymers across Earth's
- 52 surface since the 1950s (1). Of the 1.5 trillion barrels of oil (200,000 Tg) produced since the 1950s (2) about

4% (8300 Tg) has been transformed into non-biodegradable polymers, and used in predominantly single-use 53 packaging or short-lived (1-25y) technological applications (3). Produced plastics have been abundantly 54 (60%) discarded into the technosphere, the part of the environment that has been made or modified by humans: 55 urban, sub-urban, agricultural, and industrial areas, including landfills (3,4). The discarded pool of plastics 56 has been slowly mobilized by wind, runoff, rivers and ocean currents to all remote corners of planet Earth, 57 including the poles and the deep ocean (5-8). Large plastic debris tend to fragment to micro- and nano-sized 58 particles, which due to their increased surface area can absorb, adsorb or release a range of secondary natural 59 and man-made chemical compounds in the environment (9). Assessing the possible impact of plastics on 60 ecosystem and human health, and mitigating this impact, requires a solid understanding of where and when 61 62 discarded plastics end-up, and to which size range they evolve.

Over the past decades important efforts have been made to chart the abundance, size properties, and 63 bulk polymer composition of plastics in the surface ocean, soils, rivers, wetlands, biota and atmosphere. A 64 65 perceived mismatch between the relatively small quantity of plastics in the surface ocean (0.3 Tg) (10) and the large quantity delivered by global rivers $(4.8 - 12.7 \text{ Tg y}^{-1})$ (11) has fueled a 'missing marine plastics' 66 paradox (12). Solutions to this issue have been proposed in the transfer of marine plastics to the deep ocean 67 (13,14), to coastal environments, via beaching (15,16), to the subsurface ocean and marine sediments by 68 sinking (17,18), and to marine emission of microplastics to the atmosphere (19,20). Recently, a 1000-fold 69 lower global river flux of 0.0064 Tg y⁻¹ was suggested, based on alternative plastic size distribution 70 assumptions (21). Such a low river flux would imply a marine residence time of several years, and possibly 71 removes the need for a missing marine plastics sink. In parallel to rivers and ocean currents, the atmosphere 72 has been identified as a global vector of MP, in both urban (22,23) and remote environments (5,24,25), 73 including MP emission from land (19,26) and sea (19,20). In this study we use the best available estimates of 74 both plastics abundance and fluxes to construct a global plastics mass budget. This budget is implemented in 75 a global box model of plastics cycling between land, atmosphere and ocean from 1950 to 2015. We then use 76 the model to explore how plastics disperse through Earth's surface environment over times scale ranging from 77 78 decades, focusing on policy scenarios, to millennia, addressing the fate and potential burden of global plastics 79 contamination.

81 Plastics cycling box model

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In order to construct a global plastics mass and mass transfer budget (Figure 1), we use plastics observations 82 from the literature and a box modeling approach (see Methods for details). We subdivide macroplastics 83 (P, >5mm), large microplastics (LMP, >0.3mm and <5mm) and small microplastics (SMP, <0.3mm), and 84 define 'MP' as the sum of LMP plus SMP. The model is a coupled 15-reservoir numerical box model that 85 simulates how produced P and MP propagate through the terrestrial, marine, and atmospheric environments 86 upon release or emission. P fragment to LMP, and LMP fragment to SMP, and only SMP become airborne, 87 emitted from and deposited to oceans and land. Terminal P, LMP and SMP sinks are marine sediments, 88 whereas remote terrestrial (soils, barren rock, ice sheets) and deep ocean pools act as long-term temporary 89 reservoirs. The mass flux, F_{ab} (Tg y⁻¹) between two reservoirs a and b is $F_{ab} = k_{ab} \times M_a$, where M_a is the mass 90 91 of plastics in reservoir a (Tg), and k_{ab} is a first-order mass transfer (rate) coefficient (y⁻¹). In a first step, all k values are determined from published, recent, 2005-2022 observations (see Methods) and from model 92 estimates of atmospheric SMP fluxes (19). The model is then run from 1950 to 2015, with only the k_{ab} transfer 93 coefficients and plastics production and waste generation as external forcing. In the following we discuss 94 whether the simulated modern plastics distribution for 2015 corresponds to observations, which k values (and 95 therefore fluxes) need to be adjusted, and what the model implications are for our understanding of plastic 96 cycling. With the addition of atmospheric transport of plastics, the term 'emission' refers here exclusively to 97 the suspension of terrestrial and marine SMP in air. 'Release' is used as the generic term for plastics discharge 98 and mobilization to the technosphere and in-land aquatic and marine environments. Conversion of plastics 99 number concentration to mass concentration is detailed in the Methods. All uncertainties reported are 1σ 100 standard deviation (or 16th and 84th percentiles, corresponding to a 1 σ uncertainty; see Methods). 101

We start by detailing the 'base case' plastics cycling model, based on best known modern observations of reservoir sizes and fluxes between reservoirs (see Methods for details). We include plastics production (8300 Tg since 1950), waste generation and waste disposal from Geyer et al. (3) who estimated 2600 Tg of

plastics to be in use in 2015, 4900 Tg discarded (split into 4200 Tg of P, and 700 of primary LMP following 105 Lau et al. (4) and 800 Tg incinerated. In the base case we use the mid-point of the river plastics flux estimate 106 by Jambeck et al., of 8.8 Tg $y^{-1}(11)$, containing equal fractions of P and LMP. We adopt surface ocean mixed 107 layer buoyant P and LMP inventories of 0.23 and 0.04 Tg (10), and a surface mixed layer SMP inventory of 108 0.003 Tg (27). We make an order of magnitude estimate of beached LMP of 0.5 ± 0.4 Tg, based on the global 109 surface of sandy beaches (2.63 10⁵ km²; (28)), a median global beach sand LMP abundance of 2450 MP km⁻ 110 2 , and median beached LMP size of 2.0 mm (29). We estimate beached P, and shelf sediment P pools from a 111 review study (30) that estimates mean beached P and sea floor P concentrations of 2 and 5 Mg km⁻² 112 respectively (uncertainty not estimated). Multiplying by beach and continental shelf surfaces of 2.63 10⁵ and 113 $2.89 \ 10^7 \ \text{km}^2$ results in beached and shelf sediment P pools of 1.3 and 51 Tg. An estimate for the global deep 114 ocean sediment MP pool of 1.5 Tg is based on observed mean deep sediment MP concentrations of 0.72 MP 115 g^{-1} (see Methods) (31). A shelf sediment MP pool of 65 Tg (1 σ , 21 to 78Tg) is estimated from subtidal 116 sediment median MP concentrations of 100 MP kg⁻¹ (see Methods) (29). Rate coefficients for P and LMP 117 beaching (the transfer from ocean to beach), k_{beaching} of 0.15 y⁻¹ are approximated based on Onink et al. (15). 118 Surface mixed layer to deep subsurface ocean sinking rates of P, LMP, SMP lack in situ observations; we 119 estimate model sinking rate coefficients, k_{P,sinking} of 1367 y⁻¹, k_{LMP,sinking} of 196 y⁻¹ and k_{SMP,sinking} of 33 y⁻¹ for 120 the 100 m deep surface ocean mixed layer, based on the empirical results of a sinking tank study of mixed 121 phytoplankton aggregates with MP(17). We include the sinking and sedimentation of non-buoyant P over the 122 shelf, but not from open ocean waters, assuming that only buoyant P dominate open ocean P. Macroplastics, 123 P, are beached as described above, and fragmented in surface ocean waters to LMP at a rate $k_{oceP \rightarrow LMP}$ of 0.03 124 y^{-1} (16), supported by observations (32). A recent review of plastics degradation rates highlights the 125 complexity and variability of plastics degradation rates as a function of polymer type, sunlight, and physical 126 environment (33). The authors use an observed median HDPE degradation rate of 4.3 μ m y⁻¹ in the marine 127 environment, and a theoretical degradation framework to illustrate how a typical HDEP bag (film), fiber (2 128 mm diameter, 230mm long) or bead (8.8 mm diameter) would degrade at relative mass loss rates of 0.5, 0.005 129 and 0.0014 y⁻¹. The rate of 0.03 y⁻¹ (1 σ uncertainty: 0.006 to 0.06 y⁻¹) we adopt lies within this estimated 130 variability. We consider that for the purpose of our study, it is too early at present to try and incorporate more 131 detailed plastics fragmentation or degradation parameterizations. We agree with Chamas et al. (33) that more 132 robust degradation observations are needed, and we suggest that a follow-up box model that incorporates 133 variable polymer types would be a more appropriate occasion. In the absence of fragmentation rates for LMP 134 to SMP in surface, subsurface waters, beach zone, and discarded pool, and for P to LMP in subsurface water, 135 beach zone and discarded pool we adopt, in the base case, the same rate $k_{oceLMP \rightarrow SMP}$ of 0.03 v⁻¹ for all these 136 fragmentation sites. 137

The subsurface ocean pool of LMP and SMP, below the surface mixed layer, is of importance to complete the marine plastics budget and to parameterize model settling and sedimentation of plastics. Table 1 and Figure 2 summarize recent observations of subsurface marine MP. We estimate a global deep ocean MP inventory of 82 ± 47 Tg based on mean N-Pacific pelagic concentrations of $131 \pm 44 \ \mu g \ m^{-3}$ (6,34), mean N and S-Atlantic concentrations of $91 \pm 46 \ \mu g \ m^{-3}$ (35–37), and extrapolated estimates for the Indian, Southern, and S-Pacific Oceans (Table 2, Methods).

Recent studies on atmospheric MP cycling show fragment and fiber size distributions to be in the SMP 144 range <300 µm. While LMP emission and deposition occurs, these tend to deposit more rapidly back to the 145 same reservoir (e.g., marine emission followed by marine deposition) and are therefore ignored in the box 146 model. Table 3 summarizes SMP observations in the boundary layer and free troposphere, yielding a total 147 tropospheric SMP mass of 0.031 ± 0.027 Tg. This observed stock is 10x higher, though within uncertainty, of 148 a model estimate of 0.0036 Tg (19). Our estimate of 0.031 Tg is very sensitive to the assumed median SMP 149 size of 70 µm, which is where the atmospheric SMP model allocates most SMP mass(19). We adopt global 150 SMP emissions from the same model study (19): emissions from roads, 0.01 Tg y^{-1} , agricultural dust, 0.07 Tg 151 y⁻¹, population dust, 0.02 Tg y⁻¹, and oceans, 8.6 Tg y⁻¹. We use SMP deposition observations over land 152 (5,24,38) in combination with population density data for 2015 (39) to estimate global SMP deposition over 153 land of 1.1 ± 0.3 Tg y⁻¹ and an accumulated remote terrestrial SMP pool of 28 ± 10 Tg (see Methods). We 154 assume that global SMP emissions (8.6 Tg y⁻¹; (19)) equal deposition, and estimate SMP deposition over 155 oceans to be 7.6 Tg v^{-1} . 156

158 Results and Discussion

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160 The box model base case is run from 1950 to 2015 and results, in terms of plastics reservoir sizes and fluxes for the year 2015, are shown in Table 4 in comparison to the above-mentioned observations. The base case 161 reproduces observed amounts of in-use P, discarded P, LMP, SMP and terrestrial SMP to within 40%. Using 162 the upper bound river plastics flux of 13 Tg y⁻¹ (11) the base case also reproduces well the observed 163 downstream plastics mass in marine and remote terrestrial systems (surface and deep ocean, sediments, beach, 164 remote terrestrial surfaces) of 201 Tg (10, 120 to 630 Tg). Remote terrestrial surfaces are included in the 165 downstream environment, because its accumulated SMP mass is for 96% derived from the SMP river flux, 166 surface ocean LMP degradation, and the important marine emission of SMP to the atmosphere where it leads 167 to global dispersal and deposition to remote terrestrial surfaces (soil, rock, ice). We note that using the 2000-168 fold lower river plastics flux of 0.0064 Tg y⁻¹ by Weiss et al. (21) would lead to large low bias in the marine 169 and remote terrestrial reservoirs. A model river plastics flux of 13 Tg y⁻¹ (1 σ , 9 to 51 Tg) balances the overall 170 marine plastics budget, and gives satisfactory (within a factor 10x) reproduction of surface ocean P, LMP and 171 SMP, shelf sediment P, LMP and SMP, and beached LMP reservoirs. Within the marine system, the modeled 172 deep sediment MP pool is however biased high 90-fold, and beached P biased low 26-fold. We therefore 173 optimize and lower subsurface ocean specific $k_{LMP,sinking}$ from 4.9 y⁻¹ to 0.0012 y⁻¹ and $k_{SMP,sinking}$ from 0.8 y⁻¹ 174 to 0.0002 y⁻¹, and increase k_{beaching} from 0.15 y⁻¹ to 4.0 y⁻¹. We argue that the base case sinking rates and k 175 estimates for experimental biofouled LMP and SMP are inappropriate for deep ocean sedimentation because 176 remineralization of biofilm during sinking increases buoyancy, halts sinking and lowers the effective sinking 177 rate (14). The base case k_{beaching} was derived for the coastal ocean (15), which we do not explicitly separate 178 and simulate here, likely leading to its underestimation relative to whole surface ocean P cycling. It is 179 important to note that out of 23 mass transfer coefficients (k's) only 3 needed fitting. This indicates that current 180 understanding of P, LMP and SMP stocks and fluxes, that determine k's, is sufficiently accurate to formulate 181 182 and use the box model.

Figure 1 presents our best estimate of the global plastics cycle for the year 2015, based on observed inventories and fluxes (black), modeled inventories and fluxes (red), and a river plastics flux of 13 Tg y⁻¹ to the ocean (see Table 4 for uncertainties). Key properties of the global plastics cycle are:

The large mass, 1200 Tg of discarded LMP (of which 840 Tg primary LMP) and on the order of 500 Tg of
 discarded SMP in the technosphere, which are potentially mobilizable to wetlands, oceans, groundwater,
 atmosphere and remote terrestrial surfaces.

2. The substantial mass of plastics, 201 Tg, representing 3% of all plastics produced since 1950, that has been
released from the technosphere to pristine terrestrial and marine ecosystems.

3. The 65-fold larger river plastics flux (13 Tg y⁻¹) compared to the total terrestrial atmospheric SMP emission flux (0.2 Tg y⁻¹).

4. The importance of marine SMP emissions on further distributing microplastics to remote ocean waters and
to remote terrestrial surfaces (96% of the 28 Tg on remote land originates from marine emissions, and only
4% from terrestrial emissions).

5. The potentially large subsurface oceanic LMP and SMP (82 ± 47 Tg), and shelf sediment P and LMP (116 Tg) reservoirs, compared to beached P and LMP (1.8 Tg), and compared to surface ocean plastics (0.27 Tg).

The uncertainties associated with the global plastics cycle (Table 4) are large, due to an overall lack of observations and underlying plastics quantification challenges. In particular, observations of SMP number and mass in the terrestrial discarded and remote terrestrial pools, and in terrestrial and marine emissions and deposition are needed.

We use the box model to simulate and illustrate at what timescales P, LMP and SMP propagate through Earth surface reservoirs if we were to halt plastics production and waste generation in 2025. Figure 3 shows P, LMP and SMP dispersal from 1950 to the year 3000: The discarded terrestrial P pool decreases rapidly, by 90% in 2100, due to fragmentation to LMP, which in turn decreases by 90% in 2150 due to further fragmentation to SMP. LMP and SMP transport by rivers and air leads to rapid increases of LMP and SMP in the marine pools and of SMP in the remote terrestrial pool. The discarded SMP pool takes longer, 90% by 2500, to mobilize to the surface ocean, and from there via marine emission back to the remote terrestrial pool. The cyclical behavior that develops, cycles SMP for millennia back and forth between surface ocean and continents, before gradual escape to the deep ocean marine sediments (Figure S2). This scenario illustrates that even if we would entirely replace plastics by alternative materials, the legacy of historical plastics mismanagement could result in prolonged plastics dispersal over centuries (LMP) or millennia (SMP), unless we better manage present and future discarded plastics pools on land.

Next, we explore in detail how two production and waste management scenarios affect plastics cycling 214 over the period 2015 to 2050: 1. The business as usual (BAU) scenario from Geyer et al. (3) reaching 30,000 215 Tg of produced plastics in 2050, and with discard below 10% and recycling and incineration of 43% and 49% 216 in 2050, 2. The systems change scenario (SCS) from Lau et al. (4) which proposes ambitious, but realistic 217 measures to reduce, substitute, recycle, and dispose of plastics (see Methods for details, and SI). Figure 4A 218 illustrates how a 2-fold drop in plastics production from 550 Tg y⁻¹ to 250 Tg y⁻¹ in 2040 under the SCS 219 scenario significantly limits further plastics accumulation in the technosphere compared to BAU. Yet, despite 220 the projected strong decrease of mismanaged waste, and increase in safe disposal and recycling, the SCS does 221 not lead to measurable changes in key metrics, such as beached P, total river plastics flux (P+LMP+SMP) or 222 atmospheric SMP deposition to remote terrestrial surfaces by 2050 (Figure 4 B, C, D). The reason for this is 223 the persistent mobility of legacy plastic waste in the large terrestrial discarded P, LMP and SMP reservoirs. 224 To render SCS policy effective, it will have to be supported by immobilization or remediation of the terrestrial 225 discarded plastics pool. We explore the potential impact of remediation of the discarded P pool from 2025 226 onwards at a rate of 3% P isolation and safe disposal per year (Figure 4 B, C, D). Discarded P remediation 227 halts beached P dispersal by 2040, curbs total river plastics discharge to some extent but does not impact 228 atmospheric SMP deposition to land. Although technically more challenging, remediation of discarded LMP 229 and SMP pools at an identical 3% per year rate is needed to also inverse dispersal of river and atmospheric 230 plastics (Figure 4 B, C, D) and to truly limit future planetary dispersal of plastics. The fragmentation of SMP 231 to nanoplastics and ultimately to dissolved and colloidal polymers needs further study, in particular their rates 232 of production, before they can be included in the dispersion box model. 233

235 Conclusions

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In this study we define a global plastics cycling budget for the year 2015, and develop a box model of plastic 236 cycling, including the transport and fragmentation of macroplastics (P) to large (LMP) and to small 237 microplastics (SMP) within coupled terrestrial, oceanic and atmospheric reservoirs. We drive the model with 238 historical plastics production and waste data, and investigate how macroplastics (P), LMP and SMP propagate 239 through Earth surface reservoirs from 1950 to 2015 and beyond, to 2050 and to the year 3000. Based on 240 published plastics observations we estimate that important amounts of plastics are present in the deep ocean 241 (82 Tg), in shelf sediments (116 Tg), on beaches (1.8 Tg) and in the remote terrestrial surface pool (28 Tg). 242 243 The box model suggests that plastics in the remote terrestrial surface pool originate predominantly from marine SMP emissions that are transported via the atmosphere and deposited over land. Simulated zero-release 244 of plastics to land, water and air from 2025 onwards illustrates how P and LMP reservoirs recover on 245 centennial time scales, while SMP continue to cycle in air, soil, and surface ocean for millennia. Business as 246 usual or maximum feasible reduction and discard scenarios show similar, 4-fold increases in atmospheric and 247 aquatic ecosystem SMP exposure by 2050, because future plastics mobilization is controlled by releases from 248 the large terrestrial discarded plastics reservoir. We conclude that in order to limit future dispersal of plastics 249 we should, in addition to reducing plastics use and waste, anticipate remediation of the large terrestrial legacy 250 plastics pool. 251

252253 Methods

Definitions of plastics size categories are continuously debated; here we use operational definitions of macroplastics (P, >5mm), large microplastics (LMP, >0.3mm and <5mm) and small microplastics (SMP, <0.3mm). The 0.3mm distinction is based on the frequently used plankton net mesh size of approximately 0.3 mm. The 0.3mm cut-off is also a reasonable starting point for the simulation of atmospheric cycling of SMP, with nearly all remote airborn SMP particles, films and 50% of fibers falling in the 1-300 μ m range (5,24). All P, LMP, SMP reservoir sizes (i.e., inventory) and fluxes are expressed in teragrams 260 (Tg = 10^{12} grams) and Tg y⁻¹. For some reservoirs, studies do not discern LMP or SMP, in which case we 261 retain the generic 'MP' abbreviation.

LMP and SMP observations are typically expressed as MP counts per unit volume or per unit area. To estimate mass concentrations, we use, whenever reported, the full MP size distribution reported, a uniform density of 1 x $10^{-6} \mu g \mu m^{-3}$ (40), and the MP volume approximation, V = L³ x 0.1, where L are the reported length values of the size distribution.

We use global plastics production, 8300 Tg (teragrams or millions of metric tons), and waste 266 generation (discarded, recycled or incinerated) from Geyer et al. (3). Produced plastics enter the 'in-use' pool, 267 where they are mostly discarded within a single year due to the dominant use of single-use packaging. In 2015, 268 55% of non-fiber plastics are still discarded within a year, 25% incinerated and 20% recycled (3). We assume 269 fiber plastics to undergo similar relative discarding and incineration fates, leading to a 'discarded P+MP' 270 reservoir of 4900 Tg, an incinerated pool of 800 Tg (atmospheric CO₂) and an in-use pool of 2600 Tg in 2015 271 as described by Geyer et al.(3). Lau et al. (4) estimated the proportion of municipal solid waste that enters 272 aquatic and terrestrial environments as primary LMP to be 14 ± 4 % in 2016, which we apply here to all 273 discarded plastics (4). We therefore apply a primary f_{LMP} fraction of 0.14 and primary f_P fraction of 0.86 to 274 estimate transfer from the in-use to discarded reservoir for the period 2050-2015. The following mass balance 275 equations are defined for in-use and discarded pools: 276

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278
$$\frac{d(P_{use})}{dt} = P_{prod} - f_{disc} \times P_{waste} - f_{inc} \times P_{waste} \quad (\text{Eq.1})$$
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280 Where P_{use} is the mass of total plastic (P + LMP) in use, P_{prod} the mass of total plastics produced (Tg y⁻¹), 281 P_{waste} the mass of total plastic waste, and f_{disc} , and f_{inc} are the fractions of P_{use} that are discarded, incinerated 282 and recycled.

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$$\frac{d(P_{disc})}{dt} = f_{disc} \times P_{waste} \times f_P - k_{P-river} \times P_{disc} - k_{discP \to LMP} \times P_{disc} \quad (Eq.2)$$
285

286 Where P_{disc} is the mass of P discarded, f_P is the fraction of total plastic waste that are macroplastics, $k_{P-river}$ is 287 the transfer coefficient for P to the ocean, via river runoff.

289
$$\frac{d(LMP_{disc})}{dt} = f_{disc} \times P_{waste} \times f_{LMP} + k_{discP \to LMP} \times P_{disc} - k_{LMP-river} \times LMP_{disc} - k_{discLMP \to SMP} \times 290 \quad LMP_{disc} \text{ (Eq.3)}$$

Where LMP_{disc} is the mass of LMP discarded, f_{LMP} is the fraction of total plastics waste that are primary microplastics (pellets, synthetic textiles, personal care products, etc), $k_{LMP-river}$ is the transfer coefficient for LMP to the ocean, via river runoff, and $k_{LMP\rightarrow SMP}$ is the transfer coefficient for LMP degradation to SMP within the terrestrial 'discarded' pool.

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$$\frac{d(SMP_{disc})}{dt} = k_{discLMP \to SMP} \times SMP_{disc} - k_{SMP-river} \times SMP_{disc} - k_{disc-atm} \times SMP_{disc}$$
(Eq.4)
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Where SMP_{disc} is the mass of SMP discarded, $k_{SMP-river}$ is the transfer coefficient for SMP to the ocean, via river runoff, and $k_{SMP-atm}$ is the transfer coefficient for SMP emission to the atmosphere from the terrestrial 'discarded' pool, including tire wear particles (TWP).

Transfer coefficients k_{P-river}, k_{LMP-river}, and k_{SMP-river} are calculated from 2015 plastic fluxes and inventories, e.g. k_{P-river}=P_{disc}/F_{P-river} where F stands for flux (Table S1). The mid-point estimate for F_{P-river} of 8.8 Tg y⁻¹ ((11,41)) is used here, and subdivided into 50% P and 50% LMP (21). The 'discarded pool to atmosphere' transfer coefficient, k_{disc-atm}, which theoretically equals SMP_{disc}/F_{SMP_disc-atm} is unconstrained, because the SMP_{disc} pool size, in Tg, is unknown (F_{SMP_disc-atm} is 0.18 Tg y⁻¹, based on Brahney et al. (19), and was therefore fitted at 0.00037 y⁻¹ as described in the text.

The global ocean. Two previous box models have examined the plastics budget of the marine environment 309 (13,16). In addition, a number of Lagrangian oceanic or atmospheric transport models have provided insight 310 in marine plastics dispersal and surface ocean plastics mass balance (15,19,42). Koelmans et al. (13) used a 311 plastics mass budget for the surface ocean to fit a marine P to LMP fragmentation rate, and a LMP 312 sedimentation rate, under the assumption of 100% buoyant P (no settling to deep waters). To accommodate 313 the high river plastic inputs, rapid plastic fragmentation to LMP (>90% per year), and rapid LMP settling rates 314 were fitted, and suggested a short plastics and LMP residence time for the surface ocean (<3 yrs). Subsequent 315 modeling work has investigated P and LMP beaching, resuspension in coastal waters (15,16), marine SMP 316 emissions(19), and P sedimentation due to loss of buyuancy(16). Lebreton et al. (16), in their marine box 317 model study(16), argued that observations of old plastics in the surface ocean disagree with rapid 318 fragmentation and settling and fitted a plastics to LMP degradation rate of 3% per year, which we adopt here 319 for the surface mixed layer ($k_{Poce \rightarrow LMP} = 0.03 \text{ y}^{-1}$). 320

Lebreton et al. (16) fitted important beaching of coastal plastics (97% per year). In the absence of a 321 robust estimate for global beached macroplastics (43), Onink et al. (15) recently analyzed model beaching and 322 resuspension scenarios finding at least 77% of net beaching for positively buoyant plastic debris over 5 years 323 (15), which we adopt here in the base case as $k_{P,beaching} = 0.15 \text{ y}^{-1}$. Surface ocean P, LMP, and SMP equations 324 are: 325

 $\frac{d(P_{surf-oce})}{dt} = k_{P-river} \times P_{disc} - k_{Psurf-oce-beach} \times P_{oce} - k_{Psurf-oce \rightarrow LMP} \times P_{surf-oce} - k_{Psurf-oce \rightarrow sed} \times P_{surf-oce} \times f_{shelf} \text{ (Eq.5)}$ 328 329

 $\frac{d(LMP_{surf-oce})}{dt} = k_{LMP-river} \times LMP_{disc} + k_{Psurf-oce \rightarrow LMP} \times P_{oce} - k_{LMPsurf-oce \rightarrow beach} \times LMP_{surf-oce} - k_{LMPsurf-oce \rightarrow beach} \times LMP_{surf-oce \rightarrow beach}$ 330 $k_{LMPsurf-oce \rightarrow shelfsed} \times LMP_{surf-oce} \times f_{shelf} - k_{LMP-sink} \times LMP_{surf-oce} \times f_{pelagic} - k_{LMP-sink} \times LMP_{surf-oce} \times f_{pelagic}$ 331 332 $k_{LMPsurf-oce \rightarrow SMP} \times LMP_{surf-oce}$ (Eq.6) 333 d(CMD

$$334 \quad \frac{a(SMP_{surf-oce})}{dt} = k_{SMP-river} \times SMP_{disc} + k_{atm \to oce} \times SMP_{atm} + k_{terr \to oce} \times SMP_{terr} + 335 \quad k_{LMPsurf-oce \to SMP} \times LMP_{surf-oce} - k_{oce \to atm} \times SMP_{surf-oce} - k_{SMPsurf-oce \to sed} \times SMP_{surf-oce} \times 336 \quad f_{shelf} - k_{SMP-sink} \times SMP_{surf-oce} \times f_{pelagic} (Eq.7)$$

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$$\frac{d(P_{shelf-sed})}{dt} = k_{Psurf-oce \rightarrow sed} \times P_{surf-oce} \times f_{shelf}$$
(Eq.8)

d(IMD

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$$\frac{u(LMP_{shelf-sed})}{dt} = k_{LMPsurf-oce \rightarrow sed} \times LMP_{surf-oce} \times f_{shelf}$$
(Eq.9)

342
$$\frac{d(SMP_{shelf-sed})}{dt} = k_{SMPsurf-oce \rightarrow sed} \times SMP_{surf-oce} \times f_{shelf}$$
(Eq.10)

343 344

341

Where $f_{shelf} = 0.08$, is the fraction of global continental shelf surface area, and $f_{pelagic}$ is the fraction of open 345 ocean surface area. Subsurface ocean equations are: 346

347 $\frac{d(LMP_{deep-oce})}{dt} = k_{LMP-sink} \times LMP_{surf-oce} \times f_{pelagic} - k_{LMP \rightarrow SMP} \times LMP_{deep-oce} - k_{LMPdeep \rightarrow deepsed} \times deepsed \times deep$ 348 $LMP_{deep-oce}$ (Eq.11) 349 350 $\frac{d(SMP_{deep-oce})}{dt} = k_{SMP-sink} \times SMP_{surf-oce} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} - k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} + k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} + k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times LMP_{deep-oce} + k_{SMPdeep \to deepsed} \times f_{pelagic} + k_{LMP \to SMP} \times f_{pelagic} + k_{LMP \to SM$ 351 352 $SMP_{deep-oce}$ (Eq.12) 353 $\frac{d(P_{beach})}{dt} = k_{P-beach} \times P_{surf-oce} - k_{P \to LMP} \times P_{beach}$ (Eq.13) 354 355

356	$\frac{d(LMP_{beach})}{dt} = k_{LMP-beach} \times LMP_{surf-oce} + k_{P \to LMP} \times P$	Pbeach (Eq.14)
357		
358	$\frac{d(LMP_{deep-sed})}{dt} = k_{LMP-sed} \times LMP_{surf-oce} \times f_{pelagic}$	(Eq.15)
359		
360	$\frac{d(SMP_{deep-sed})}{dt} = k_{SMP-sed} \times SMP_{surf-oce} \times f_{pelagic}$	(Eq.16)
361		

Estimation of shelf sediment, deep sediment and beached P, and MP, based on reviews of literature data 362 reporting MP counts per surface area and particle size statistics, is relatively straightforward. The beached MP 363 pool is estimated at 0.5 Tg, based on the global surface of sandy beaches $(2.63 \cdot 10^5 \text{ km}^2; (28))$, a median global 364 beach sand MP abundance of 2450 MP km⁻² (IQR, 613 - 2700), and median MP size of 2.0 mm (IQR, 1.1 - 2700) 365 3.8) (29). Reviews of deep ocean MP and shelf sediment MP pools report numbers of MP counts per mass 366 unit, which leads to more intricate pool mass estimates: Barrett et al. (31) reported mean deep sediment MP 367 concentrations of 0.72 MP g⁻¹ for cored and grab sediment samples of 9cm depth. Deep sea sedimentation 368 rates are typically on the order of 0.1-1 cm per 1000 years, suggesting that the majority of such composite 369 sediment samples pre-date the plastics mass production period <1950. Yet, the measurement (0.72 MP g^{-1}) is 370 expressed relative to the bulk of the composite sample mass, representing on average 9 cm of deep sea 371 sediment (31). In this case we used the following data to estimate the global deep sea MP pool mass: depth in 372 cm, dry sediment bulk density of 1.37 g cm⁻³, a water to sediment mass ratio of 3.0, the mean MP size of 0.1 373 mm reported (31), a MP density of 1 x 10^{-6} µg µm⁻³, and an open ocean seafloor surface area of 3.36 x 10^{8} 374 km². Similarly: the shelf sediment MP pool is estimated from subtidal sediment median MP concentrations of 375 100 MP kg⁻¹ (IQR, 32-120), reviewed and reported by Shim et al. (29), a corresponding median MP size of 376 2.0 mm (IQR, 1.1 - 3.8), a dry sediment bulk density of 1.37 g cm⁻³, a typical shelf sedimentation rate of 1 377 mm y⁻¹, 65 years of MP accumulation (1950 – 2015), a water to sediment mass ratio of 3.0, and a shelf seafloor 378 surface area of 3.53×10^7 km². The final estimates for the deep ocean and shelf sediment MP pools are 1.5 Tg 379 and 65 Tg (1σ , 21 to 78Tg) respectively. We acknowledge that plastic litter concentrates in given areas of the 380 seafloor, and therefore, sediment sampling data could be biased depending on the sampling site. This is 381 382 ultimately reflected in the large budget uncertainties.

The global atmosphere. Brahney et al. (19,24) estimated the global atmosphere to contain 0.0036 Tg of SMP. 384 They also estimated global emissions from roads, 0.096 Tg y⁻¹, agricultural dust, 0.069 Tg y⁻¹, population dust, 385 0.018 Tg y⁻¹, and oceans, 8.6 Tg y⁻¹, which we adopt here. Atmospheric SMP deposition to remote terrestrial 386 surfaces has been investigated by Allen et al. (5) in France, finding a median SMP deposition of 0.011 Mg 387 $km^{-2} y^{-1}$, and by Brahney et al. (24). who observed a median of 0.0012 Mg $km^{-2} y^{-1}$ in the western USA. 388 Similar sampling and analysis techniques were used, and similar SMP particle and fiber size distributions 389 found, suggesting that the 9x difference reflects the difference in population density of both areas, 100 390 inhabitants per km² in SW Europe vs. 16 per km² in the western USA. In (sub-)urban environments in 391 Hamburg (Germany, 240 inhabitants per km²) mean SMP deposition of 0.016 \pm 0.006 Tg km⁻² y⁻¹ was 392 observed (38). Precursor studies on atmospheric plastics observed mostly the LMP fiber fraction (0.3 to 5mm) 393 with for example 0.014 Tg LMP km⁻² y⁻¹ in Dongguan (China) (23), but only 0.002 Tg km⁻² y⁻¹ in Paris 394 (France) (22). For simplicity we do not include LMP emission to the atmosphere in the box model, since the 395 short residence time of LMP likely leads to immediate deposition back to the broad terrestrial discarded LMP 396 397 reservoir. We regress SMP deposition over land, from the three detailed recent studies mentioned above, as a function of population density (Figure S1). We then extrapolate the observed relationship globally using 398 population density and surface area data per country for the year 2015 (44), capping SMP deposition at 0.016 399 Tg km⁻² y⁻¹ based on the Hamburg observations. Doing so leads to a global SMP deposition estimate over land 400 of 1.1 ± 0.5 Tg y⁻¹. SMP deposition over oceans is unconstrained by observations. We assume that global SMP 401 emissions (8.6 Tg y⁻¹; (19)) equal deposition, and estimate SMP deposition over oceans to be 7.5 Tg y⁻¹ (total 402 deposition of $8.6-1.1 \text{ Tg y}^{-1}$ deposition over land). 403

The mass inventory, emission and deposition flux estimates for 2015 serve to approximate the mass transfer coefficients associated with emission, $k_{oce \rightarrow atm}$ and deposition, $k_{atm \rightarrow oce}$, $k_{atm \rightarrow terr}$, in the following mass balance equation:

408 $\frac{d(SMP_{atm})}{dt} = k_{terr \to atm} \times SMP_{terr} + k_{disc \to atm} \times SMP_{disc} + k_{oce \to atm} \times SMP_{surf-oce} - k_{atm \to terr} \times$ 409 $SMP_{atm} - k_{atm \to oce} \times SMP_{atm} \qquad (Eq.17)$

410

414

411 We assume $k_{terr \rightarrow atm}$ to be equal to $k_{disc \rightarrow atm}$ which was optimized so that the modeled 2015 SMP emission flux 412 from the discarded pool fitted the flux of 0.18 Tg y⁻¹ (sum of road, population and agricultural SMP emission) 413 derived from the 3D global aerosol model for SMP dispersal by Brahney et al. (19).

Remote terrestrial pool. In the box model, agricultural and urban soils are included in the discarded plastics 415 pool. We use a separate box for remote terrestrial surfaces, outside of the technosphere, that are solely supplied 416 by atmospheric SMP. These include pristine soils, barren rock and land, ice sheets and remote inland waters. 417 We estimate the approximate amount of SMP in the remote terrestrial pool by making use of the quasi-linear 418 increase in global plastics production, discard and dispersal fluxes: global SMP deposition of 1.15 Tg y^{-1} in 419 2015 suggests a mean SMP deposition flux that is about half, 0.58 Tg y^{-1} since 1965, which multiplied by a 420 land surface area of 1.49 10⁸ km² amounts to 28 Tg of remote terrestrial SMP. SMP in this pool is mobilized 421 by rainfall to river runoff to the surface ocean, with the same k_{SMP→river-oce} that we derived for SMP runoff 422 from the discarded SMP pool. The remote terrestrial pool mass balance is: 423

424

425
$$\frac{d(SMP_{terr})}{dt} = k_{atm \to terr} \times SMP_{atm} - k_{terr \to atm} \times SMP_{terr} - k_{SMP \to river \to oce} \times SMP_{terr}$$
(Eq. 18)
426

BAU and SCS model scenarios. Both future, 2015 – 2050, model scenarios, business as usual (BAU), and 427 systems change scenario (SCS, from Lau et al. (4)), use the same mass transfer coefficients, k, but different 428 production, and waste management strategies summarized in the SI. BAU uses exponentially increasing 429 production, and quasi-linearly increasing incineration and recycling, and decreasing discard from Gever et al. 430 (3). Lau et al. (4) developed a detailed model of plastics stocks and flows from municipal solid waste (MSW) 431 and four sources of LMP. Their CSC scenario presents the most complete, yet feasible plastics management 432 strategy over the period 2016 – 2040 for MSW, including a decrease in plastics production by 2040 to 433 220 Tg y⁻¹. We digitized their disposal (incineration + safe landfilling), recycling and discard model output 434 (Tg y⁻¹), expressed these as fractions of MSW production, and extrapolated these to the year 2050 to compare 435 to BAU. To do so, we anchored (by normalization) the SCS disposal fractions for the period 2015 - 2050 to 436 the disposal fraction for 2050 - 2015 by Geyer et al.(3), in order to maintain a relatively smooth transition. 437 We acknowledge that the SCS waste disposal estimates deviate to some extent from the original (4) estimates, 438 but the overall trends are preserved: SCS disposal and recycling towards 2050 increase to 24 and 66 %, while 439 discard declines to 10%. Extrapolation of current waste disposal trends under the BAU scenario leads to 440 surprisingly similar numbers as SCS, though the real difference lies in the plastics production numbers that 441 reach 991 Tg y⁻¹ under BAU, and drop to 168 Tg y⁻¹ in the SCS by 2050 (SI). 442 443

Budget and model uncertainty. The model assumes no temporal evolution of the mass transfer coefficients, k, implying that fragmentation, sedimentation, emission, deposition and release dynamics are considered timeinvariant. While we argue that to first order these processes have remained similar through time, we acknowledge that reality is more complex. As more observational and mechanistic studies become available over the next decade, more appropriate parameterizations for plastics cycling can be tested, including the fragmentation of SMP to nanoplastics and ultimately dissolved and colloidal polymers with potential biological breakdown, i.e., as an energy source to biota.

Plastics data in the literature are predominantly reported as 'items per mass, volume, or surface area'. We converted these data to mass numbers by taking into account, where possible, the reported particle size distribution, or the reported median (or mean) particle size. In the case of fibers, reported length and diameter were used. Studies that did not report particle size properties were not included in the budget estimates. Particles were assumed to be flake shaped (45), with volume V defined as $V = L^{3*}0.1$, where L is the observed effective diameter, and have a mean density of 1 g cm⁻³. In summary, for each particle size class, reported L

was used to compute flake volumes, then multiplied by particle/fiber number, and multiplied by density to 457 obtain particle/fiber mass. The obtained masses were summed to obtain total P, LMP or SMP mass in a sample. 458 Table 4 summarizes 1_o (one relative standard deviation, in %) expanded uncertainties of observed P, 459 LMP and SMP pools (Tg) and fluxes (Tg y⁻¹), based on reported data, or conservatively approximated as 460 500%. The latter corresponds to a 2σ uncertainty of 1000%, which amounts to a factor 10. In other words, we 461 consider that a large number of plastics pools and fluxes are at the moment only known to within a factor of 462 10. In the future, as more observations on plastics pools, fluxes and degradation become available, we will 463 develop a formal Monte Carlo uncertainty analysis for the model. 464 465 Funding 466 We acknowledge financial support via the ANR-20-CE34-0014 ATMO-PLASTIC project, the Plasticopyr 467 project within the Interreg V-A Spain-France-Andorra program, a CNRS 80prime PhD scholarship, and a 468 MSCA ITN GMOS-Train PhD scholarship via grant agreement No 860497. 469 470 Acknowledgements 471 We thank the anonymous reviewers for their constructive comments. 472 473 **Author contributions** 474 JES designed the study. JES, AK and JLT developed the model. All authors reviewed literature data, and 475 contributed to model data interpretation and writing. 476 477 Availability of data and material 478 The authors declare that the data supporting the findings of this study are available within the paper and its 479 supplementary information files. 480 481 Ethics approval and consent to participate 482 NA 483 484 485 **Consent for publication** The authors provide consent for publication 486 **Competing interests** 487 The authors declare no competing financial or other interests. 488 489 490 References 491 Carpenter EJ, Smith KL. Plastics on the Sargasso sea surface. Science. 1972 Mar 17;175(4027):1240-492 1. 1. 493 Hughes L, Rudolph J. Future world oil production: growth, plateau, or peak? Current Opinion in 2. 494 Environmental Sustainability. 2011 Sep 1;3(4):225-34. 495 Gever R, Jambeck JR, Law KL. Production, use, and fate of all plastics ever made. Science Advances. 496 3. 2017 Jul 19;3(7):e1700782. 497 Lau WWY, Shiran Y, Bailey RM, Cook E, Stuchtey MR, Koskella J, et al. Evaluating scenarios toward 4. 498 zero plastic pollution. Science. 2020 Sep 18;369(6510):1455-61. 499 5. Allen S, Allen D, Phoenix VR, Le Roux G, Durántez Jiménez P, Simonneau A, et al. Atmospheric 500 transport and deposition of microplastics in a remote mountain catchment. Nat Geosci. 2019 501 May;12(5):339-44. 502 Peng X, Chen M, Chen S, Dasgupta S, Xu H, Ta K, et al. Microplastics contaminate the deepest part of 6. 503 the world's ocean. Geochemical Perspectives Letters. 2018 Nov 1;9:1-5. 504

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Table 1. Subsurface ocean microplastics (MS) observations. MS include fragments and fibers in the 0.3 - 5 mm (LMP) and <0.3 mm (SMP) range. Reported data in # m⁻³ were converted to mass concentrations, taking into account the full particle/fiber size distribution (see Methods).

Ocean basin	Location	depth	LMP+SMP	Reference
		m	μg m ⁻³	
N-Pacific	Korean East Sea	206	125	(34)
N-Pacific	Korean East Sea	2100	177	(34)
N-Pacific	Mariana Trench	2673	90	(6)
Mean			131	
1σ			44	
N- and S- Atlantic	-53° S to 47° N	160	134	(36)
N-Atlantic	Rockall Trough	2200	97	(35)
S-Atlantic	Gyre		43	(37)
Mean			91	
1σ			46	
Arctic Ocean	Central basin	5 to 1000	6	(46)
Arctic Ocean	Central basin	1769	66	(47)
Arctic Ocean	Fram Strait	300 to 5570	0.2	(48)
Mean			24	
1 σ			36	

612 Table 2. Global subsurface ocean microplastics budget. Atlantic, N-Pacific and Arctic Ocean data from 613 Table 1. Microplastics (MP) include fragments and fibers in the 0.3 – 5 mm (LMP) and <0.3 mm (SMP) range. 614 Data for the S-Pacific and Southern Ocean are extrapolated based on surface Ocean data from Shim et al.(29) 615 with uncertainties set to 10x. No data exists for the Indian Ocean, where concentrations were assumed equal 616 to the S-Atlantic observations by Eo et al.(34)(Table 1). Subsurface oceanic budgets in Tg include do not 617 include the mixed layer (upper 0.1 km).

Ocean basin	Area	Volume	MP	MP	1σ
	km ²	km ³	μg m ⁻³	Tg	Tg
Arctic Ocean	15558000	18750000	24	0.4	0.6
North Atlantic	41490000	146000000	91	13.0	3.0
South Atlantic	40270000	16000000	91	14.3	3.3
Indian Ocean	70560000	264000000	43	11.0	11.0
North Pacific	77010000	331000000	131	42.2	14.1
South Pacific	84750000	329000000	4	1.2	12.0
Southern Ocean	21960000	71800000	4	0.3	3.0
Total				82	47

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Table 3. Atmospheric small microplastics (SMP) budget. Mean $\pm 1\sigma$ SMP concentrations in the BL (boundary layer) (144 ± 124 ng m⁻³ for outdoors locations) and FT (free troposphere) (0.3 \pm 0.2 ng m⁻³) are from Allen et al. (25), assuming a mean SMP size of 70 µm for SMP in the BL, based on Brahney et al. (19).

	Mean global BL height	Mean global FT height	Area	BL SMP	FT SMP
	km	km	km ²	Tg	Tg
ocean	0.25	13	3.62 10 ⁸	0.013	0.0014
land	0.75	13	$1.48 \ 10^8$	0.016	0.0005
Total				0.0)31

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Table 4. Comparison of observed and modeled plastics mass (M, in Tg) and fluxes (F, in Tg y⁻¹) for the 625 year 2015. Plastics are divided in macroplastics, P (>5 mm), large microplastics, MP (0.3 – 5mm), and small 626 microplastics, SMP (<0.3 mm). Uncertainties (1 σ) on observations are based on the literature, except when 627 not reported, in which case we assigned an uncertainty, denoted by '*'. Uncertainties (1σ) on model estimated 628 pools and fluxes are conservatively estimated to be 500% (denoted by '**'), corresponding to a 2σ uncertainty 629 of a factor of 10 (see Methods). The second column with M and F abbreviations correspond to parameter 630 nomenclature used in mass balance equations 1-18. 631

Reservoir mass (M) or flux (F)	Abbreviation	Observed	Uncertainty	Box model	
			1σ		
M P produced	P _{prod}	8300	10%*	8297	
M P in-use	Puse	2600	10%*	3320	
M P discarded	Pdisc	4214	22%	2351	
M LMP discarded	LMP _{disc}	686	22%	1196	
M SMP discarded	SMP _{disc}			524**	
M P Surface Ocean	P _{surf-oce}	0.23	75%*	0.025	
M LMP Surface Ocean	LMP _{surf-oce}	0.031	75%*	0.044	
M SMP Surface Ocean	SMP _{surf-oce}	0.0028	196%	0.010	
M LMP Deep Ocean	LMP _{deep-oce}	0.2	F 70/	91	
M SMP Deep Ocean	SMP _{deep-oce}	82	57%	39	
M SMP atmosphere	SMPatm	0.03	500%	0.012	
M SMP remote terrestrial	SMPterr	28	37%	44	
M P beach	Pbeach	1.3	500%*	1.3	
M LMP beach	LMPbeach	0.53	100%	3.4	
M P shelf sediment	P _{shelf-sed}	51	500%*	51	
M LMP shelf sediment	LMP _{shelf-sed}	65	100%	11	
M SMP shelf sediment	SMP _{shelf-sed}			0.3**	
M LMP deep ocean sediment	LMP _{deep-sed}	1.0	F000/*	1.5	
M SMP deep ocean sediment	SMP _{deep-sed}	1.0	500%*	0.1	
M P Incinerated	Pincin	800	20%*	663	
M P Recycled	Precyc	750	20%*	554	
F Puse to LMPdisc		42	22%	41	
F Puse to Pdisc		118	22%	130	
F Puse to Pincin		74	20%*	63	
F Puse to Precyc		56	20%*	57	
F Pdisc to LMPdisc				71**	
F LMP _{disc} to SMP _{disc}				36**	
F P river				2.8**	
F LMP river				8.9**	
F SMP river (from SMP _{disc})				3.9**	
F SMP river (from P _{terr})				0.3**	
F river total		0.006-13		13	
F Surface Ocean P to LMP				0.0007**	
F Surface Ocean MP to SMP				0.001**	
F SMP Ocean to atmosphere		8.6	500%*	30	
F SMP Atmosphere to ocean		7.6	500%*	26	
F P beaching				0.1**	
F LMP beaching				0.2**	
F beach P to LMP				0.04**	
F LMP surface to deep ocean				8.0**	
F SMP surface to deep ocean				0.3**	
F P surface to shelf sediments				2.7**	
F LMP surface to shelf sediments				0.7**	
F SMP surface to shelf sediments				0.03**	
F LMP deep ocean to sediments				0.1**	
F SMP deep ocean to sediments				0.01**	

F SMP terrestrial to atmosphere			0.02**
F SMP atm to terrestrial pool	1.1	37%	3.8
F SMP Discard to atmosphere	0.087	500%*	0.2

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634

633 Figures



Figure 1. Global plastics budget and cycle for the year 2015. Reservoir sizes are shown in teragrams (Tg), 635 and fluxes in Tg y⁻¹ (arrows). Three plastics size classes are considered: macroplastics > 5mm (P), 636 microplastics from 0.3 to 5mm (LMP), and small microplastics <0.3mm (SMP) that can become airborne. The 637 discarded (Disc) plastic pools represent the terrestrial technosphere, including urban-industrial areas, landfills, 638 agricultural soils impacted by mulching or waste disposal, wetlands, and other impacted ecosystems. The 639 remote terrestrial reservoir lies outside the technosphere and is only impacted by airborne SMP deposition, 640 re-emission and runoff. Numbers in black are based on observations, and numbers in red on box model 641 optimization. Uncertainties are provided in Table 4. 642



Figure 2. Subsurface Ocean microplastics observations. (A) MP number concentrations per m³ of sea water. (B) MP mass concentrations for datasets where particle/fiber size distribution was reported (see Methods). The shaded vertical bar indicates the range of mean $\pm 1\sigma$ mass concentrations estimated for the Pacific and Atlantic Oceans (45 to 175 µg m⁻³, Table 1).



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Figure 3. Plastics dispersal through Earth surface reservoirs from 1950 to the year 3000, following a 650 halt on production and discard in 2025. This unrealistic model scenario illustrates over what timescales 651 discarded microplastic (P, >5mm), large microplastic (LMP) and small microplastic (SMP, <0.3mm) 652 potentially disperse via rivers and air into oceans, remote terrestrial surfaces, beach and marine sediments. (A) 653 P and LMP disappear in all transitory reservoirs within 100 and 200 years due to fragmentation at an annual 654 rate of 3%. The prolonged dispersal of SMP in all reservoirs is driven by cyclical marine emissions to air, 655 deposition to terrestrial surfaces, runoff to surface oceans, and re-emission to air. Only a small fraction of 656 SMP sinks to shelf sediments and to the deep ocean, followed by slow sedimentation to deep ocean sediments. 657 SMP mass, and concentrations, in the surface ocean and atmosphere, where human SMP exposure is relevant, 658 only return to 2025 levels towards the year 5000 (Figure S2). 659





Figure 4. Box model results for plastics cycling from 1950 to 2050. From 1950 to 2015 the model estimates 663 the dispersal of P, LMP and SMP in different Earth surface reservoirs, based on known plastics production 664 and waste generation. From 2015 to 2050 the model illustrates plastics production (A), amount of beached 665 macroplastics, P (B), the total, P+LMP+SMP, annual river plastics flux (C), and atmospheric deposition (atmo 666 dep) to remote land surfaces (**D**), for two different scenarios with different plastics production and waste 667 disposal trajectories: business as usual (BAU, grey dashed line) (3), and systems change scenario (CSC, orange 668 short dashed line) (4), the latter representing feasible plastics policy implementation. Despite the large 669 difference in plastics production towards 2050, 991 vs 168 Tg y⁻¹ in BAU and SCS, environmental stocks and 670 fluxes recover only slowly due to the large mobilization of mismanaged plastics from the terrestrial discarded 671 plastics pool that continue to cycle between land, ocean and atmosphere. Two remediation scenarios are 672 simulated for the 2025 to 2050 period: Discarded P remediation at a rate of 3% per year (yellow solid line), 673 and combined discarded P, LMP and SMP remediation at a rate of 3% per year. See Methods and SI for details 674 on BAU and SCS. 675

677 Supporting Information

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679 Modeling present and future plastics dispersal in the global environment and recommendations for 680 cleanup scenarios

- 681
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- 693
- 694

695 Table S1. Box model mass transfer coefficients k (y⁻¹).

	k y ⁻¹
k_beached P to LMP	3.00E-02
k_SurfOce LMP to DeepOce	1.96E+02
k_SurfOce SMP to DeepOce	3.25E+01
k_P_Disc to river to SurfOce	1.21E-03
k_LMP_Disc to river to SurfOce	7.41E-03
k_SMP_Disc to river to SurfOce	7.41E-03
k_Disc P to LMP	3.00E-02
k_Disc LMP to SMP	3.00E-02
k_Disc SMP to atm	3.66E-04
k_SurfOce P to beach	4.00E+00
k_SurfOce LMP to beach	4.00E+00
k_SurfOce P to ShelfSed	1.37E+03
k_SurfOce LMP ShelfSed	1.96E+02
k_SurfOce SMP ShelfSed	3.25E+01
k_SurfOce P to LMP	3.00E-02
k_SurfOce LMP to SMP	3.00E-02
k_SMP SurfOce to atm	3.13E+03
k_SMP terr to atm	3.66E-04
k_SMP atm to SurfOce	2.13E+03
k_SMP atm to terrestrial	3.08E+02
k_SMP terrestrial to SurfOce	7.41E-03
k_DeepOce LMP to DeepSed	1.23E-03
k_DeepOce SMP to DeepSed	2.03E-04

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Figure S1. Atmospheric small microplastic deposition versus population density. SMP deposition to
 remote locations for the USA from Brahney et al.⁸, for France (FR) from Allen at al.⁷, and for urban locations
 around Hamburg, Germany (DE) from Klein et al.³⁵. Error bars are 1σ standard deviations.



Figure S2. Plastics dispersal through Earth surface reservoirs from 1950 to the year 20,000, following 705 a halt on production and discard in 2025. This is the same model scenario that is shown in Figure 3, and 706 illustrates over what timescales discarded microplastic (P, >5mm), large microplastic (LMP) and small 707 microplastic (SMP, <0.3mm) potentially disperse via rivers and air into oceans, remote terrestrial surfaces, 708 and marine sediments. P and LMP disappear in all transitory reservoirs within 100 and 200 years due to 709 fragmentation at an annual rate of 3%. The prolonged dispersal of SMP in all reservoirs is driven by cyclical 710 marine emissions to air, deposition to terrestrial surfaces, runoff to surface oceans, and re-emission to air. 711 Only a small fraction of SMP sinks to shelf sediments and to the deep ocean, followed by slow sedimentation 712 to deep ocean sediments. 713

- 515 SI Model forcing data. Prod=production, Disc=discarded, Incin=incinerated, Recyc=recycled, SD=Safe Disposed
- 716 (incinerated and landfilled). BAU=business as usual scenario from Geyer et al., 2017; SCS=systems change scenario
- 717 from Lau et al., 2020.

	BAU	BAU	BAU	BAU	BAU	BAU	BAU	BAU	SCS	SCS	SCS	SCS	SCS	SCS	SCS	SCS
Year	Prod	Waste	Disc	Incin	Recyc	Disc	Incin	Recyc	Prod	Waste	Disc	SD	Recyc	Disc	Disp	Recyc
	Tg/y	Tg/y	fraction	fraction	fraction	Tg/y	Tg/y	Tg/y	Tg/y	Tg/y	fraction	fraction	fraction	Tg/y	Tg/y	Tg/y
1950	2.0	1.0	1.00	0.00	0.00	1.0	0.0	0.0	2.0	1.0	1.00	0.00	0.00	1.0	0.0	0.0
1951	2.0	1.0	1.00	0.00	0.00	1.0	0.0	0.0	2.0	1.0	1.00	0.00	0.00	1.0	0.0	0.0
1952	2.0	1.0	1.00	0.00	0.00	1.0	0.0	0.0	2.0	1.0	1.00	0.00	0.00	1.0	0.0	0.0
1953	3.0	1.5	1.00	0.00	0.00	1.5	0.0	0.0	3.0	1.5	1.00	0.00	0.00	1.5	0.0	0.0
1954	3.0	1.5	1.00	0.00	0.00	1.5	0.0	0.0	3.0	1.5	1.00	0.00	0.00	1.5	0.0	0.0
1955	4.0	2.0	1.00	0.00	0.00	2.0	0.0	0.0	4.0	2.0	1.00	0.00	0.00	2.0	0.0	0.0
1956	5.0	2.5	1.00	0.00	0.00	2.5	0.0	0.0	5.0	2.5	1.00	0.00	0.00	2.5	0.0	0.0
1957	5.0	2.5	1.00	0.00	0.00	2.5	0.0	0.0	5.0	2.5	1.00	0.00	0.00	2.5	0.0	0.0
1958	6.0	3.0	1.00	0.00	0.00	3.0	0.0	0.0	6.0	3.0	1.00	0.00	0.00	3.0	0.0	0.0
1959	7.0	3.5	1.00	0.00	0.00	3.5	0.0	0.0	7.0	3.5	1.00	0.00	0.00	3.5	0.0	0.0
1960	8.0	4.0	1.00	0.00	0.00	4.0	0.0	0.0	8.0	4.0	1.00	0.00	0.00	4.0	0.0	0.0
1961	9.0	4.5	1.00	0.00	0.00	4.5	0.0	0.0	9.0	4.5	1.00	0.00	0.00	4.5	0.0	0.0
1962	10	5.4	1.00	0.00	0.00	5.4	0.0	0.0	10	5.4	1.00	0.00	0.00	5.4	0.0	0.0
1963	12	6.4	1.00	0.00	0.00	6.4	0.0	0.0	12	6.4	1.00	0.00	0.00	6.4	0.0	0.0
1964	13	7.6	1.00	0.00	0.00	7.6	0.0	0.0	13	7.6	1.00	0.00	0.00	7.6	0.0	0.0
1965	15	8.9	1.00	0.00	0.00	8.9	0.0	0.0	15	8.9	1.00	0.00	0.00	8.9	0.0	0.0
1966	17	10	1.00	0.00	0.00	10	0.0	0.0	17	10	1.00	0.00	0.00	10	0.0	0.0
1967	19	12	1.00	0.00	0.00	12	0.0	0.0	19	12	1.00	0.00	0.00	12	0.0	0.0
1968	22	14	1.00	0.00	0.00	14	0.0	0.0	22	14	1.00	0.00	0.00	14	0.0	0.0
1969	24	15	1.00	0.00	0.00	15	0.0	0.0	24	15	1.00	0.00	0.00	15	0.0	0.0
1970	27	17	1.00	0.00	0.00	17	0.0	0.0	27	17	1.00	0.00	0.00	17	0.0	0.0
1971	30	19	1.00	0.00	0.00	19	0.0	0.0	30	19	1.00	0.00	0.00	19	0.0	0.0
1972	34	22	1.00	0.00	0.00	22	0.0	0.0	34	22	1.00	0.00	0.00	22	0.0	0.0
1973	37	24	1.00	0.00	0.00	24	0.0	0.0	37	24	1.00	0.00	0.00	24	0.0	0.0
1974	41	27	1.00	0.00	0.00	27	0.0	0.0	41	27	1.00	0.00	0.00	27	0.0	0.0
1975	45	29	1.00	0.00	0.00	29	0.0	0.0	45	29	1.00	0.00	0.00	29	0.0	0.0
1976	49	32	1.00	0.00	0.00	32	0.0	0.0	49	32	1.00	0.00	0.00	32	0.0	0.0
1977	53	35	1.00	0.00	0.00	35	0.0	0.0	53	35	1.00	0.00	0.00	35	0.0	0.0
1978	58	38	1.00	0.00	0.00	38	0.0	0.0	58	38	1.00	0.00	0.00	38	0.0	0.0
1979	62	42	1.00	0.00	0.00	42	0.0	0.0	62	42	1.00	0.00	0.00	42	0.0	0.0
1980	67	45	1.00	0.00	0.00	45	0.0	0.0	67	45	0.98	0.02	0.00	44	0.7	0.0
1981	72	49	1.00	0.00	0.00	49	0.0	0.0	72	49	0.98	0.02	0.00	48	0.8	0.0
1982	78	53	1.00	0.00	0.00	52	0.2	0.0	78	53	0.98	0.02	0.00	52	0.9	0.0
1983	83	57	0.99	0.01	0.00	56	0.5	0.0	83	57	0.98	0.02	0.00	55	1.1	0.0
1984	89	61	0.99	0.01	0.00	60	0.9	0.0	89	61	0.98	0.02	0.00	59	1.2	0.0
1985	95	65	0.98	0.02	0.00	64	1.2	0.0	95	65	0.98	0.02	0.00	64	1.5	0.0
1986	101	70	0.98	0.02	0.00	68	1.7	0.0	101	70	0.98	0.02	0.00	68	1.7	0.0
1987	108	74	0.97	0.03	0.00	72	2.2	0.0	108	74	0.97	0.03	0.00	72	2.0	0.0
1988	114	79	0.96	0.03	0.00	76	2.7	0.3	114	79	0.97	0.03	0.00	77	2.3	0.0
1989	121	84	0.95	0.04	0.01	80	3.3	0.9	121	84	0.97	0.03	0.00	82	2.7	0.0
1990	128	90	0.94	0.05	0.02	84	4.0	1.6	128	90	0.96	0.04	0.00	86	3.2	0.2
1991	136	95	0.92	0.05	0.03	88	4.8	2.4	136	95	0.94	0.04	0.02	90	3.8	1.6
1992	143	101	0.91	0.06	0.03	92	5.6	3.2	143	101	0.93	0.04	0.03	93	4.4	3.0
1993	151	107	0.90	0.06	0.04	96	6.6	4.2	151	107	0.91	0.05	0.04	97	5.1	4.4

	Non-peer reviewed EarthArXiv preprint															
1994	159	113	0.89	0.07	0.05	100	7.6	5.2	159	113	0.89	0.05	0.05	101	6.0	5.9
1995	167	119	0.87	0.07	0.05	104	8.7	6.4	167	119	0.88	0.06	0.06	104	7.0	7.5
1996	175	125	0.86	0.08	0.06	108	9.9	7.6	175	125	0.86	0.06	0.07	108	8.1	9.1
1997	183	132	0.85	0.09	0.07	112	11	9.0	183	132	0.85	0.07	0.08	112	9.3	11
1998	192	139	0.83	0.09	0.07	116	13	10	192	139	0.83	0.08	0.09	116	11	12
1999	201	146	0.82	0.10	0.08	120	14	12	201	146	0.82	0.08	0.10	120	12	14
2000	210	153	0.81	0.10	0.09	124	16	14	210	153	0.80	0.09	0.10	123	14	16
2001	220	161	0.79	0.11	0.10	128	18	16	220	161	0.79	0.10	0.11	127	16	18
2002	229	169	0.78	0.12	0.10	132	20	17	229	169	0.78	0.11	0.11	131	18	19
2003	239	177	0.77	0.12	0.11	136	22	20	239	177	0.76	0.12	0.12	135	21	21
2004	249	185	0.75	0.13	0.12	139	24	22	249	185	0.75	0.13	0.12	139	23	23
2005	259	193	0.74	0.14	0.12	143	26	24	259	193	0.74	0.14	0.13	142	26	25
2006	269	202	0.73	0.14	0.13	147	29	27	269	202	0.72	0.15	0.13	146	29	27
2007	280	211	0.71	0.15	0.14	150	32	29	280	211	0.71	0.16	0.14	149	33	29
2008	291	220	0.70	0.16	0.15	154	34	32	291	220	0.69	0.17	0.14	153	37	31
2009	302	230	0.68	0.16	0.15	157	38	35	302	230	0.68	0.18	0.14	156	41	33
2010	313	239	0.67	0.17	0.16	160	41	38	313	239	0.66	0.19	0.15	159	45	35
2011	325	249	0.65	0.18	0.17	163	44	42	325	249	0.65	0.20	0.15	162	50	37
2012	336	259	0.64	0.19	0.17	166	48	45	336	259	0.64	0.21	0.15	165	55	40
2013	348	270	0.63	0.19	0.18	169	52	49	348	270	0.62	0.22	0.16	168	60	42
2014	360	280	0.61	0.20	0.19	171	56	53	360	280	0.61	0.23	0.16	170	66	44
2015	372	291	0.60	0.21	0.20	174	60	57	372	291	0.59	0.25	0.16	173	72	47
2016	385	303	0.58	0.21	0.20	176	65	61	375	303	0.58	0.26	0.16	175	78	50
2017	398	314	0.57	0.22	0.21	178	70	66	369	313	0.56	0.27	0.17	176	84	53
2018	410	326	0.55	0.23	0.22	180	75	71	362	322	0.55	0.28	0.17	176	91	55
2019	424	338	0.54	0.24	0.22	181	80	76	356	331	0.53	0.29	0.18	176	97	58
2020	437	350	0.52	0.25	0.23	183	86	81	349	339	0.52	0.30	0.18	175	103	61
2021	450	362	0.51	0.25	0.24	184	92	87	343	345	0.50	0.32	0.18	173	109	63
2022	464	375	0.49	0.26	0.25	184	98	92	336	351	0.48	0.33	0.19	170	115	66
2023	478	388	0.48	0.27	0.25	185	105	98	330	357	0.47	0.34	0.19	167	120	69
2024	492	402	0.46	0.28	0.26	185	112	105	323	361	0.45	0.35	0.20	164	125	72
2025	507	415	0.45	0.29	0.27	185	119	111	317	364	0.44	0.36	0.21	159	130	75
2026	521	429	0.43	0.30	0.27	184	127	118	311	367	0.42	0.37	0.21	155	134	78
2027	536	443	0.41	0.30	0.28	183	135	125	304	368	0.41	0.37	0.22	149	138	81
2028	551	458	0.40	0.31	0.29	182	143	132	298	369	0.39	0.38	0.23	144	141	84
2029	566	473	0.38	0.32	0.30	181	152	140	291	369	0.37	0.39	0.24	138	144	87
2030	582	488	0.37	0.33	0.30	179	161	148	285	368	0.36	0.40	0.25	132	146	91
2031	597	503	0.35	0.34	0.31	176	171	156	278	366	0.34	0.40	0.26	126	147	94
2032	613	519	0.33	0.35	0.32	173	181	165	272	364	0.33	0.40	0.27	119	147	97
2033	629	535	0.32	0.36	0.32	170	191	174	265	360	0.31	0.41	0.28	113	147	101
2034	645	551	0.30	0.37	0.33	166	202	183	259	356	0.30	0.41	0.29	106	146	104
2035	662	568	0.28	0.38	0.34	162	214	192	253	351	0.28	0.41	0.31	99	144	107
2036	678	585	0.27	0.39	0.35	157	225	202	246	344	0.27	0.41	0.32	93	141	111
2037	695	602	0.25	0.40	0.35	152	238	212	240	337	0.26	0.41	0.34	86	137	114
2038	712	619	0.24	0.40	0.36	146	251	223	233	330	0.24	0.40	0.35	80	133	117
2039	730	637	0.22	0.41	0.37	139	264	234	227	321	0.23	0.40	0.37	74	128	120
2040	747	655	0.20	0.42	0.37	132	278	245	220	311	0.22	0.39	0.39	68	121	122
2041	765	674	0.18	0.43	0.38	124	292	257	214	301	0.21	0.38	0.41	62	115	124
2042	783	693	0.17	0.44	0.39	116	307	269	208	290	0.19	0.37	0.44	56	107	126
2043	801	712	0.15	0.45	0.40	107	323	282	201	277	0.18	0.36	0.46	51	99	127

	Non-peer reviewed EarthArXiv preprint															
2044	819	731	0.13	0.46	0.40	98	339	295	195	264	0.18	0.34	0.48	46	90	128
2045	838	751	0.12	0.47	0.41	87	356	308	188	251	0.17	0.32	0.51	42	81	128
2046	857	772	0.10	0.48	0.42	76	374	322	182	236	0.16	0.30	0.54	38	71	127
2047	876	792	0.08	0.49	0.42	64	392	336	175	220	0.15	0.28	0.57	34	61	125
2048	895	813	0.06	0.50	0.43	52	410	351	169	204	0.15	0.25	0.60	30	52	122
2049	914	834	0.05	0.52	0.44	38	430	366	162	186	0.14	0.22	0.63	27	42	117
2050	934	856	0.03	0.53	0.45	24	450	381	156	168	0.14	0.19	0.66	24	32	112