Supplementary Material

All is not lost:

Deriving a top-down mass budget of plastic at sea

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Detailed motivation of model assumptions and default parameter values

Detailed support for model assumptions - There are ten assumptions underlying the model approach (Figure 1):

1. Input of plastic to the oceans can be estimated from annual global plastic production data.
2. A constant fraction of this annual production reaches the oceans and is initially buoyant, entering at the surface.
3. The plastic particles emitted to and present in the oceans are separated into three operationally defined size fractions: macroplastic, microplastic and nanoplastic\(^1\) particles.
4. Macro-, micro-, and nanoplastics comprise constant fractions of the total annual input to the oceans.
5. Particles are first considered as spheres, and then are corrected for non-sphericity using (approximate) shape correction factors.
6. Macro- and microplastics are subject to degradation, abrasion and fragmentation processes (hereafter collectively referred to as ‘fragmentation’) occurring either in the water or inside organisms, the mass-based rates of which are proportional to the surface area of the particles.
7. Macro- and microplastics are subjected to sedimentation, which can occur either as immediate settling for the fraction of emitted plastic with a density greater than seawater, or as a result of biofouling or by entrainment in biological aggregates (including fecal pellets after ingestion) for plastics with a density lower than seawater.
8. The mass balance for initially buoyant macroplastics has the macroplastic fraction of the annual plastic production as gain, and sedimentation and fragmentation to microplastics as loss processes.
9. The mass balance for initially buoyant microplastics has the microplastic fraction of annual emission plus fragmentation from macroplastics as inputs, and sedimentation plus fragmentation to nanoplastic particles as loss processes.
10. At all times, the difference between the cumulative mass of plastic emitted to the oceans and the mass of macro- and microplastics in the OSL equals the sum of (a) emitted plastic with a density greater than seawater, (b) the mass of settling microplastics, and (c) the mass of < 0.335 mm ‘nanoplastics’.

\(^1\) ‘Nanoplastic’ is pragmatically defined as particles < 0.335 mm, but we do not explicitly include particles exhibiting colloidal behavior in a fluid.
The above implies that plastic debris on beaches and ingested plastic are not explicitly modeled, that is, with separate differential equations. This is motivated as follows. Plastic debris on shorelines is part of the fraction residing on land and thus is omitted from the analysis through assumption 2. Uncertainty in this fraction is covered in the uncertainty analysis.

Ingested plastic is omitted from the mass balance equations because of two main reasons:

(a) First and most importantly, the modeled loss rate through ingestion is small compared to the other loss mechanisms. For instance, ingestion by birds has been estimated at ~100 tons per year at present (Van Franeker 2011), and ingestion by mesopelagic fishes in the North Pacific is an estimated 12-24 thousand tons per year (Davison and Asch 2011), whereas our modeled removal of plastic is an excess of 9.4 million tons for 2016. Hence, even if we would take the estimate of 12-24 thousand tons per year for mesopelagic fish ten times higher to cover the world ocean, the ingested quantities still would have a negligible effect on the mass balance. As for ingestion by zooplankton; Desforges et al (2015) report marine zooplankton in the OSL of North Pacific food webs to ingest a negligible < 0.07% of the microplastic number concentration. Hence, these available data clearly suggest an overall small effect on the microplastic mass balance.

(b) Second, ingestion of plastic debris by marine organisms is not considered a permanent sink. This can be motivated for various groups of organisms as follows. Phytoplankton does not ingest plastic but plays a role in forming aggregates or marine snow (Long et al, 2015), which however just would settle with the rate estimated by the model. The gut passage time in zooplankton is in the order of hours to days (eg., Cole et al, 2013), which implies that there will be a marginal (unmeasurable) time delay in settling of plastic. The egested plastics are known to settle in fecal pellets (Cole et al, 2013, 2014, 2016), which is accounted for in assumption 7. After ingestion and gut passage by fish, microplastics also are shown to be egested (Choy and Drazen 2013; Grigorakis et al, 2017; Rummel et al, 2016). In the paper by Choy & Drazen (2013), 159 debris pieces found in fish stomachs of multiple species were used for buoyancy trials. All these plastic particles appeared to be buoyant in seawater representing depths of 0–50m. Therefore they argued that ‘.. it is reasonable to assume that plastics are present in the water column either as neutrally drifting pieces or slowly sinking particles. However, biofouling organisms would be digested in an acidic fish stomach
after ingestion and the debris buoyancy properties would change.’ These data thus show that the biofouling layer is stripped off in the fish gut after which particles would surface again. As for seabirds, Van Franeker et al. (2011) provided an estimate of the loss rate of 75% of ingested plastic in one month, thus implying that plastics ingested by seabirds do not represent a permanent sink either. In summary, after ingestion and gut passage, microplastics are egested and return to the OSL (Choy and Drazен 2013), which would result in a time-delay in the OSL abundance that can be assumed to be small and relatively constant in time. Alternatively, egested plastics are known to settle in fecal pellets (Cole et al, 2013, 2014, 2016), which is accounted for in assumption 7.

Detailed support for default parameter values -

Global plastic production - The time dependent annual world plastic production \( (P_t; \text{MT \times yr}^{-1}) \) represents one of the most reliable terms in the model, as global production values are reported annually by PlasticsEurope (PlasticsEurope 2013). \( P_t \) can be accurately captured by a 2nd order polynomial \( (R^2=0.999; \text{Table 1}) \), as was shown in our previous study (Koelmans et al 2016). The cumulative world plastic production since 1950, \( \int_{1950}^t P_t \) (Eq 4) (4808 MT in 2016), is obtained by integrating the polynomial defining \( P_t \).

Fraction of annually produced plastic entering the sea – Source types of plastic and microplastics have been reviewed by GESAMP 2016 and include: pellets & fragments, Greenhouse-sheets, pots, pipes, nutrient prills, fishing gear, packaging, buoys, lines, nets, PVC pipes, EPS, packaging, pellets, tyres, tyre dust, paints, pipes, clothes, plastic-blasting, cargo, consumer goods, microbeads, textile fibres, synthetic turf, containers, plastic bags, bottles, caps, cups, plates, straws, spoons, etc., microbeads, packaging, toothbrushes, etc. Because exact emission inventories and pathway analysis are lacking, the fraction of annually produced plastic entering the oceans is uncertain. For some time it was assumed that about 10% of the annual world plastic production enters the oceans (Thompson 2006). Recent work based upon estimates of plastic waste generation on land suggested that about 4.8-12.7 MT of plastic entered the ocean from a total of 275 MT produced in 2010, which translates to 1.7-4.6 % of total produced plastic entering the ocean (Jambeck et al 2015). Although there will be some variability among years, we assume that on a global scale on average no substantial changes in the proportion of plastic getting into the oceans occurred over the past decades (e.g. constant relative rates of plastics being lost/discard from fisheries, catchment
hydrology and waste management streams). This would imply a more or less constant relative retention of produced plastic debris on the continents. Based on these data and arguments, parameter ‘A’ was set at 0.03 (range 0.017-0.046). Note that, because production has been exponential, the integrated emissions are heavily weighted towards the last few years, for which the 2010 land-based waste calculation is the best available estimate.

**Fraction of buoyant plastic** – Some plastics will be removed from the ocean surface layer by sinking immediately upon entering the ocean, which is why the input fraction $A \times P_t$ is reduced to only buoyant plastics by multiplication with parameter $B$ ($0 < B < 1$). Plastics with densities less than that of seawater are polypropylene (PP), low density polyethylene (LDPE/LLDPE), and high density polyethylene (HDPE), with production fractions of 0.24, 0.21 and 0.17, respectively (Andrady 2015), and expanded polystyrene with an estimated production fraction of 0.03 (PlasticEurope 2013). Nylon has a density of 1.02-1.05 g/cm$^3$ and is nearly buoyant, but the production fraction is $< 0.03$ (Andrady 2015) and therefore nylon was omitted. Based on these reported fractions, parameter ‘B’ was set at 0.65.

**Fraction of primary nano- and microplastic** - No data exist on the fraction of the plastic emission to sea in the form of primary microplastics smaller than 5 mm. However, it has been calculated recently that for plastics originating from Europe, about 11% would be comprised of microplastics from cosmetics when waste water treatment plant (WWTP) retention was not taken into account (Gouin et al, 2015). Retention in a typical European WWTP would lower the emissions by an extra factor of ten, leading to an estimated percentage of the plastic emission to sea in the form of primary microplastics of 1%. The actual percentage could be somewhat higher due to less efficient WWTPs elsewhere or due to the emission of other primary microplastics besides cosmetic products, but the percentage also could be lower due to the river retention that recently has been shown to play a role (Besseling et al 2014, 2017; Nizzetto et al, 2016). Based on these data and arguments this fraction was set at 0.01. Part of this fraction will however relate to the particles with a size less than 335 µm, here referred to as nanoplastics. Assuming equal mass fractions for particles $< 335$ µm and $0.335 – 5$ mm, the parameters ‘C’ and ‘D’ were each set at 0.005.

**Macro- and microplastic size and surface area** – In the model we distinguish between two particle sizes: macro- and microplastic. These are the two main categories of plastic considered in the literature with respect to sources, fate and effects of plastic debris (Andrady 2011). Although more detailed size distributions would better reflect reality, insufficient data
exist to define these distributions, which also vary over space and time. Therefore, to avoid over-parameterization, we approximate the two main categories as if they are monodisperse pools of plastic debris. From reported data for particle sizes in the OSL (Eriksen et al 2014), we approximated representative average particle equivalent sphere radii of 10 cm and 1.2 mm for macroplastic \( r_{MA} \) and microplastic \( r_{MI} \) respectively. To account for irregular shapes, surface roughness and fractal geometry (Douglas 1989, Farin et al 1987), the dimensionless non-sphericity surface correction factor \( \alpha_{MI} \) was set at 2. This means that the average microplastic particle was assumed to have a surface of two times that of a perfect sphere with the same particle volume. Macroplastic was assumed to be more irregular and non-spherical than microplastic, because of larger proportions of nearly whole items such as plastic sheets, fragments of cups, caps, bottles, plates, crates, nets, ropes and wires. The surface correction factor \( \alpha_{MA} \) therefore was assigned a value of 30. These \( \alpha \) values (\( \alpha_{MI} = 2 \) and \( \alpha_{MA} = 30 \)) are motivated in detail in the next section.

Values for the dimensionless non-sphericity shape correction factors (\( \alpha_{MA} \) and \( \alpha_{MI} \)) - Modeled surface areas assuming spherical particles were converted to surfaces for more realistic shapes representing plastic debris, using dimensionless non-sphericity surface correction factors \( \alpha_{MA} \) or \( \alpha_{MI} \). This factor corrects for the surface of actual geometrical shapes relative to that of the surface area of spherical particles. For instance, an \( \alpha \) value of two means that the average microplastic particle was assumed to have a surface of two times that of a perfect sphere with the same particle volume. Here we provide some example calculations showing some typical values for these factors.

For macroplastic, it appears that cubes and spheroids with the same volume as a sphere have approximately 1.2 times more surface (Table S1). For sheets, plates, threads and wires however, non-sphericity can yield an increase in apparent surface area of up to one to two orders of magnitude and these shapes are far more abundant than macro-sized cubes or spheroids. No actual data on the relative abundances of particle shapes in the OSL are available. However, based on the values in Table S1, \( \alpha_{MA} \) was set at 30.
**Table S1**: Dimensionless non-sphericity surface correction factor $\alpha_{MA}$ for specific geometric shapes, based on common types of macroplastics.

| Geometrical shape | Volume (m$^3$) | Surface (m$^2$) | $\alpha_{MA}$ | Comment |
|-------------------|----------------|-----------------|---------------|---------|
| Sphere            | 0.00419        | 0.126           | 1             | Based on a default macroplastic particle radius of 0.1 m. |
| Cube              | 0.00419        | 0.156           | 1.24          | The cube volume translates to a cube side length of 0.161 m. |
| Rectangular prism | 0.00419        | 8.39            | 66.7          | Based on an assumed polymer sheet thickness of 1 mm |
| Rectangular prism | 0.00419        | 3.36            | 26.8          | Based on an assumed polymer plate thickness of 2.5 mm |
| Rectangular prism | 0.00419        | 0.86            | 6.87          | Based on an assumed polymer block thickness of 10 mm |
| Cylinder          | 0.00419        | 8.38            | 66.7          | Based on an assumed thread/wire radius of 1 mm |
| Cylinder          | 0.00419        | 3.35            | 26.7          | Based on an assumed wire/rope radius of 2.5 mm |
| Prolate spheroid  | 0.00419        | 0.148           | 1.18          | Based on prolate spheroid radii $3r_1=3r_2=r_3$. (rugby ball shape) |

a) Volume of a sphere with radius 0.1 m, which was kept constant for all shapes considered.

Like for macroplastic, for microplastic no data on the relative abundances of particle shapes in the OSL are available. However, due to their smaller size and higher degree of weathering, microplastic particles are generally more close to sphericity. Therefore, we selected the dimensions of the example rectangular prism to be closer to the dimensions of a cube than to the dimensions of a plate or thin sheet, and also considered almost spherical oblate spheroids (Table S2). Apart from fibres, $\alpha_{MI}$ for some of the representative microplastic particle shapes ranges between 1 and 1.4 based on geometric shape alone. However, the actual surface area is higher due to surface fractal dimensions of weathered polymers being between 2 and 3. Therefore, and because of presence of fibers, $\alpha_{MI}$ was set at a value of 2.
Table S2: Dimensionless non-sphericity surface correction factor $\alpha_{MI}$ for specific geometric shapes, based on common types of microplastics.

| Geometrical shape | Volume ($m^3$) | Surface ($m^2$) | $\alpha_{MI}$ | Comment |
|-------------------|---------------|----------------|---------------|---------|
| Sphere            | $7.24 \times 10^{-9}$ | $1.81 \times 10^{-5}$ | 1 | Based on a default microplastic particle radius of 0.0012 m. |
| Cube              | $7.24 \times 10^{-9}$ | $2.25 \times 10^{-5}$ | 1.24 | The cube volume translates to a cube side length of 1.9 mm. |
| Rectangular prism | $7.24 \times 10^{-9}$ | $2.52 \times 10^{-5}$ | 1.39 | Based on a typical microplastic particle with thickness of 1 mm and area $2.7 \times 2.7$ mm |
| Cylinder          | $7.24 \times 10^{-9}$ | $5.83 \times 10^{-5}$ | 3.22 | Based on an assumed fibre radius of 0.25 mm, length 37 mm. |
| Prolate spheroid  | $7.24 \times 10^{-9}$ | $1.95 \times 10^{-5}$ | 1.08 | Based on prolate spheroid radii $2r_1=2r_2=r_3$. (rugby ball shape) |
| Prolate spheroid  | $7.24 \times 10^{-9}$ | $2.13 \times 10^{-5}$ | 1.18 | Based on prolate spheroid radii $3r_1=3r_2=r_3$. (rugby ball shape) |

a) Volume of a sphere with radius 0.0012 m, which was kept constant for all shapes considered.

**Fragmentation of plastic** - Size reduction of particulates or aggregates has been described in terms of two main categories of processes: rupture and erosion processes (Rwei et al 1991). Here we argue that for removal of plastic due to degradation, fragmentation, wear or abrasion, a surface area based (‘onion peeling’) erosion mechanism is most likely, because the processes hypothesized to act in the degradation of plastic all occur at the surface. For instance, solar UV-induced photo-oxidation is considered a dominant plastic degradation mechanism in the environment. UV radiation and oxygen have limited penetration depth in the polymer, causing oxidation, erosion and cracking to occur at the surface of the particle (Andrady 2011, Andrady 2015, Gewert et al 2015, Gardette et al 2013, Weinstein et al 2016, Lambert and Wagner, 2016; Song et al, 2017). Also, if any biodegradation of plastic would occur under marine conditions (Harshvardhan and Jha 2013; Restrepo-Flórez et al 2014), this would involve growth of microorganisms on the surface that obtain their carbon nutrient from
the plastics. This leads to a loss of polymer integrity at the surface and eventually loss of polymer mass (Sudhakar et al 2007). Recent work reported a range of plastic surface microtextures, including pits and grooves conforming to the shape of microorganisms, supporting the occurrence of surface degradation weathering due to the presence of biota (Reisser et al 2014). Physical wear and abrasion may be relevant for beached plastic particles that afterwards may be washed back into the ocean, and inside seabirds microplastics are worn down or fragmented into sizes small enough to pass into the intestines (Van Franeker and Law 2015) and presumably be excreted. Tests and theory for the abrasive wear of polymers is well developed and based on the production of loose particles from the surface due to disruption of material at the contact point at the surface (Budinsky 1997, Cenna et al 2003). Hence, abrasion is also a surface area-based loss process.

Loss of mass due to all these processes can be assumed to occur at a rate per unit of surface area. It is not known what the relative share of each of these individual processes to overall fragmentation is, nor how this varies in time and space and how this may differ among plastic types in different exposure conditions. Nevertheless, we can define an apparent ‘ocean average’ surface area-normalized fragmentation rate constant ($k_F$) that can be used to calculate the effect of surface fragmentation on the scale of total mass (M) in the ocean, i.e. $\frac{dM}{dt} = -k_F A_{M,t} M$, like we suggest in Equations 1 and 2 for macro- and microplastics respectively. Consequently, the apparent first order rate constant for fragmentation ($k_F A_{M,t}$) is dependent on the total surface area $A_{M,t}$, which in turn depends on the mass, size and shape of the plastic. Another implication is that at equal mass, degradation of smaller plastics proceeds faster than that of larger plastics, because small plastic particles have a higher surface to volume ratio (Gewert et al 2015, ter Halle 2016). Note that the parameter $k_F$ (m$^{-2}$ × yr$^{-1}$) reflects the fragmentation rate per unit of surface, reflecting the average in situ condition in the world ocean, averaging variability due to temperature, degree of biofouling, particle size, local
turbulence, UV irradiance, oxygen concentration and temporary beaching. Therefore, this parameter cannot be calculated from first principles and is a calibration parameter.

*Biofouling and sedimentation* – Plastics entering the sea are removed from the OSL by immediate settling of dense plastics (fraction 1-B), or by biofouling of initially buoyant plastics (fraction B) and subsequent settling. Because the biofilm/plastic mass ratios are higher for smaller particles, this process is especially important for microplastic, as shown in a recent experimental study (Fazey et al, 2016). Consequently, in the model the buoyant macroplastic is assigned a settling rate ($V_{SM}^A$) of zero; that is, settling of buoyant macroplastic is assumed negligible compared to removal due to fragmentation. In principle, the settling rate of microplastics can be estimated from Stokes law; however, particle densities, biofilm thickness and biofilm properties, as well as water density and OSL thickness and turbulence are highly variable and difficult to define. Therefore, the apparent settling rate of microplastics ($V_{SM}^I$) from the OSL is another calibration parameter. The thickness of the OSL usually is between 25 and 200 m, but can vary between 10 and > 1000 m (de Boyer Montégut 2004). Based on reported spatial distributions of OSL thicknesses across the oceans we selected an average OSL mixing depth of 100 m (de Boyer Montégut 2004).
Detailed explanation of the calculation of the surface area of macro- and microplastic from the modeled mass of macro- and microplastic (MAₜ, MIₜ)

Equations 1 and 2 in the main manuscript simulate the masses of macroplastic (MA) and microplastic (MI) over time. Here we explain how the surface areas of MA and MI are calculated, given these masses. First, the total volume (m³) of macro- and microplastic in the world ocean OSL is calculated as:

\[ V_{MA,t} = \frac{MA_t 10^6}{\sigma_{PL}} \text{ and } V_{MI,t} = \frac{MI_t 10^6}{\sigma_{PL}} \]  (S1)

The factor \(10^6\) is required because mass of plastic (MAₜ, MIₜ) is modeled in MT, whereas density \(\sigma_{PL}\) is in T/m³. Assuming spherical particles, the number of particles (NMAₜ, NMIₜ) then is:

\[ N_{MA,t} = \frac{V_{MA,t}}{V_{Particle}} \text{ and } N_{MI,t} = \frac{V_{MI,t}}{V_{Particle}} \]  (S2)

where \(V_{Particle}^{MA}\) and \(V_{Particle}^{MI}\) are the volumes of the individual particles (m³). With the sphere volume \(V = \frac{4}{3} \pi r^3\) for individual particles and a combination of Equations (S1) and (S2) this gives:

\[ N_{MA,t} = \frac{MA_t 10^6}{\frac{4}{3} \pi r_{MA}^3 \sigma_{PL}} \text{ and } N_{MI,t} = \frac{MI_t 10^6}{\frac{4}{3} \pi r_{MI}^3 \sigma_{PL}} \]  (S3)

Equation (S3) expresses the total number of macro- and microplastic spheres in the OSL as a function of macro- and microplastic total mass in the OSL, radius, and plastic density. Because the sphere surface is \(A = 4\pi r^2\), the total surface area of these numbers of spherical particles can be obtained by multiplication \((A_{MA,t} = 4\pi r_{MA}^2 N_{MA,t})\) and \((A_{MI,t} = 4\pi r_{MI}^2 N_{MI,t})\), which yields:

\[ A_{MA,t} = \frac{3MA_t 10^6}{\sigma_{PL} r_{MA}^2} \text{ and } A_{MI,t} = \frac{3MI_t 10^6}{\sigma_{PL} r_{MI}^2} \]  (S4)

Equation S4 provides the total surface areas of macro- and microplastic particles if they were perfect spheres. Because the actual particles will be non-spherical, the calculated areas are corrected for non-sphericity of the particles through multiplication with a correction factor \((\alpha_{MA}, \alpha_{MI})\):
A representative radius for microplastic particles $r_{MI}$ was estimated as follows. Abundance data for two microplastic size classes (0.33-1 and 1.01-4.75 mm) as provided by Eriksen et al. (2014) were combined to calculate one surface equivalent average size for microplastic. We used these data to be consistent with the abundance data used in the modeling that also were taken from this study. First, size ranges as reported were approximated by a median particle size (0.0006 and 0.003 m, Table S3). For all particles with this size, a total surface area was calculated using reported number count and surface area per particle. Then total number count and total surface area of the two reported size classes were combined to calculate an average surface area per particle. From the average surface area per particle, an average radius was calculated assuming spherical particles. This way the total surface area for microplastic that follows from the data was preserved, albeit assigned to one single average size class for microplastic ($r_{MI}= 1.2 \times 10^{-3} \text{ m}$, Table S3). For macroplastic, size classes of 4.76-200 mm and > 200 mm were reported. Because no upper limit was reported, a similar calculation is not possible for macroplastic. Therefore, for macroplastic an intermediate value of $r_{MA} = 0.1 \text{ m}$ was selected.

### Table S3. Calculation of a surface equivalent monodisperse size for microplastics (data from Eriksen et al.\cite{Eriksen2014})

| Size class (mm) | Selected representative size (m) | Number count | Surface area (m²) | Total Number count (both sizes) | Total Surface area (m²) (both sizes) |
|----------------|-------------------------------|--------------|--------------------|-------------------------------|-------------------------------------|
| 0.33-1         | 0.0006                        | 1.83E+12     | 2.070E+06          | 4.85E+12                      | 8.746E+07                           |
| 1.01-4.75      | 0.003                         | 3.02E+12     | 8.539E+07          |                               |                                     |

| Surface per particle | Radius per particle |
|----------------------|---------------------|
| 1.8033E-05 m²        | 0.001198 m          |
|                       | 1.2 mm              |
Extending the model to include $<0.335$ mm particles (‘nanoplastic’)

The masses of macro- and microplastic are modeled with Equations 1 and 2 respectively. For ‘nanoplastic’ in the OSL (defined here as plastic particles $< 335$ µm), a third differential equation can be defined. Nanoplastics in the OSL ($NP_t$) is assumed to originate from primary sources and from fragmentation of $0.335 – 5$ mm microplastic and is assumed to be removed from the OSL by agglomeration - or heteroaggregation-sedimentation. If aggregation is sufficiently fast, the latter process can be approximated by first order removal (Quik et al, 2014, whereas the settling aggregates can be assumed to have the same apparent settling rate as microplastics (Velzeboer et al, 2014), with settling rate $V_S^{MI}$:

$$\frac{dN_P}{dt} = A \times B \times D \times P_t + k_F A_{MI,t} M I_t - \frac{V_S^{MI}}{H} N P_t \quad (S6)$$

A simulation with the model including Equation S6, shows a steadily increasing nanoplastic mass in the OSL, with 5430 tons estimated for 2010 and 7750 tons for 2016 (Figure S1).

**Figure S1**: Macroplastic (MA, $\circ$) and Microplastic (MI, $\circ$) like in Figure 2A, with subsequent nanofragmentation of the microplastic to $<0.335$ mm Nanoplastic (NP, $\circ$) modeled according to Equation S6 and parameters as in Table 1. To improve the readability of the curve for Nanoplastic, a log scale is used for the Y-axis. According to the simulation, the 2010 quantity of nanoplastic in the OSL would be 5430 tons. For micro- and macroplastic, these simulated quantities are 36 340 tons and 233 300 tons, respectively, equal to the quantities reported by Eriksen et al, 2014 (indicated with marker). The dashed areas indicate the propagated model uncertainty expressed as $\pm 1$ SD.
Supplementary Material Uncertainty analysis

Our results are based on parameter values taken from literature data and/or from best educated guesses. However, these parameters as well as the measured plastic abundance data are uncertain. Therefore, the primary calculations were performed again, now using parameter sets randomly sampled from predefined distributions of parameter values, each of which quantifies the uncertainty in that parameter value. This probabilistic assessment of model uncertainty was performed in two steps.

Step 1. Distributions of the unknown parameters $k_F$ and $V_{SMI}^*$ were obtained by fitting them to 5000 sets of parameters / measured data randomly drawn from preset distributions as defined and motivated in Table S4. The resulting frequency distributions were log-triangular distributed between -7.3 and -5.7 for $k_F$ (Figure S2), and log-normal distributed for $V_{SMI}^*$ (Figure S3).

![Histogram of log10(kf)](image)

**Figure S2.** Distribution of log $k_F$ values obtained from n=5000 model fits of $k_F$ and $V_{SMI}^*$ to plastic debris abundance data while drawing parameter values from the distributions of model parameters as specified in Table S4.
Step 2. Distributions for predicted MA, MI, NP and LP were calculated for 1950 – 2016 by Monte Carlo model simulations (10 000 iterations) using the same preset parameter distributions (Table S4). The distributions for $V_s^{MI}$ and $k_F$ were obtained from the uncertainty estimates for these parameters obtained from step 1. Uncertainties in the resulting abundances of MA, MI and NP were presented as ± 1 SD (Figure S2).
| Parameter | Value; range/distribution | Comment / Motivation |
|-----------|---------------------------|----------------------|
| $P_t$     | $P_t = 0.0843t^2 - 0.8015t + 3.0191$ | Annual production data show excellent fit to 2nd order polynomial model (Koelmans et al, 2016 ). Uncertainty was assumed small compared to other sources of uncertainty. |
| A         | 0.03; range 0.017-0.046; triangular distribution | Value and range provided by Jambeck et al (2015) |
| B         | 0.65; range 0.55-0.75, uniform distribution | The default value of 0.65 is based on production data for plastic with density lower than that of seawater, i.e. buoyant plastic. However, thin PE/PP film macroplastic (bags & sheets) with density lower than seawater might still rapidly foul and sink, in which case B=0.65 could be too high. On the other hand, some higher than water density microplastics may not sink and reside in the Ocean Surface Micro Layer, in which case B=0.65 would be too low. The percentages of thin films produced for LDPE/LLDPE, HDPE and PP are 35.4, 15 and 8.9 % (plastics.americanchemistry.com). Accounting for this would yield a ~ 0.1 lower buoyancy fraction. Based on these data, uncertainty was defined as 0.1, uniform in both directions. |
| C and D   | 0.005; range 0.0025 – 0.01; triangular distribution | The fraction of primary, buoyant microplastic particles < 4.75 mm was estimated as 0.01 with uncertainty 0.005 – 0.02. This fraction however, also includes a fraction <0.335 mm, which in this paper is referred to as nanoplastic. No accurate data on the relative proportion of masses in the size fractions < 0.335 and 0.335-4.75 mm is available, which is why we assigned them equal share (0.005, range 0.0025-0.01). |
| $r_{MA}$  | 0.1 m | Based on measured data, uncertainty in role of radius on estimate of plastic surface area is accounted for through uncertainty in $\alpha$ |
| $r_{MI}$  | 0.0012 m | Based on measured data, uncertainty in role of radius on estimate of plastic surface area is accounted for through uncertainty in $\alpha$ |
| $\alpha_{MA}$ | 30; SD=7.5, normal distribution with delimiters at values 5 and 55. | An SD of 7.5 implies that ~ 95% of the sampled values are between 15 and 45, which is considered a good representation of the range in Table S2. Delimiters are used to prevent $\alpha_{MA}$ to take values defined for microplastics, and/or to become |
An SD of 0.5 implies that ~95% of the sampled values are between 1 and 3, which is considered a good representation of the range in Table S3. Delimiters are used to prevent $\alpha_{MI}$ to take values $<1$, which are by definition not possible because 1 represents sphericity, and $>3$, which would cause an asymmetrical distribution and would imply that values are sampled that represent macroplastics.

Shape of distribution and concomitant boundaries were obtained from Step 1, see Figure S2.

Shape of distribution and concomitant SD were obtained from Step 1, see Figure S3.

| Abundance data | Value; range/distribution | Comment / Motivation |
|----------------|---------------------------|----------------------|
| MA in 2010     | 0.2334 MT; log-normal distribution, SD=0.21 | Uncertainties in measured data were obtained from L. Lebreton, based on the data analysis in Eriksen et al, 2014. |
| MI in 2010     | 0.03554 MT; log-normal distribution, SD=0.70 | Uncertainties in measured data were obtained from L. Lebreton, based on the data analysis in Eriksen et al, 2014. |
Macro- micro- and < 0.335 mm ‘nanoplastic’ concentrations at constant plastic annual production rate: Steady state solution

Over the past decades, the total production of plastic has been increasing following a second order polynomial (Koelmans et al, 2016; PlasticsEurope, 2013), which is incorporated in the model. However, if annual plastic production were to become constant ($P_t = P_{SS}$), an analytical solution to the model can be calculated, which provides the concentrations of macro-, micro, and nanoplastic at steady state ($MA_{SS}$, $MI_{SS}$, $NP_{SS}$).

**Macroplastic** - The steady state solution to Equation 1 is:

$$MA_{SS} = \frac{A_{MA} \times B \times (1-C) \times P_{SS}}{k_F A_{MA,SS} + \frac{V_{MA}}{H}}$$

in which $A_{MA,SS} = \frac{3MA_{SS} \times a_{MA} \times 10^6}{\sigma_{PLTMA}}$ (S7)

Elimination of $A_{MA,SS}$ yields a quadratic equation with the steady state solution for $MA_{SS}$:

$$MA_{SS} = \frac{-V_{MA} + \sqrt{V_{MA}^2 + 4 \left( \frac{3k_F A_{MA} \times B \times (1-C) \times P_{SS} \times 10^6}{\sigma_{PLTMA}} \right)}}{2k_F \times 3a_{MA} \times 10^6}$$

(S8)

**Microplastic** – The steady state solution to Equation 2 is:

$$MI_{SS} = \frac{A \times B \times C \times P_{SS} + k_F A_{MA,SS} \times MA_{SS}}{k_F A_{MI,SS} + \frac{V_{MI}}{H}}$$

(S9)

Elimination of $A_{MI,SS}$ yields a quadratic equation with the steady state solution for $MI_{SS}$:

$$MI_{SS} = \frac{-V_{MI} + \sqrt{V_{MI}^2 + 4 \left( \frac{2k_F A_{MI} \times B \times C \times P_{SS} + k_F A_{MA,SS} \times MA_{SS} \times 10^6}{\sigma_{PLTMI}} \right)}}{2k_F \times 3a_{MI} \times 10^6}$$

(S10)

in which $A_{MA,SS} = \frac{3MA_{SS} \times a_{MA} \times 10^6}{\sigma_{PLTMA}}$ (main manuscript, Equation 3) and $MA_{SS}$ is given by Equation S8.

**Nanoplastic** - The steady state solution to Equation S6 is:

$$NP_{SS} = \frac{A \times B \times D \times P_{SS} + k_F A_{MI,SS} \times MI_{SS}}{V_{MI} \times H}$$

(S11)

in which $A_{MI,SS} = \frac{3MI_{SS} \times a_{MI} \times 10^6}{\sigma_{PLTMI}}$ (main manuscript, Equation 3) and $MI_{SS}$ is given by Equation S10.
Figure S4. Sensitivity of the model output (simulated mass of macroplastic and microplastic in the OSL in 2010), to a 10% change in each of the parameters $A$, $B$, $C$, $D$, $H$, $r_{MA}$, $r_{MI}$, $\alpha_{MA}$, $\alpha_{MI}$, $k_F$ and $V^M_S$. 


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