

Export of Plastic Debris by Rivers into the Sea

Christian Schmidt,^{*,†,‡} Tobias Krauth,^{†,‡} and Stephan Wagner[§]

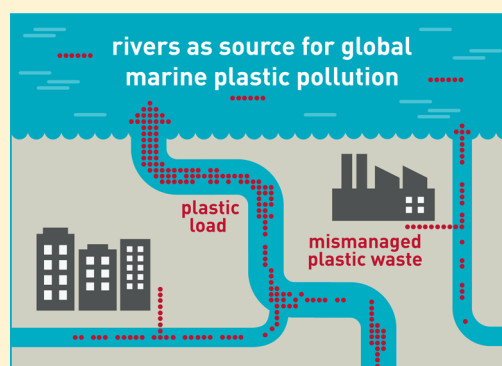
[†]Department of Hydrogeology, Helmholtz-Centre for Environmental Research - UFZ, Permoserstrasse 15, 04318 Leipzig, Germany

[‡]Department of Environmental Engineering, University of Applied Sciences Weihenstephan-Triesdorf, Markgrafenstrasse 16, 91746 Weidenbach, Germany

[§]Department of Analytical Chemistry, Helmholtz-Centre for Environmental Research - UFZ, Permoserstrasse 15, 04318 Leipzig, Germany

S Supporting Information

ABSTRACT: A substantial fraction of marine plastic debris originates from land-based sources and rivers potentially act as a major transport pathway for all sizes of plastic debris. We analyzed a global compilation of data on plastic debris in the water column across a wide range of river sizes. Plastic debris loads, both microplastic (particles <5 mm) and macroplastic (particles >5 mm) are positively related to the mismanaged plastic waste (MMPW) generated in the river catchments. This relationship is nonlinear where large rivers with population-rich catchments delivering a disproportionately higher fraction of MMPW into the sea. The 10 top-ranked rivers transport 88–95% of the global load into the sea. Using MMPW as a predictor we calculate the global plastic debris inputs from rivers into the sea to range between 0.41 and 4×10^6 t/y. Due to the limited amount of data high uncertainties were expected and ultimately confirmed. The empirical analysis to quantify plastic loads in rivers can be extended easily by additional potential predictors other than MMPW, for example, hydrological conditions.



1. INTRODUCTION

Pollution of the marine environment with plastic debris is widely recognized and is of increasing ecological concern¹ because of the chemical persistence of plastics and their mechanical fragmentation to so-called microplastics,² which can be ingested by even small organisms such as zooplankton.³ Beyond the long recognized occurrence of plastic debris in the marine environment, plastic debris has been more recently detected in freshwater environments and can be found even in pristine, remote locations.⁴ The plastic pollution of freshwater systems, particularly rivers and of the marine environment is interlinked because rivers ultimately discharge into the marine environment. So far land-based inputs into the sea have been attributed to a 50 km⁵ and 200 km⁶ wide coastal zone. Coastal inputs have been assumed to be proportional to the amount of mismanaged plastic waste (MMPW) generated in that coastal zone but rivers have not been explicitly accounted for.⁷ This implicitly assumes that further inland plastic sources remain unconnected to the sea. Undoubtedly, most of the global population lives in coastal areas⁷ but river networks may facilitate transport of plastic debris over long distances into the sea, as it is has been shown for terrestrial sediments, organic carbon,⁸ nitrogen,⁹ and various other solutes. Thus, rivers connect most of the global land surface to the marine environment. Since land-based sources are considered to be a major contributor to marine plastic debris,⁵ rivers are a major pathway for plastic transport into the seas. Recently, Lebreton

et al. (2017)¹⁰ estimated that ~3–19% of the costal plastic emissions are facilitated by river transport and additional ~0.8–1.5 × 10⁶ t/y reach the ocean from inland areas.

Plastic loads and concentration in rivers depend on the characteristics of the catchment. Urban land use and population density have been shown to be positively related to plastic concentrations.¹¹ However, our data and knowledge base is still insufficient to link the sources, transport pathways, and fate of plastic debris in both marine and freshwater environments. This understanding is required to assess the plastic pollution in the aquatic environment and to develop efficient prevention strategies for pollution. One prominent example is the so-called “missing plastic” problem—the mismatch between the large estimated plastic inputs into the sea and the low amount actually observed.²

The aim of the study is to compile available data of plastic debris in rivers from available data, to identify patterns of plastic concentrations and loads and to provide an estimate of the amount of plastic exported from river catchments into the sea assuming that the entire river catchment is connected to the coastal sea via the river network. We combine plastic concentrations in rivers with the amount of MMPW generated

Received: May 8, 2017

Revised: August 17, 2017

Accepted: September 11, 2017

Published: October 11, 2017

in the catchments. This approach is similar to a recently published approach by Lebreton et al. (2017).¹⁰ Here, we compiled a larger data set and treated microplastic (particles <5 mm) and macroplastic (particles >5 mm) separately. Both approaches provide empirical frameworks to predict plastic inputs via rivers into the sea.

2. MATERIALS AND METHODS

2.1. Compilation of the River Plastic Data Set. Peer reviewed papers were systematically searched in Web of Science on Apr. second 2017 applying the key word combinations “plastic AND stream” and “plastic AND river”. These search terms were intentionally kept broad to ensure that no relevant studies are overlooked. We scanned through the titles of 1870 search results and extracted 73 articles that deal with aquatic systems. From these articles full texts were obtained and assessed if they reported plastics in streams and rivers and if the plastics had been sampled from the water column. We included studies reporting particle and/or mass concentrations or loads regardless of the size fraction considered (e.g., microplastics only, various size fractions or total plastics only). Given the limited observational data, our strategy was to include as many data as possible, although the diverse sampling and analysis techniques certainly limit the comparability among data sets. We focused our search on data from the water column as we were interested in the fraction of plastic that is mobile in river systems and not retained in sediments. Data from sampling on beaches, river banks, and river bed sediments have thus not been considered. Also data from estuaries were excluded because discharge cannot be obtained and the tidal mixing with seawater may result in a mixing of plastic particles that originate either from the river or from the sea.

Additional data from two reports and from an article in a nonpeer reviewed journal was discovered in a web search and a search within cited references. In total data from the 10^{11–20} peer-reviewed articles and three^{21–23} nonpeer-reviewed publications have been extracted for further analysis. Either stationary drift nets (hand-held or floating) or drift nets attached to a vessel comprised the vast majority of sampling techniques. In one study¹⁷ particles were extracted by pumping and subsequent sieving. The typical mesh size was 300–500 μm , the minimum was 50 μm ,¹⁷ 100 μm mesh size was used by.¹⁴ In order to ensure comparable size distributions the smallest fractions in^{14,17} have been omitted from further load estimations. The dimensions of the net openings did not exceed 1 m width, height was between 0.1 and 0.5 m. One study used channel spanning retention booms.¹³ Mesh size and net dimensions for each reference are provided in [Supporting Information \(SI\) Data S1](#).

Data reported in units of particle or mass “per area” were converted to “per volume” by dividing with the submerged height of the net. This approach assumes that plastic particles are uniformly distributed across the water column. If river discharges have not been obtained in conjunction with the plastic sampling, missing discharges were extracted from available data, for example, from online database from the closest gauging station or by contacting the corresponding author.

We grouped the particle sizes into two classes: “microplastic” which comprise particles of 5 mm and below and “macroplastic” which are all particles larger than 5 mm in diameter. A third class “total plastics” was introduced to account for data from studies which reported total plastics only. Data from

studies that reported micro- and macroplastic concentrations are considered separately in the micro- and macroplastic size class and as sum of the two fractions in the total plastic class. We selected 5 mm as size cutoff because it is typically considered as microplastic and studies focused on microplastic frequently do not report larger particle sizes.

Data from studies that exclusively reported particle concentrations have been converted to masses and mass concentrations. For this we used studies reporting both particle concentration and mass concentrations and calculated the arithmetic mean of particle masses for microplastic and macroplastic, respectively. All data are provided in the [SI \(Data S1\)](#).

2.2. Estimation of Plastic Loads. The load of a solute or particulate matter can be estimated from concentration $C(t)$ and discharge time series $Q(t)$ (eq 1).

$$L_{\tau} = \int_0^{\tau} C(t)Q(t)dt \quad (1)$$

where L_{τ} is the load over time interval τ . By applying eq 1 we assumed that the observed concentrations are uniform and representative for the entire river water column. Further, we assumed that all plastic particles are transported in the water column and transport with the sediment bedload is negligible. We used concentrations in units of mass per volume resulting in loads as mass per time. eq 1 could be equivalently applied to particle count data. Because river discharge and plastic concentrations have been rarely measured simultaneously and with high, quasi continuous frequency, loads were estimated from the available, discrete and sparse data via a variety of methods.²⁴ Currently, we are not aware that regular monitoring programs have been established. All load estimates have thus to rely on data from single sampling campaigns, typically covering not more than three or four consecutive measurements over the course of a few months. Loads were estimated from the instantaneous plastic concentration C_i and weighted by the instantaneous discharges Q_i and the mean discharge \bar{Q} to account for bias introduced by sampling at very high or low discharges (eq 2).

$$L = \frac{\sum_n^i (C_i Q_i)}{\sum_n^i Q_i} \bar{Q} \quad (2)$$

Equation 2 was applied separately to the three size classes “microplastic”, “macroplastic” and ‘total plastic’ to estimate the loads for each of them. When only mean discharges were available loads have been estimated from mean discharge and the mean concentration. Single concentration measurements at one point in time were combined with mean discharges.

2.3. Catchment and Population Data. The river network and the global catchment boundaries have been taken from the HydroSHEDS data sets²⁵ (<http://hydrosheds.cr.usgs.gov>). We used 1494 catchments with an outlet to the sea as provided by Milliman & Farnsworth (2011),²⁶ regardless of their size. For each sampling point in the plastic data set, the upstream catchments were estimated from the hydrologically conditioned digital elevation model distributed with the HydroSHEDS data set which is based on data from the Shuttle Radar Topography Mission. Global population was taken from the 2010 data set Gridded Population of the World (GPWv3) (<http://sedac.ciesin.columbia.edu/data/collection/gpw-v3>).²⁷ Population data are provided on a grid with 2.5 arc-minutes resolution. This data set was intersected with the catchments boundaries to

Table 1. Summary of Particle and Mass for Instantaneous Concentrations

particle concentration	median [particle/1000m ³]		mean [particle/1000m ³]		standard dev. [particle/1000m ³]	
	with 0s	without 0s	with 0s	without 0s	with 0s	without 0s
MicP	13.4	18.8	37 659.7	38 208.1	272 998.1	274 950.1
MacP	0.01	0.5		3154.7	16 945.1	22 819.1
mass concentration	median [g/1000m ³]		mean [g/1000m ³]		standard dev. [g/1000m ³]	
	with 0s	without 0s	with 0s	without 0s	with 0s	without 0s
MicP	0.6	0.6	298.6	298.6	1760.5	1760.5
MacP	0.3	0.4	864.7	933.9	5461.3	5674.0

estimate the population in each catchment. Country boundaries from the GPWv3 data set were used throughout the study.

2.4. Estimation of Mismanaged Plastic Waste Generation. To estimate the amount of MMPW generated per person and time (kg/person/day) in each catchment, we extended the data from Jambeck et al. 2015⁵ which contained 192 coastal countries (including sovereign, UN – listed states and other independent territories) by 41 countries which have a share on river catchments that discharge into the sea. Plastic waste is considered to be mismanaged if it is inadequately disposed or littered. The waste generation rate and the fraction of plastic in the waste stream for these countries have been taken from Hoornweg and Bhada-Tata, (2012).²⁸ To estimate the fraction of plastic waste that is mismanaged in the 41 additional countries, we applied the average values of MMPW for each World Bank economic classification²⁹ (HIC = high income; UMI = upper middle income; LMI = lower middle income; LI = low income, see SI Data S1) taken from the countries covered in.⁵ The country-based data were intersected with the catchment boundaries of global river catchments²⁶ and the global population data set.²⁷ The MMPW generation rate in each catchment was estimated from the area-weighted average of the country-based population and the MMPW generation per capita. The same procedure was applied to estimate the MMPW generated in the catchments upstream of each observation location. MMPW per person and time for each country and catchment are provided in SI Data S1.

2.5. Regression Analysis. An initial evaluation of the data revealed a nonlinear relationship between MMPW and L . To account for the nonlinearity we use a power-law model. The regression coefficients b_1 (slope) and b_0 (intercept) were estimated by linear least-squares regression of $\log_{10}(\text{MMPW})$ and $\log_{10}(L)$ (eq 3).

$$\log_{10}(L) = b_0 + b_1 \log_{10}(\text{MMPW}) \quad (3)$$

Particularly when the regression was extrapolated for estimating global plastic loads into the sea, uncertainties propagate into the estimation results. To address the underlying uncertainty we performed bootstrapping by case resampling for the regressions of L and MMPW and estimated b_1 and b_0 , for 5000 bootstrap samples. Then the 5000 combinations of b_1 and b_0 were applied to the global MMPW data set to generate a range of plausible values for the global plastics loads.

2.6. Global Estimation of Plastic Loads. To estimate the plastic load delivered by rivers into the sea we apply the regression models of MMPW and loads to the global river data set. Because of the inherent uncertainty in the underlying data sets we use two different approaches to estimate the global plastic loads. “Model 1” is based on the microplastic data set which includes all microplastic measurements. “Model 2” takes data sets into account where both microplastic and macroplastic fraction were measured. The macroplastic data is the

same in both models. Summing the microplastic and macroplastic models yield estimates the total mass of plastic debris discharged per time regardless of its size. A graphical overview on the methodological concept, the data set and the data flow of the analysis is provided in SI Figure S1.

3. RESULTS

3.1. Data Set. The analyzed data set of plastic in rivers consists of 240 individual samples, at 79 sites covering 57 rivers. The sampled rivers are not uniformly distributed across river sizes. Plastics have been predominantly sampled either in small rivers (mean discharge $< \sim 50 \text{ m}^3/\text{s}$) or in larger rivers with mean discharges $> \sim 1000 \text{ m}^3/\text{s}$. An explanation for the underrepresentation of medium-sized rivers could be that small rivers are easily wadeable, whereas large rivers can be sampled by trawls with vessels adapted from marine sampling. Population in the catchments ranged between ~ 1900 and 309×10^6 inhabitants.

3.2. Observed Concentrations. Microplastic particles were detected in 98.5% of the samples with mean and median concentrations with zero detections excluded of $\sim 38\,200$ and ~ 19 particles/1000 m^3 and zero detections included of $\sim 37\,700$ and ~ 13 particles/1000 m^3 , respectively (Table 1). The range of concentrations varied over almost 8 orders of magnitude (Figure 1a).

Macroplastic particles were detected in 55% of the samples. We attribute this relatively large number of zero detection to the sampling design which is not specifically adapted to sample the relatively low counts of macroplastic. Relatively small nets of ~ 0.25 to 0.5 m^2 cross-sectional and short exposure times ($< 1 \text{ h}$) may not be appropriate to representatively capture macroplastic concentration. We thus report both, concentrations with and without zero detection. For the subsequent calculation of loads we used the data with zero detection included. Depending on whether zeros are included in the calculation of mean and median, the resulting concentrations differ. Excluding zeros from calculation the mean and the median concentration was about 3000 and 0.5 particles/1000 m^3 and thus approximately 1 order of magnitude lower than for microplastic. The observed range of concentration also spanned more than 7 orders of magnitude. The total plastic particle concentration comprises in average of 95% microplastic particles (Figure 1d).

Mass concentrations have been obtained less frequently in comparison to particle counts. From the total 240 samples only $\sim 20\%$ have been reported mass concentrations. Studies providing masses detected microplastic in all samples. Median concentration of microplastics was 0.6 g/1000 m^3 . Mean mass concentration was ~ 300 g/1000 m^3 , also indicating a right-tailed distribution (Table 1).

Macroplastic mass concentrations revealed a similar range with a median of 0.3 and 0.46 g/1000 m^3 with and without zero

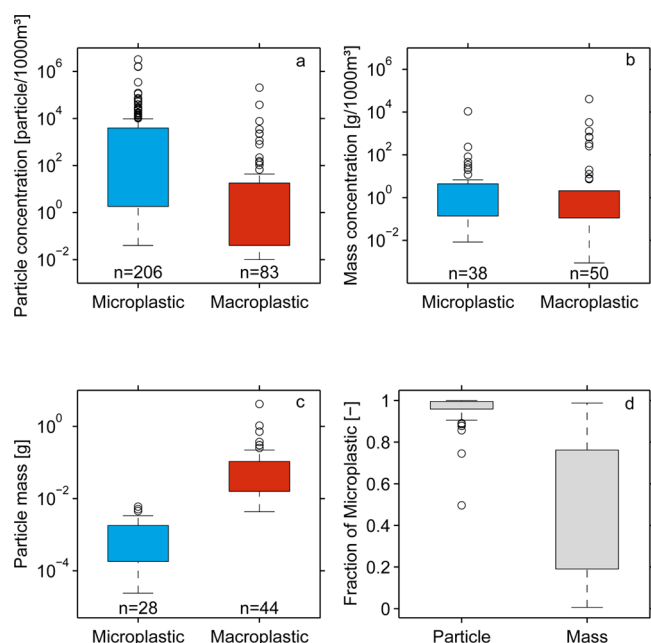


Figure 1. Boxplots of particle concentration (a) and mass concentration (b), masses of single particles (c) for microplastic and macroplastic. (d) shows the fraction of microplastic in terms of particles and mass. In each boxplot the horizontal line represents the median the box the 25th and 75th percentile, dots are data outside the edges of the box are the 25th and 75th percentiles, the whiskers represent 1.5 times the range between the 25th and 75th percentiles and dots are outside this range.

detection, respectively. Mean mass concentrations ranged between 870 and 930 g/1000 m³, depending on the inclusion of zero detection. Macroplastic masses were detected in 93% of the macroplastic samples.

The fraction of microplastic mass concentration in total plastic varied strongly between samples. A few larger particles contribute a large fraction of the total plastic concentration. The mean mass fraction of microplastics in the samples was 0.48 (median 0.47) ($n = 33$).

The average particle masses of micro- and macroplastic have been estimated from studies where both mass and particle concentrations were available (Figure 1c and SI Data S1). The mean mass of microplastic particles is 0.0018 g and for macroplastic 0.2226 g. These values have been used further to estimate mass concentrations.

3.3. Plastic Export from River Catchments. Catchment area and average discharge covered by the data set range from 15 km²–1.46 × 10⁶ km² (microplastic), 101 km²–8.2 × 10⁵ km² (macroplastic), 15 km²–8.2 × 10⁵ km² (total plastic) and 0.5 m³/s to 2.5 × 10⁴ m³/s (microplastic), 1.2 m³/s to 6500 m³/s (macroplastic), 0.0450 m³/s to 6500 m³/s (total plastic). The overall MMPW generation in the individual catchments ranges between 4.7 t/y and 10.4 × 10⁶ t/y. Specific MMPW generation (MMPW generated per unit area and time) ranges between 14 kg/y/km² and 10.7 t/y/km².

We evaluated the loads separately for microplastic, macroplastic and total plastics because they comprise different, yet partially overlapping subdata sets. Loads are reported in terms of masses. Observed annual loads in the individual catchments range over 10 orders of magnitude from 0.18 kg/y to 2.3 × 10⁶ t/y for microplastic, 0.13 kg/y–1400 t/y for macroplastic, and 0.18 kg/y–1800 t/y for total plastic. Note that e.g. for the

Yangtze characterized by the highest observed microplastic loads, no data on macroplastic were available which explains the different range of micro- and macroplastic loads.

Specific loads provided insight into the plastic transport independent from the catchment size by normalizing both the load and MMPW with the catchment area.

The observed specific loads (load per unit catchment area) cover a wide range between $\sim 0.17 \times 10^{-7}$ and 1.6 (t/y/km²) for microplastic and $\sim 0.12 \times 10^{-7}$ and 0.6 (t/y/km²) for macroplastics (Figure 2d–e). Particular low specific loads have been observed for the tributaries to the Great Lakes.⁹ Maximum specific loads were observed for the Yangtze River (1.6 t/y/km²) close to Wuhan.¹⁵ Notably, the second highest specific loads have been found the Hanjiang River (1.1 t/y/km²) in the same study. For macroplastic the San Gabriel river, a highly urbanized river crossing Los Angeles shows the highest specific loads.¹⁹

The observed plastic loads are generally positively related to the MMPW generated annually in the catchment. MMPW is a good predictor for microplastic loads ($r^2 = 0.78$). Also the relationships for macroplastic and total plastic are highly significant, yet with lower r^2 (Figure 2a–c). A similar behavior can be observed for the specific loads and their relationship to the area weighted MMPW generation. The slopes of the linear regressions are consistently larger than 1 (Figure 2d–e) indicating that an increasing fraction of MMPW, both total and area weighted, is found being transported in rivers with increasing MMPW generation (Figure 2a–e).

To explore this further we introduce a metric which can be referred to as plastic delivery ratio. It is the ratio between the observed plastic loads in rivers and MMPW generated in the catchment. This is conceptually the same as the conversion rates of mismanaged plastic waste to marine debris as applied in Jambeck et al. 2015⁷. The observed delivery ratios range between $\sim 5 \times 10^{-7}$ and 0.23 for microplastic, $\sim 1 \times 10^{-6}$ and 0.06 for macroplastic and $\sim 2 \times 10^{-6}$ for total plastic.

The delivery ratio tends to increase with increasing total MMPW generation (Figure 2g–i). This effect is particularly notable for microplastic while macroplastic and total plastic show also a positive, yet less steep and insignificant relationship at an $\alpha = 0.05$ level (p values: macroplastic 0.077; total plastic 0.13). Mean and median of the delivery ratio are 1.3×10^{-2} and $\sim 9 \times 10^{-5}$ for microplastic, 3×10^{-3} and 6×10^{-5} for macroplastic and 3×10^{-3} and 1×10^{-4} for total plastics.

3.4. Global Plastic Inputs from Rivers into the Sea.

From the 1494 rivers and the data set of Milliman and Farnsworth, 2011,²⁶ we analyzed the 1350 rivers where the estimated MMPW generation was nonzero. The rivers in the data set range from small first order streams to large rivers which all discharge to the sea. The catchment sizes ranged between <1 and 5.9×10^6 km² and total population in the catchments between 0 and 621 million. Drawing from the classification scheme of Jambeck et al. 2015⁵ there is an amount of approximately 76 million tons/y of plastic waste generated in these catchments which is mismanaged and potentially available for transport into and within river systems.

To estimate the plastic loads we use two different regression models which yield different results pointing on the uncertainty of the underlying data. “Model 1” comprises all microplastic measurements. “Model 2” takes only data sets into account where both microplastic and macroplastic have been measured in conjunction. The macroplastic data is the same in both models. Summing the each microplastic model and with the

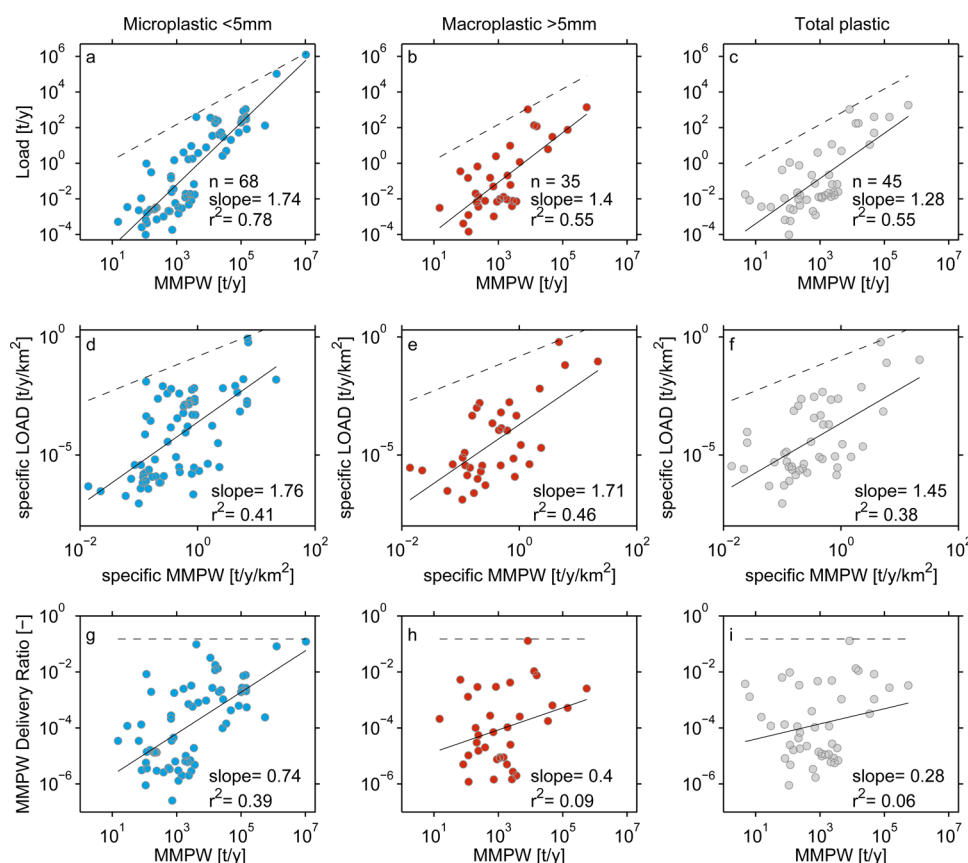


Figure 2. Regression of MMPW generated in the catchments and measured plastic loads (a–c) and regression of specific (area-weighted) load and MMPW (d–f) for micro-, macro and total plastic. The bottom row shows the relationship between MMPW and the MMPW delivery ratio. Dashed lines represent a hypothetical proportional model where 15% of the MMPW to illustrate the disproportionality of plastic loads associated with larger MMPW-rich catchments.

macroplastic model yield estimates the total mass of plastic debris discharged per time regardless of its size.

Model 1 yields a median of 0.16×10^6 t/y of microplastic with 25 and 75% prediction intervals of 0.06×10^6 and 0.45×10^6 t/y, respectively. Model 2 yields a global median of microplastic load of $\sim 2.31 \times 10^6$ t/y, with 25 and 75% prediction intervals of 1.41×10^6 and 3.73×10^6 t/y. Macroplastic loads are estimated to be at 0.15×10^6 t/y (25 and 75% prediction interval: 0.051×10^6 and 0.44×10^6 t/y) (Figure 3).

The resulting total plastic loads differ based on the input data. Model 1 results in a median total load of 0.47×10^6 t/y with (25 and 75% prediction interval: 0.21×10^6 and 1.12×10^6 t/y). Model 2 yields 2.75×10^6 t/y as median estimate with a 25 and 75% prediction interval of 1.72×10^6 and 4.38×10^6 t/y (Figure 3).

The different underlying data sets do not have only consequences for the total estimates but also for the spatial distribution of plastic inputs into the sea. The spatial distribution of plastic export from rivers is highly nonuniform because of the skewed distribution of river and catchment sizes. Because spatially nonuniform plastic delivery ratios tend to increase toward larger, population rich catchments, this disproportional pattern is even amplified. For Model 1 the top 10 catchments contribute 88% of the total estimated load and 94% for Model 2. For comparison, a spatially uniform delivery ratio would result in 58%. Of the 10 catchments delivering the highest loads to the ocean, 8 are located in Asia,

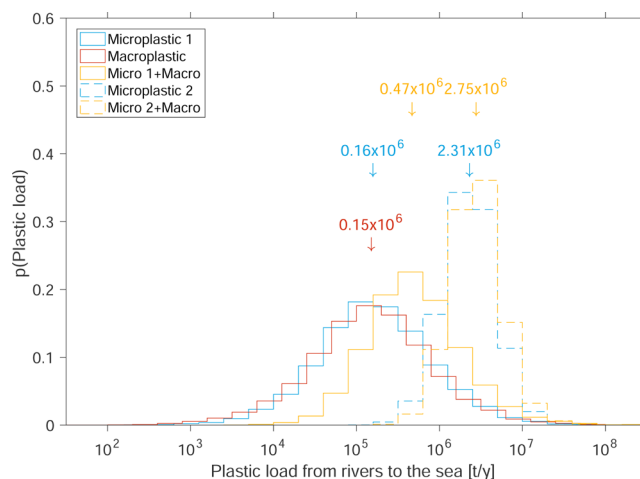


Figure 3. Probability of global microplastic loads for Model 1 and Model 2, macroplastic loads and their combinations. Text insets depict the median values.

with mostly middle-income countries²⁶ such as China (SI Data S1, Table S1), where high rates of MMPW generation prevail.

4. DISCUSSION

The enormous concentration variability of ~ 7 (macroplastic) to ~ 8 (microplastic) orders of magnitude (mass concentrations) is not unusual for particulate matter and has been observed for suspended sediments³⁰ and coarse particulate

organic matter.³¹ Like concentration in rivers, the plastic concentrations at the sea-surface vary over a wide range covering 4 orders of magnitude.³² High particle concentrations have been found in the North Atlantic with up to 580 000 particles/km²³³ equivalent to 1160 particles/1000 m³ assuming 0.5 m submergence of the trawl net. Mass concentrations have been reported to be up to 8000 g/km² or 16 g/1000 m³.³² Thus, mean concentration in rivers is roughly 40–50 times higher than the maximum concentration observed in the open ocean.

Observed differences between concentrations in rivers and in the seas, as well as among different rivers likely arise also from differences in the sampling program design, sample collection and analysis methods and target variables such as particles or mass or the size and type classification. Generally, reporting masses and mass distributions of particles would provide a more unbiased, conservative measure of plastic debris, for example, if large plastic debris is fragmented, the sum of the fragment masses is constant. Moreover, the abundance of plastic in rivers would ideally be reported in conjunction with discharge in order to derive concentration and load. Both are absolutely crucial for establishing mass balances, comparison between sites and for understanding fragmentation.³⁴ Despite these uncertainties, the presented empirical quantification of plastic loads in rivers provides a testable framework which can be extended to other catchment properties such as land use or hydrologic conditions than the amount of mismanaged plastic waste alone, for further assessing plastic transport and accumulation in rivers worldwide.

Microplastic and macroplastic loads are related to the MMPW generation in the catchments. The steeper slopes for microplastic of the log–log linear regression suggest that microplastic is more efficiently transported than macroplastic and total plastics in river systems whose catchments have a high MMPW generation. This is in line with the increased correlation (Pearson correlation coefficient) of microplastic loads with the mean discharge (microplastic: 0.79; macroplastic: 0.4, total plastic: 0.49) (SI Figure S2). Not only loads but also concentrations are positively related to larger rivers. A further indication of an increased transport control of microplastic is that the r^2 is considerably lower for the relationship between specific MMPW and specific loads than for MMPW and loads. Specific MMPW can be interpreted as source strength within the river catchment. However, on the one hand larger rivers may facilitate the transport of microplastics, on the other hand there are additional sources which are not accounted for in the MMPW data, such as wastewater discharge, inland navigation and industrial activities located preferentially at larger rivers may directly contribute to microplastic inputs such as preproduction pellets.¹²

The delivery ratios are not uniform and are related to the river size. The delivery ratio is the ratio between the observed plastic loads and MMPW. Again the relationship between the delivery ratio is stronger for microplastics, supporting the idea of the higher transport efficiency for microplastic, particularly in larger catchments. Spatially uniform delivery ratios as reported by⁵ thus reflect the fraction of plastic MMPW potentially available for transport but not necessarily the actual fraction transported in rivers. On the other hand, if a high fraction of MMPW enters the river systems but only a relatively small amount is found being transported, suggest that plastic debris accumulates in river systems with potentially adverse effects on aquatic life, similarly to the oceans.³⁵ Although there is no data

to quantitatively confirm the immobilization, plastic debris has been documented at river banks³⁶³⁷ and in river bed sediments.³⁸ Also transport simulations suggest that plastic particles are efficiently retained in rivers.³⁹

The estimates of land-based plastic inputs into the sea in Jambeck et al. (2015)⁵ range between 4.8×10^6 and 12.7×10^6 t/y. Their estimates consider a 50 km coastal zone with an MMPW production of 32×10^6 t/y. The area of the global river catchments is of course larger and 76×10^6 t/y MMPW are produced in the catchments. One would expect higher loads originating from global rivers compared to the coastal zone alone. Our estimates of river inputs are remarkably lower for Model 1 with a median of 0.48×10^6 t/y and are within a similar range for Model 2 with a median of 2.75×10^6 t/y. Our results bracket those of a recent study by Lebreton et al. (2017)¹⁰ who found that rivers export between 1.15 and 2.41×10^6 t/y. These similar results are not surprising, as both studies use the same underlying MMPW data set and partially overlapping data of plastic in rivers.

Generally, our estimated values are uncertain because of the sparse, both spatially and temporarily and fairly heterogeneous source data. However, assuming Model 1 being realistic would suggest that pathways other than rivers such as direct stormwater runoff, wind dispersal and littering potentially account for a considerable fraction of total land-based inputs. If Model 2 would best represent reality, rivers would indeed be a major pathway for land-based plastic inputs to the sea.

The high discrepancy between the total plastic loads in Model 1 and 2 mainly arise from the different slopes of the underlying log–log -linear regression equations. Steeper slopes result in largely increased load estimates in large rivers and thus also in the global loads. However, with the current database it is hardly possible to better constrain the river inputs into the sea.

Independent of the estimate, be it millions or hundreds of thousand tons, it is not possible to close the mass balance between the plastic debris loads delivered to the sea and the amount found at the sea surface. The estimates of floating plastic debris in the oceans diverge greatly between 7000 and 35 000 tons,³² 268 940 tons,⁴⁰ and between 93 000–236 000 tons⁴¹ and broadly agree with the annual load supplied by the global river networks. However, the ocean data represents plastic debris discharged over an unknown time period, yet longer than annual time period and no exhaustive information on plastic fate in the ocean can be derived from these data. We have also tested if plastic loads in rivers reveal a temporal trend. Our data set comprises samples taken between 2004 and 2016. We considered loads of microplastics, macroplastics and total plastics. There was no observable trend in the data. However, we had to aggregate data across river catchments because no multiyear time-series are available yet from a single river. We compared loads normalized by population to account for the different catchments. The composition, size, and mass distribution of floating plastic debris in the sea appears similar to that of the mobile plastic debris in rivers.³⁴ This suggests that plastic debris may not be significantly fragmented within the time scales required for transport from rivers to the large ocean gyres or it simply indicates that certain size fractions are preferentially captured by the drift net sampling applied in both marine and freshwater systems and also that certain size fractions are preferentially transported in river systems which is then reflected in the composition of marine plastic debris.

The spatial pattern of river inputs are generally similar to the estimates of Jambeck et al.⁵ However, in our estimates the

fraction contributed from larger rivers is considerably higher. Note that despite differences in the MMPW generation per capita among the different countries as a result of their economic status, the catchment size has a main control on the total amount of MMPW generated (SI Figure S2).

Rivers from the 10 top-ranked catchments alone contribute between 88% and 94% (depending on the underlying model) of the total plastic load. A proportional relationship between MMPW and the load would result in 58%. The high fraction of a few river catchments contributing the vast majority of the total load implies that potential mitigation measures would be highly efficient when applied in the high-load rivers. Reducing plastic loads by 50% in the 10 top-ranked rivers would reduce the total river-based load to the sea by 45%.

Our analysis reveals that plastic loads of large rivers disproportionately increase in relationship to the increase of plastic debris available for transport.⁶

■ ASSOCIATED CONTENT

■ Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.7b02368.

Data on measured plastics in rivers, global catchment data, mismanaged plastic waste in the catchments, additional plots representing the methodological concept and the data flow, a map of catchments with measured data, a table summarizing the results for the top-ten ranked river catchments (PDF)

A spreadsheet containing all data used in study (ZIP)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: christian.schmidt@ufz.de.

ORCID

Christian Schmidt: 0000-0001-9787-8327

Author Contributions

C.S. and S.W. designed the research; T.K. performed the GIS analysis. C.S. performed the statistical analysis. All authors wrote the manuscript.

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

We are grateful to Marie J. Kurz and Andreas Musolf for their critical comments and Susan Walter for supporting the TOC figure design. We also thank the three reviewers for their valuable inputs which helped to improve the manuscript. This research was supported by the Helmholtz Research Programme "Terrestrial Environment", Topic 3: "Sustainable Water Resources Management".

■ REFERENCES

- (1) Rochman, C. M.; Browne, M. A.; Underwood, A. J.; van Franeker, J. A.; Thompson, R. C.; Amaral-Zettler, L. A. The ecological impacts of marine debris: unraveling the demonstrated evidence from what is perceived. *Ecology* **2016**, 97 (2), 302–312.
- (2) Thompson, R. C.; Olsen, Y.; Mitchell, R. P.; Davis, A.; Rowland, S. J.; John, A. W. G.; McGonigle, D.; Russell, A. E. Lost at sea: Where is all the plastic? *Science* **2004**, 304 (5672), 838–838.
- (3) Cole, M.; Lindeque, P.; Fileman, E.; Halsband, C.; Goodhead, R.; Moger, J.; Galloway, T. S. Microplastic Ingestion by Zooplankton. *Environ. Sci. Technol.* **2013**, 47 (12), 6646–6655.

- (4) Free, C. M.; Jensen, O. P.; Mason, S. A.; Eriksen, M.; Williamson, N. J.; Boldgiv, B. High-levels of microplastic pollution in a large, remote, mountain lake. *Mar. Pollut. Bull.* **2014**, 85 (1), 156–163.
- (5) Jambeck, J. R.; Geyer, R.; Wilcox, C.; Siegler, T. R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K. L. Plastic waste inputs from land into the ocean. *Science* **2015**, 347 (6223), 768–771.
- (6) van Sebille, E.; England, M. H.; Froyland, G. Origin, dynamics and evolution of ocean garbage patches from observed surface drifters. *Environ. Res. Lett.* **2012**, 7 (4), 044040.
- (7) Small, C.; Nicholls, R. J. A Global Analysis of Human Settlement in Coastal Zones. *J. Coastal Res.* **2003**, 19 (3), 584–599.
- (8) Ludwig, W.; Probst, J.-L.; Kempe, S. Predicting the oceanic input of organic carbon by continental erosion. *Global Biogeochem. Cycles* **1996**, 10 (1), 23–41.
- (9) Gruber, N.; Galloway, J. N. An Earth-system perspective of the global nitrogen cycle. *Nature* **2008**, 451 (7176), 293–296.
- (10) Lebreton, L. C. M.; Zwet, J.; van der Damsteeg, J.-W.; Slat, B.; Andrady, A.; Reisser, J. River plastic emissions to the world's oceans. *Nat. Commun.* **2017**, 8, ncomms15611.
- (11) Baldwin, A. K.; Corsi, S. R.; Mason, S. A. Plastic Debris in 29 Great Lakes Tributaries: Relations to Watershed Attributes and Hydrology. *Environ. Sci. Technol.* **2016**, 50 (19), 10377–10385.
- (12) Lechner, A.; Keckeis, H.; Lumesberger-Loisl, F.; Zens, B.; Krusch, R.; Tritthart, M.; Glas, M.; Schludermann, E. The Danube so colourful: A potpourri of plastic litter outnumbers fish larvae in Europe's second largest river. *Environ. Pollut.* **2014**, 188, 177–181.
- (13) Carson, H. S.; Lamson, M. R.; Nakashima, D.; Toloumu, D.; Hafner, J.; Maximenko, N.; McDermid, K. J. Tracking the sources and sinks of local marine debris in Hawaii. *Mar. Environ. Res.* **2013**, 84, 76–83.
- (14) Dris, R.; Gasperi, J.; Rocher, V.; Saad, M.; Renault, N.; Tassin, B. Microplastic contamination in an urban area: a case study in Greater Paris. *Environ. Chem.* **2015**, 12 (5), 592–599.
- (15) Faure, F.; Demars, C.; Wieser, O.; Kunz, M.; de Alencastro, L. F. Plastic pollution in Swiss surface waters: nature and concentrations, interaction with pollutants. *Environ. Chem.* **2015**, 12 (5), 582–591.
- (16) Mani, T.; Hauk, A.; Walter, U.; Burkhardt-Holm, P. Microplastics profile along the Rhine River. *Sci. Rep.* **2016**, 5, 17988.
- (17) Wang, W.; Ndungu, A. W.; Li, Z.; Wang, J. Microplastics pollution in inland freshwaters of China: A case study in urban surface waters of Wuhan, China. *Sci. Total Environ.* **2017**, 575, 1369–1374.
- (18) McCormick, A.; Hoellein, T. J.; Mason, S. A.; Schluep, J.; Kelly, J. J. Microplastic is an Abundant and Distinct Microbial Habitat in an Urban River. *Environ. Sci. Technol.* **2014**, 48 (20), 11863–11871.
- (19) McCormick, A. R.; Hoellein, T. J.; London, M. G.; Hittie, J.; Scott, J. W.; Kelly, J. J. Microplastic in surface waters of urban rivers: concentration, sources, and associated bacterial assemblages. *ECO-SPHERE* **2016**, 7 (11), e01556.
- (20) Rech, S.; Macaya-Caquilpan, V.; Pantoja, J. F.; Rivadeneira, M. M.; Campodonico, C. K.; Thiel, M. Sampling of riverine litter with citizen scientists - findings and recommendations. *Environ. Monit. Assess.* **2015**, 187 (6), 10.1007/s10661-015-4473-y
- (21) Moore, C. J.; Lattin, G. L.; Zellers, A. F. Quantity and type of plastic debris flowing from two urban rivers to coastal waters and beaches of Southern California. *Journal of Integrated Coastal Zone Management* **2011**, 11 (1), 65–73.
- (22) Hohenblum, P. *Plastik in der Donau (Plastics in the Danube)*, REPORT REP-0547; Umweltbundesamt: Wien, 2015.
- (23) van der Wal, M. Identification and Assessment of Riverine Input of (Marine) Litter; 2015.
- (24) Quilbe, R.; Rousseau, A. N.; Duchemin, M.; Poulin, A.; Gangbazo, G.; Villeneuve, J. P. Selecting a calculation method to estimate sediment and nutrient loads in streams: Application to the Beaurivage River (Quebec, Canada). *J. Hydrol.* **2006**, 326 (1–4), 295–310.
- (25) Lehner, B.; Verdin, K.; Jarvis, A. New Global Hydrography Derived From Spaceborne Elevation Data. *Eos Trans; AGU*, 2008, 89 (10), 93–94.

(26) Milliman, J. D.; Farnsworth, K. L. *River Discharge to the Coastal Ocean: A Global Synthesis*; Cambridge University Press: Cambridge, 2011.

(27) Center for International Earth Science Information Network - CIESIN - Columbia University; United Nations Food and Agriculture Programme - FAO; Centro Internacional de Agricultura Tropical - CIAT. Gridded Population of the World, Version 3 (GPWv3): Population Count Grid. NASA Socioeconomic Data and Applications Center (SEDAC) 2005.

(28) Hoornweg, D.; Bhada-Tata, P. *What a Waste: A Global Review of Solid Waste Management*. 2012.

(29) Serajuddin, U.; Fantom, N. J. *The World Bank's Classification of Countries by Income*; WPS7528; The World Bank, 2016; pp 1–52.

(30) Meybeck, M.; Laroche, L.; Dürr, H. H.; Syvitski, J. P. M. Global variability of daily total suspended solids and their fluxes in rivers. *Global and Planetary Change* **2003**, 39 (1–2), 65–93.

(31) Turowski, J. M.; Badoux, A.; Bunte, K.; Rickli, C.; Federspiel, N.; Jochner, M. The mass distribution of coarse particulate organic matter exported from an Alpine headwater stream. *Earth Surf. Dyn.* **2013**, 1 (1), 1–11.

(32) Cózar, A.; Echevarría, F.; González-Gordillo, J. I.; Irigoien, X.; Úbeda, B.; Hernández-León, S.; Palma, Á. T.; Navarro, S.; García-de-Lomas, J.; Ruiz, A.; et al. Plastic debris in the open ocean. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, 111 (28), 10239–10244.

(33) Law, K. L.; Moret-Ferguson, S.; Maximenko, N. A.; Proskurowski, G.; Peacock, E. E.; Hafner, J.; Reddy, C. M. Plastic Accumulation in the North Atlantic Subtropical Gyre. *Science* **2010**, 329 (5996), 1185–1188.

(34) ter Halle, A.; Ladirat, L.; Gendre, X.; Goudouneche, D.; Pusineri, C.; Routaboul, C.; Tenailleau, C.; Duployer, B.; Perez, E. Understanding the Fragmentation Pattern of Marine Plastic Debris. *Environ. Sci. Technol.* **2016**, 50 (11), 5668–5675.

(35) Eerkes-Medrano, D.; Thompson, R. C.; Aldridge, D. C. Microplastics in freshwater systems: A review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. *Water Res.* **2015**, 75, 63–82.

(36) Hoellein, T.; Rojas, M.; Pink, A.; Gasior, J.; Kelly, J. Anthropogenic Litter in Urban Freshwater Ecosystems: Distribution and Microbial Interactions. *PLoS One* **2014**, 9 (6), e98485.

(37) Klein, S.; Worch, E.; Knepper, T. P. Occurrence and Spatial Distribution of Microplastics in River Shore Sediments of the Rhine-Main Area in Germany. *Environ. Sci. Technol.* **2015**, 49 (10), 6070–6076.

(38) Wagner, M.; Scherer, C.; Alvarez-Muñoz, D.; Brennholt, N.; Bourrain, X.; Buchinger, S.; Fries, E.; Grosbois, C.; Klasmeier, J.; Marti, T.; et al. Microplastics in freshwater ecosystems: what we know and what we need to know. *Environ. Sci. Eur.* **2014**, 26 (1), 12.

(39) Besseling, E.; Quik, J. T. K.; Sun, M.; Koelmans, A. A. Fate of nano- and microplastic in freshwater systems: A modeling study. *Environ. Pollut.* **2017** 22054010.1016/j.envpol.2016.10.001

(40) Eriksen, M.; Lebreton, L. C. M.; Carson, H. S.; Thiel, M.; Moore, C. J.; Borerro, J. C.; Galgani, F.; Ryan, P. G.; Reisser, J. Plastic Pollution in the World's Oceans: More than 5 Trillion Plastic Pieces Weighing over 250,000 Tons Afloat at Sea. *PLoS One* **2014**, 9 (12), e111913.

(41) van Sebille, E.; Wilcox, C.; Lebreton, L.; Maximenko, N.; Hardesty, B. D.; van Franeker, J. A.; Eriksen, M.; Siegel, D.; Galgani, F.; Law, K. L. A global inventory of small floating plastic debris. *Environ. Res. Lett.* **2015**, 10 (12), 124006.