Environmental Science & Technology

Article

Cite This: Environ. Sci. Technol. 2019, 53, 11810−11817

pubs.acs.org/est

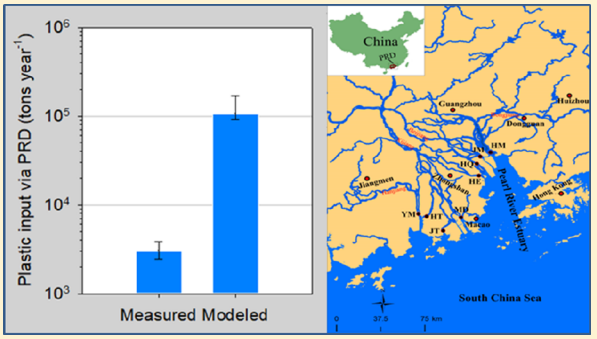
Riverine Microplastic Pollution in the Pearl River Delta, China: Are Modeled Estimates Accurate?

Lei Mai, Shan-Ni You, Hui He, Lian-Jun Bao, Liang-Ying Liu, and Eddy Y. Zeng*

Guangdong Key Laboratory of Environmental Pollution and Health, School of Environment, Jinan University, Guangzhou 510632, China

Supporting Information

ABSTRACT: Plastic pollution has caused increasing global concern. Currently, model estimates of the riverine plastic inputs to the global oceans based on the concept of Mismanaged Plastic Waste (MPW) varied substantially, and no field measurements of riverine inputs were available. We conducted sampling at the eight major river outlets of the Pearl River Delta, South China with rapid economic growth and urbanization to provide field measured data for fine-tuning modeling results. Floating microplastics (MPs) were collected with a Manta net (mesh size of 0.33 mm) five times during 2018. Microplastic particles (0.3−5.0 mm) widely occurred in all sampling sites. The number and mass concentrations of MPs were in the ranges of 0.005−0.7 particles m^{-3} and 0.004−1.28 mg m^{-3} and were positively correlated with water discharges. The annual riverine input of MPs from the Pearl River Delta was estimated at 39 billion particles or 66 tons, which converts to 2400−3800 tons of plastic debris based on calculations described in Text S2. These values were substantially below the MPW-based model estimates (91,000−170,000 tons). The large difference between measured and modeling results may have derived from the large uncertainty in the MPW values assigned to the world's countries/regions.



INTRODUCTION

Past decades have witnessed the growing convenience of plastic products in human daily life. The global production of plastic materials reached 348 million tons in 2017.[1] Once discarded, approximately 4.8 to 12.7 million tons of land-based plastic waste may enter the oceans as estimated in 2010.[2] Due to the buoyancy of most plastics, they are distributed abundantly in water surface. River networks, which often flow through large urban areas with numerous anthropogenic activities, receive plastic wastes from land-based sources. The Global River Discharge Database[3] estimates that the annual global riverine discharge from the continents to the oceans is 2.14×10^{13} m^3. In this context, riverine discharge can be an important vector for transporting plastics from terrestrial environments to the coastal seas.[4] Using an empirical model, Lebreton et al.[5] obtained an annual input of up to 2.4 million tons of plastic waste from rivers to the world's oceans. Coupled with plastic waste generated during maritime activities, large amounts of plastic debris can be accumulated in the ocean gyres through oceanic circulation, resulting in serious plastic pollution in the open oceans.[6,7]

The relative abundance of floating plastics increases with decreasing particle size,[8] and plastic particles of smaller sizes can be ingested by a wider range of organisms. Thus, microplastics (MPs), with particle size of ≤5 mm, are of utmost concern in the environment.[9] Ingestion of MPs by organisms has been widely reported, from small planktonic organisms[10] to large mammals,[11] which may pose potential damage to organisms or change the microbial environment inside their bodies.[12] Due to their strong sorption capability, MPs may sorb hydrophobic chemicals (e.g., polycyclic aromatic hydrocarbons and polychlorinated biphenyls) and heavy metals (e.g., zinc) from surrounding environments,[13−15] thus acting as a facilitator for transport of toxic chemicals in the environment[16,17] and also chemical transformation in organisms.[18] Evidence for toxic effects of MP ingestion by aquatic organisms has also been documented.[19,20]

Although China is the largest producer of plastic materials in the world,[21] the export of plastic waste, especially MPs, from rivers to the coastal seas remains largely unknown. Modeling results based on the concept of mismanaged plastic waste (MPW) combined with population density and other socio-economic data suggested that China accounted for a large portion of the global riverine input to the ocean.[5] Despite the initial (and largely preliminary) success of modeling the riverine inputs of plastics to the global oceans,[2] field measurements over an extended time period are needed to provide the ultimate validation.

Received: August 10, 2019
Revised: September 17, 2019
Accepted: September 18, 2019
Published: September 18, 2019

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

The river network within the Pearl River Delta (PRD) drains into the South China Sea (SCS) through eight major riverine outlets (Supporting Information, Figure S1), and Hong Kong at the downstream of the major portion of the PRD was reported to be a hotspot of MPs.[22] Therefore, the PRD is an ideal site for field measurement of riverine MP inputs to coastal oceans. In the present study, we sampled five times at the eight outlets of the PRD in 2018 and obtained number and mass concentrations of MPs and calculated their correlations with river discharge. The objectives were to (1) document the occurrence of MPs in the riverine system of the PRD and riverine MP inputs to the SCS; (2) examine the correlation between riverine MP inputs and water discharges, and (3) provide field-measured data for calibrating model estimates for global riverine inputs of plastics (by converting the inputs of MPs to those of plastics).

METHODS AND MATERIALS

Sample Collection. Field sampling was conducted at the outlets of Humen (HM), Jiaomen (JM), Hongqimen (HQ), Hengmen (HE), Modaomen (MD), Jitimen (JT), Hutiaomen (HT), and Yamen (YM) (Figure S1). Samples were collected by performing triplicate surface water trawling at each site (Figure S2) during five discrete sampling events on January 16, April 19, June 15, August 11, and November 9, 2018 within 1 h before the intraday lower tide (a neap tide to obtain the net flux from river to the sea). A Manta trawl (1.0 m × 0.5 m rectangle mouth; 0.33 mm mesh) was used to collect floating plastics at a speed of approximately 2 knots for a period of 15−20 min, with a removable cod-end to retain samples. The detailed sampling conditions are provided in Table S1. The trawling distance was obtained from the digital reading of a flowmeter (Hydro-Bios, Germany) attached in the middle of the rectangle mouth. After each towing, the trawl was rinsed thoroughly with local water, and all retained materials were combined and transferred to a 500 mL brown wide-mouth glass bottle. Samples were immediately transported to the laboratory within 4 h and stored in a cold room at 2−4 °C prior to analysis. To maximize sample homogeneity, samples from three replicate tows were combined to make one sample. A flowchart depicting the analytical process of MPs is presented in Figure 1.

Sample Extraction and Identification. Materials decanted from each glass bottle were combined with saturated sodium chloride solution (~1.2 g cm^{-3})[23,24] prepared in the laboratory with distilled water and analytical pure sodium chloride (precombusted in a muffle furnace at 450 °C). Subsequently, floating plastic particles were filtered through a 0.3 mm stainless steel sieve and picked out with a precleaned tweezers. The attached organic matter was rinsed thoroughly with distilled water using the same procedures as described in literature studies.[6,25] This procedure was repeated three times, and the leftover in the glass bottle was removed and examined thoroughly for any potential MPs. A 40× optical microscope was used to inspect fine particles, and the criteria reported by Norén[26] were applied to visually identify MPs. Identified MP particles were rinsed with distilled water, wrapped with aluminum foil, and freeze-dried. All dried samples were further classified into three physical shapes, that is, fragments, lines (include fibers), and pellets. Fragments were further divided into small (0.3−2 mm) and large (2−5 mm) sizes. Plastic particles with size larger than 5 mm were excluded in the present study.

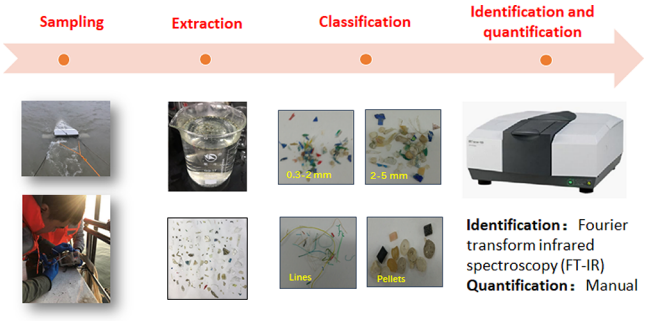


Figure 1. Flowchart depicting the full process for microplastic analysis used in the present study, including sampling, extraction, classification and identification, and quantification.

An attenuated total reflectance Fourier transform infrared spectrometer (Shimadzu model 8300) was employed to determine the polymer types for all MPs by comparing the sample and standard reference spectra (Figure S3), with similarity values mostly higher than 80% (Table S2). The accuracy of visual identification for plastic debris was demonstrated to be approximately 98% (Table S3). Identified MP particles in each classification were further counted manually. For mass measurement, identified MPs were carefully transferred to a piece of preweighed clean filter paper placed close to the operating platform of the FTIR. The total mass was weighed with an analytical balance to the nearest 0.1 mg and recorded, and the total mass minus the clean filter paper mass was taken as the net weight of MPs.

Quality Assurance and Quality Control. To minimize potential cross-contamination, latex gloves and cotton lab coats were worn during the entire sampling process. Each field blank was prepared by placing 4 L of distilled water in a glass bottle and taken to the sampling site (a total of 40 field blanks were used). The glass bottle lid remained open during sampling. These field blanks were transported back to the laboratory and analyzed along with the field samples. In the laboratory, one additional laboratory blank was prepared for each sampling event (n = 5 in total) by filling a precleaned beaker with distilled water and exposing the beaker to the air during sample analysis. The sample treatment was performed in a clean fume hood. No MPs were found in the field blanks, but few textile fibers were occasionally identified in the laboratory blanks (one fiber was found on 15 June and two fibers on 11 August). They were excluded from data analysis because they may have derived from airborne deposits in the laboratory.[27]

Data Analysis. The abundance of MPs from each sampling event was calculated by dividing the number (particles) or mass (milligrams) of identified plastic particles by the filtered water volume obtained by multiplying the rectangle mouth area with the towing distance and presented as particles m^{-3} or mg m^{-3}. A riverine input (F_{ij}) of MPs from the ith outlet during the jth season was calculated by

$$F_{i,j} = C_{i,j} \times Q_{i,j} \times 10^9$$

where $C_{i,j}$ is the concentration of MPs obtained from the ith outlet during the jth season, where January, April, June and August, and November represent winter, spring, summer, and autumn, respectively, and $Q_{i,j}$ (m^3) is the total river discharge with December to February, March to May, June to August,

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

and September to October representing winter, spring, summer, and autumn seasons, respectively. A previously reported method[28] was used to estimate Q_{ij} with hydrologic data obtained from the government hydrologic website http://www.gdsw.gov.cn/tjxx_sqjb.html. Detailed protocols are presented in Text S1 and Table S4. The total annual input (F_i) from the ith outlet is the sum of the data for four seasons. Pairwise comparison (t test) and Pearson correlation data analysis were performed with SPSS (IBM SPSS Statistics 22).

RESULTS AND DISCUSSION

Distribution of Microplastics in Surface Water. Microplastics widely occurred at all sampling sites, with concentrations ranging from 0.005 to 0.704 particles m^{-3} (average 0.127 particles m^{-3}) or from 0.004 to 1.28 mg m^{-3} (average 0.21 mg m^{-3}) (Table S5). The median concentration of MPs at HM outlet (0.275 particles m^{-3} or 0.65 mg m^{-3}) was comparable to that obtained in the surface waters of Hong Kong (median concentration of 0.424 particles m^{-3} or 0.114 mg m^{-3}), which was reported to be under the influence of HM outlet at the PRD.[29] The increased number concentration and decreased mass concentration of microplastics in Hong Kong waters indicated possible fragmentation of plastic particles during transport from river to the coastal sea. Lin et al.[30] reported extremely high MP concentrations (380−7900 particles m^{-3}) in bulk water samples (60 L each) from the upstream of HM outlet. However, Lusher et al.[31] demonstrated that the volume of filtered water can significantly affect the number of particles collected, which decreased with increasing filtered water volume. The filtered water volumes in the present study (Table S1) were four to five orders of magnitude larger than those collected by Lin et al.[30] Because sampling methodologies are critical for collecting MPs,[32] it would not be reasonable to directly compare the results from the two studies employing substantially different methods. Using a trawling net of the same mesh size (0.33 mm) as used in the present study, Baldwin et al.[24] found a median plastic concentration of 1.9 particles m^{-3} in the surface water of 29 Great Lakes tributaries (98% of them were MPs), relatively higher than that obtained in the present study.

Spatially, the average MP concentrations varied greatly among sampling sites (Figure 2). Higher concentrations occurred at the four eastern outlets (HM, JM, HQ, and HE) than at all western outlets (JT, HT, and YM) except for MD, which has the largest water discharge among all outlets. A plausible explanation for this spatial pattern was that HM and HE outlets receive water draining from several highly urbanized districts, such as Guangzhou, Dongguan, Huizhou, and Zhongshan. Similarly, a significant correlation was found between plastic concentrations and urbanization levels of the watersheds in the 29 Great Lakes tributaries.[24] These districts are home to a large population, estimated at 50 million by the Guangdong Statistical Yearbook 2018,[33] resulting in large consumption of plastic products. In contrast, the areas around JT and HT are dominated by agricultural and forestry lands. Paired t test suggested no significant statistical variation between the concentrations of MPs from most sampling sites ($p > 0.05$), except for four pairs, that is, HM and HT, HE and HQ, HE and JT, and HE and HT (Table S6). Yonkos et al.[34] found that the concentrations of MPs were positively correlated with population densities but negatively correlated with the agricultural and forestry land use proportions in a half-year study measuring MPs at different estuaries in Chesapeake Bay, USA.

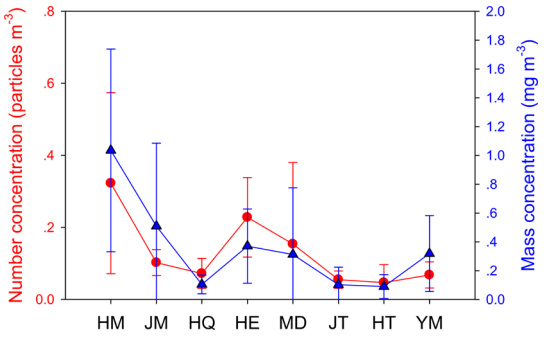


Figure 2. Number and mass concentrations of microplastics in surface water of the eight major river outlets of the Pearl River Delta, China: Humen (HM), Jiaomen (JM), Hongqimen (HQ), Hengmen (HE), Modaomen (MD), Jitimen (JT), Hutiaomen (HT), and Yamen (YM). All data points are averages of five measurements in January, April, June, August, and November 2018, and the error bars are the standard deviations.

The present study showed a clear temporal pattern in the concentrations of MPs by both number and mass (Figure S4), with relatively higher concentrations in June and August. Paired t test suggested significant greater concentrations in August than in January ($p < 0.05$). The database on Guangdong Meteorological Service of 2018 indicates that the precipitations are significantly greater in June (390 mm) and August (389 mm) than in January (112 mm), April (80 mm), and November (60 mm) (t test; $p < 0.05$). Distribution patterns of MPs in surface water may vary with natural conditions such as the frequency and strength of precipitations. For example, Faure et al.[35] reported one order of magnitude higher MP concentrations in the wet weather season (64 ± 35 particles m^{-3} or 12 ± 0.92 mg m^{-3}) than in the dry weather season (6.5 ± 5.3 particles m^{-3} or 1.6 ± 2.6 mg m^{-3}) in Venoge River (Swiss). Higher abundance of MPs was also found during rainy season in June to August (0.14 particles m^{-3}) than during dry season in September to November (0.0039 particles m^{-3}) in the surface water of Goiana Estuary in Brazil.[36] Extreme metrological events may also bring out abundant MPs, for example, a study in India reported 3-fold higher concentrations of MPs during postflood than during preflood.[37] Heavy rainfalls can facilitate the transport of MPs from lands to coastal seas via estuaries.[38]

The hydrodynamic conditions in the present study region varied spatially and temporally, as reflected in the large range of monthly water discharges (0.27−8.86 × 10^9 m^3; Table S7) and may have caused the variability in the concentrations of MPs. The water discharges from the three heavily polluted outlets (MD, HM, and HE) account for approximately 25% (MD), 20% (HM) and 14% (HE) of the total discharge.[28] Lima et al.[39] indicated that MPs waft with mainstream water flows and are more closely associated with water discharge than with environmental variables (e.g., salinity and precipitation). In the present study, a positive logarithmic correlation between the number ($r^2 = 0.19$; $n = 40$) or mass ($r^2 = 0.3$; $n = 40$) concentrations of MPs and the river discharges was identified (Figure 3). Furthermore, this positive Pearson correlation was statistically significant for number concentrations ($r = 0.329$, $p = 0.038$) but not for mass concentrations ($r = 0.271$, $p = 0.09$). No dilution effect was found accompanying with increased river discharge. These results

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

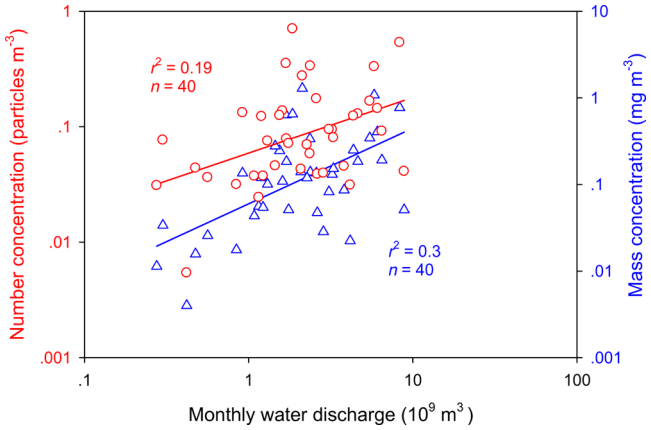


Figure 3. Positive correlations between the number (red open circle) and mass (blue open triangles) concentrations of microplastics and water discharges at the eight major river outlets of the Pearl River Delta, China, sampled in January, April, June, August, and November 2018.

demonstrated that intensified rainfalls may have mobilized increased amounts of MPs deposited within various environmental compartments of the PRD region into the aquatic system. Hurley et al.[40] also demonstrated that an increased export of MPs from rivers can be caused by flooding.

Characteristics of Microplastic Particles. Polyethylene (PE) and polypropylene (PP) dominated the plastic types for MPs, followed by polyamide, polystyrene, poly(ethylene) terephthalate, polyformaldehyde, acrylonitrile butadiene styrene, and poly(vinyl chloride) (Figure 4a). This is consistent with the result obtained in surface water of the Three Gorges Reservoir in China, that is, low-density MPs accounted for the largest fraction of the total plastic pieces.[41] Not surprisingly, low-density MPs were always the most abundant type in surface water samples because of their light weights. Among the 10 polymer types identified in sediments of Venice Lagoon, PE and PP also accounted for more than 80% of the total MPs.[42] Apparently the predominance of PE and PP in environmental samples is attributed to the widespread use of their products with short life cycles. This in turn leads to high demand for PE and PP raw materials; for example, the demand for PE and PP in Europe in 2015 was 16.5 million tons, accounting for 34% of the total plastic materials.[21]

The MP particles obtained in the present study were classified into fragments, lines, and pellets based on their morphological characteristics. The fragments were further divided into small (0.3−2 mm) and large (2−5 mm) sizes. This classification protocol is similar to the one adopted by Yonkos et al.[34] The number concentrations of MPs followed the order of 0.3−2 mm fragments > 2−5 mm fragments > lines > pellets (Figure S5). The relative abundance of each MP classification was consistent among all eight sampling sites (Figure 4b), and fragments were apparently much more abundant than lines and pellets. This was similar to the classification of MPs found in Xiangxi River, where fragment and sheet are the dominant shapes, accounting for more than 70% of the total MPs.[41] Lines generally originate from fishing lines, and pellets are commonly derived from industrial plastic resins. Different from lines and pellets, fragments are generated from breakup of large plastic pieces, such as plastic carrying bags, packaging bags, and plastic bottles. In the present study,

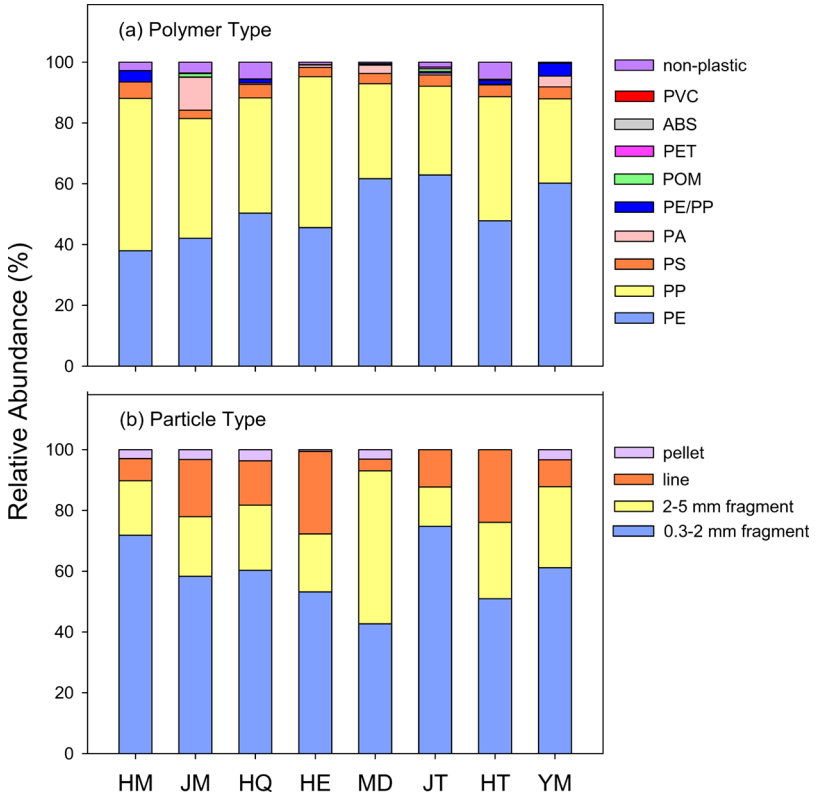


Figure 4. Relative abundances of (a) polymer type and (b) particle type of microplastics collected from the eight major river outlets of the Pearl River Delta, China. PE: polyethylene; PP: polypropylene; PS: polystyrene; PA: polyamide; PE/PP: polyethylene/polypropylene; POM: polyformaldehyde; ABS: acrylonitrile butadiene styrene; and PVC: poly(vinyl chloride).

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

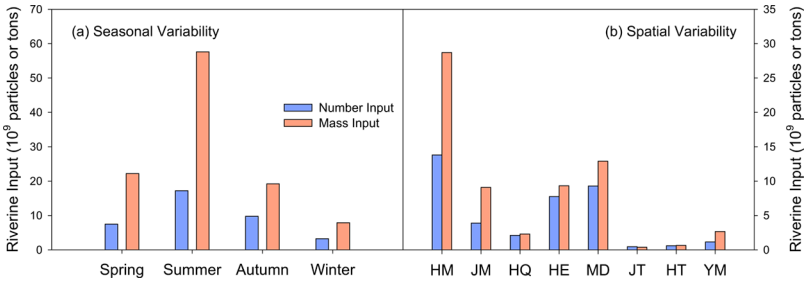


Figure 5. (a) Seasonal and (b) spatial variations in the number and mass inputs of microplastics from the eight major river outlets of the Pearl River Delta to the South China Sea.

the abundance of fragments increased with decreasing particle size, and the 0.3−2 mm fragments accounted for more than half of the total MPs (Figure 4b). Pazos et al.[43] also found a decrease trend of MP abundances going from 500 μm to 5 mm. Obviously, progressive fragmentation of large plastic pieces into more and smaller ones can result in a gradual increase of smaller-sized fragments.[6]

Occurrence of Microplastics in the World's Rivers. The occurrence of freshwater MPs has been widely documented (Table S8), but it remains a challenge to compare MP pollution in the world's rivers due to the lack of standardized sampling and analytical protocols. An average concentration of 1.07 ± 0.64 particles m^{-3} sampled with a 0.33 mm mesh net was reported in Lake Winnipeg of North America.[44] Also in North America, higher concentrations (2.57 ± 2.95 particles m^{-3}) were documented in Snake and Lower Columbia Rivers with a 0.1 mm mesh net than in Lake Winnipeg.[45] Clearly, mesh size is an important parameter for sampling water surface MPs by trawling. In the present study, the units of MP abundances used in 11 previous studies using similar sampling methods (surface trawling with a net mesh size of approximately 0.33 mm) were converted to "particles m^{-3}", based on the height of the trawl's opening mouth. The concentrations of MPs varied greatly among 22 rivers around the globe (Figure S6). The average MP concentrations at the eight river outlets were lower than most of the previously reported values (Figure S6).

No obvious regional difference in MP concentrations was found, with the highest and lowest concentrations documented in the Chicago River (USA) and Thames River (UK), respectively (Figure S6). For example, an average concentration of 0.028 particles m^{-3} was reported in Thames River,[46] which was lower than those at the eight river outlets of the PRD. On the contrary, a higher abundance was reported in Chicago River (18 particles m^{-3}).[47] Concentrations of MPs in the rivers at Chesapeake Bay ranged from 0.27−1.04 particles m^{-3},[34] and were mostly higher than those found in the present study, with the exception of HM outlet (Figure S6 and Table S8). Only a few studies were conducted in South Africa and South America, which acquired an average concentration of 3.11 particles m^{-3} in the Durban Estuary[48] (Table S8).

Clearly, some standardized sampling and analytical methods are needed to better compare the occurrences of MPs in surface waters around the world, especially the need for standardizing the trawling net mesh size and minimum filtered water volume. Based on the experience accumulated in the present study, a net mesh size of 0.33 mm and at least 100 m^3 filtered water are recommended for collecting MPs from rivers with large surface areas. Although Kapp and Yeatman[45] reported that the majority of MPs found was less than 0.33 mm in size, no standard procedure for sampling fine plastic particles in water has presented challenges for comparison of different studies. The 0.33 mm mesh neuston net is the most commonly used device for field sampling of MPs from surface waters,[24,34,49,50] and this method has been recognized as a "lower bound" by the National Oceanic and Atmospheric Administration for reporting microplastic pollution in water.[51] Later, a standard analytical method for samples collected using 0.33 neuston nets was established also by the National Oceanic and Atmospheric Administration.[23]

Riverine Inputs of Microplastics. The riverine inputs of MPs were calculated based on the concentration data (Table S5) and riverine water discharge (Table S7). Concentrations of MPs were expected to decrease exponentially with increasing vertical depth in sea waters.[52] Eo et al.[53] reported higher microplastic concentrations in surface water than in midwater at 1 m above the benthic sediment. Because the concentrations of MPs in surface water were used in the present study, the riverine MP inputs estimated thus may be regarded as the upper limits. In an attempt to estimate the annual inputs of MPs from the PRD region to the SCS via the eight major outlets, the seasonal inputs from each river outlet were in the range of 0.009−4.36 × 10^9 particles or 0.007−18.3 tons per season (Table S9). The sum of MP inputs in four seasons from one river outlet was taken as the annual MP input for that outlet. Individual annual MP inputs from the eight river outlets were summed to yield the annual riverine input of MPs from the PRD to the coastal ocean, which was 39 billion particles or 66 tons (Table S9).

The seasonal riverine inputs of MPs varied considerably, with the summer and winter seasons having the highest and lowest values, respectively (Figure 5a). This variation was reasonably attributed to the large difference between the water discharges in the summer and winter seasons (37 and 14.2% of the total, respectively). The annual inputs from each river outlet also varied spatially, with HM showing the highest number and mass weight (Figure 5b), because the HM outlet not only has large discharge but also drains through Guangzhou, a highly urbanized city, before flowing into the SCS. In general, the eastern outlets (HM, JM, HQ, and HE) transport more MPs than all western outlets (JT, HT, and YM) but MD (Figure 5b). This was probably the combined effect of MP concentrations and river discharges. The MD outlet carries the largest monthly discharges (Table S7), which have contributed to its large riverine MP input. In contrary, JT, HT, and YM outlets receive water draining through small towns with relatively low population densities and carry smaller water discharges than the eastern outlets (Table S7), resulting in lower MP inputs. This finding was consistent with

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

the riverine inputs of several groups of persistent organic pollutants reported in our previous study.[28]

Modeled and Measured Riverine Inputs of Plastics. Several models have recently been established to estimate land-derived inputs of plastic debris from rivers to the oceans.[4,5,54] In particular, six rivers in China were predicted to be among the top 20 rivers contributing to the global plastic input.[5] Three rivers (Xi, Dong, and Zhujiang rivers) eventually drain to the SCS via the eight major river outlets investigated herein. To compare with these modeling estimates, the riverine inputs of MPs in the present study were converted to those of plastic debris. A conversion procedure (Text S2) was used to estimate the riverine inputs of plastic debris from the PRD to the SCS, with the lower, midpoint, and upper mass inputs at 2400, 2900, and 3800 tons year^{-1}, respectively (Table S10).

The riverine plastic inputs based on field measurements were approximately two orders of magnitude lower than model estimates (Table S10). Several explanations for this large difference are provided herein. First, the model estimates were derived from the concept of MPW, initially introduced by Jambeck et al.[2] who determined the MPW collection rate for each country using the World Bank's recorded data on economic classification, geographic region, and waste disposal method. However, only 81 countries have direct records.[55] The authors had to divide all other countries without recorded data into several categories based on per capita income and geographic region. The MPW collection rate for countries in each category was an average value calculated using a regression model derived from the countries with recorded data.[2] However, countries in the same category may vary greatly in waste disposal practices. For example, the percent of dumping waste (considered as mismanaged) ranged from 0 for Chile to 100% for Suriname in 2010,[55] although these two countries belong in the same economic classification of Upper Middle Income and geographic region of Latin America and the Caribbean.

Second, the percent of MPW for China was pegged at 76% in 2010.[2] This may have been an overestimated number, as China's Statistical Yearbook recorded a domestic waste treatment rate of 78% in 2010,[56] equal to 22% for the percent of MPW. Finally, the time of field sampling in the present study (2018) was eight years apart from that of the data used in model estimation (2010); the treatment rate of domestic waste in Guangdong Province embracing the PRD increased from 72.1% in 2010 to 98% in 2017 (Figure S7).

Herein, we further demonstrate the subjectivity of MPW. Jambeck et al.[2] set the percent of MPW at 76% for China based on a regression model. Consequently, the annual riverine plastic inputs from the PRD were estimated at 91,000−170,000 tons,[5] which is substantially greater than our field measurements (2,400−3,800 tons). A reverse deduction using our field measured data yielded the fraction of MPW as 5.9−7.2%, which was quite close to the sum (4%) of littering fraction (2%) and untreated fraction (2%) of municipal solid waste reported in the Statistics Year Book of China.[57] Apparently, artificially inflated MPW values assigned to underdeveloped nations[2] would put these nations into an unfavorable position in ratifying and implementing any international treaty on plastics.

Field measurements are necessary for providing important constraints for fine-tuning model estimates and perhaps formulating more objective models in the future. Although modeling can be a valuable tool for predicting riverine MP inputs, extensive field studies are necessary to calibrate modeling results. A combined modeling and field measurement approach should increase the accuracy of predicting the patterns of MP pollution, particularly riverine MP inputs, which is critical for understanding the cycling of MPs in the environment. To establish and optimize such an approach, long-term monitoring programs are highly desirable for global rivers. In addition, the percent of inadequately managed plastic waste cannot be objectively obtained using a regression model based on geographic locations, level of income, and waste disposal method. More parameters, for example, population density, socioeconomic and technological development, human living standard, and level of education, obviously need to be taken into account.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.9b04838.

Procedures for estimating water discharge at the eight major river outlets (Text S1); conversion from MPs to plastic debris (Text S2); field sampling conditions (Table S1); comparison of FTIR spectra of MPs in field samples and reference spectra (Table S2); composition of collected particles (Table S3); percent discharge relative to the total discharge (Table S4); mean, median, and range of number and mass MP concentrations of MPs (Table S5); paired variation *t* test between sampling sites for particle concentrations of MPs (Table S6); population density within the watersheds and monthly water discharges during each sampling event (Table S7); global comparison of river MP concentrations (Table S8); seasonal inputs from each outlet (Table S9); comparison of plastic riverine inputs from the PRD between model estimates and field measurements (Table S10); map of the sampling sites (Figure S1); trawling design for sampling at HM and MD (Figure S2); FTIR spectra of MP samples and reference spectra (Figure S3); temporal patterns in the concentrations of MPs (Figure S4); abundances of MPs of various sizes and polymer types (Figure S5); concentrations of microplastics in several riverine surface waters around the world (Figure S6); and domestic waste treatment rates in Guangdong Province (Figure S7) (PDF)

■ AUTHOR INFORMATION

Corresponding Author

*E-mail: eddyzeng@jnu.edu.cn.

ORCID

Lei Mai: 0000-0001-6858-2543

Lian-Jun Bao: 0000-0002-0634-0829

Eddy Y. Zeng: 0000-0002-0859-7572

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

The present study was financially supported by the Ministry of Science and Technology of China (no. 2016YFC1402200), the National Natural Science Foundation of China (no. 21806053), and the Natural Science Foundation of Guang-

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

dong Province (no. 2018030310550). We are grateful to the sampling team consisted of mostly graduate students from Jinan University for assistance in field sampling. We would also like to thank Derek Muir for valuable comments and suggestions.

REFERENCES

(1) PlasticsEurope.. Analysis of European plastics production, demand and waste data. In *Plastics - the Facts 2018*; PlasticsEurope, 2018.
(2) 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*, 768−771.
(3) Milliman, J. D.; Farnsworth, K. L. *River discharge to the coastal ocean: A global synthesis*; Cambridge University Press, 2013.
(4) Schmidt, C.; Krauth, T.; Wagner, S. Export of plastic debris by rivers into the sea. *Environ. Sci. Technol.* **2017**, *51*, 12246−12253.
(5) Lebreton, L. C. M.; van der Zwet, J.; Damsteeg, J.-W.; Slat, B.; Andrady, A.; Reisser, J. River plastic emissions to the world's oceans. *Nat. Commun.* **2017**, *8*, 15611.
(6) Cózar, A.; Echevarría, F.; González-Gordillo, J. I.; Irigoien, X.; Úbeda, B.; Hernández-León, S.; Palma, A. T.; Navarro, S.; García-De-Lomas, J.; Ruiz, A.; Fernandez-de-Puelles, M. L.; Duarte, C. M. Plastic debris in the open ocean. *Proc. Natl. Acad. Sci. U. S. A.* **2014**, *111*, 10239−10244.
(7) Seltenrich, N. New link in the food chain? Marine plastic pollution and seafood safety. *Environ. Health Perspect.* **2015**, *123*, A34−A41.
(8) 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*, e111913.
(9) *Microplastic contamination in aquatic environment: An emerging matter of environmental urgency*; Zeng, E. Y. Ed.; Elsevier: Singapore, 2018; pp 409.
(10) Frias, J. P. G. L.; Otero, V.; Sobral, P. Evidence of microplastics in samples of zooplankton from Portuguese coastal waters. *Mar. Environ. Res.* **2014**, *95*, 89−95.
(11) Fossi, M. C.; Panti, C.; Guerranti, C.; Coppola, D.; Giannetti, M.; Marsili, L.; Minutoli, R. Are baleen whales exposed to the threat of microplastics? A case study of the Mediterranean fin whale (Balaenoptera physalus). *Mar. Pollut. Bull.* **2012**, *64*, 2374−2379.
(12) Rist, S.; Baun, A.; Hartmann, N. B. Ingestion of micro- and nanoplastics in *Daphnia magna* - Quantification of body burdens and assessment of feeding rates and reproduction. *Environ. Pollut.* **2017**, *228*, 398−407.
(13) Mai, L.; Bao, L.-J.; Shi, L.; Liu, L.-Y.; Zeng, E. Y. Polycyclic aromatic hydrocarbons affiliated with microplastics in surface waters of Bohai and Huanghai Seas, China. *Environ. Pollut.* **2018**, *241*, 834−840.
(14) Brennecke, D.; Duarte, B.; Paiva, F.; Caçador, I.; Canning-Clode, J. Microplastics as vector for heavy metal contamination from the marine environment. *Estuarine, Coastal Shelf Sci.* **2016**, *178*, 189−195.
(15) León, V. M.; García, I.; González, E.; Samper, R.; Fernández-González, V.; Muniategui-Lorenzo, S. Potential transfer of organic pollutants from littoral plastics debris to the marine environment. *Environ. Pollut.* **2018**, *236*, 442−453.
(16) Koelmans, A. A.; Bakir, A.; Burton, G. A.; Janssen, C. R. Microplastic as a vector for chemicals in the aquatic environment. Critical review and model-supported re-interpretation of empirical studies. *Environ. Sci. Technol.* **2016**, *50*, 3315−3326.
(17) Chua, E. M.; Shimeta, J.; Nugegoda, D.; Morrison, P. D.; Clarke, B. O. Assimilation of polybrominated diphenyl ethers from microplastics by the marine *Amphipod, Allorchestes compressa*. *Environ. Sci. Technol.* **2014**, *48*, 8127−8134.
(18) Batel, A.; Borchert, F.; Reinwald, H.; Erdinger, L.; Braunbeck, T. Microplastic accumulation patterns and transfer of benzo[a]pyrene to adult zebrafish (*Danio rerio*) gills and zebrafish embryos. *Environ. Pollut.* **2018**, *235*, 918−930.
(19) Lu, Y.; Zhang, Y.; Deng, Y.; Jiang, W.; Zhao, Y.; Geng, J.; Ding, L.; Ren, H. Uptake and accumulation of polystyrene microplastics in Zebrafish (*Danio rerio*) and toxic effects in liver. *Environ. Sci. Technol.* **2016**, *50*, 4054−4060.
(20) Khan, F. R.; Boyle, D.; Chang, E.; Bury, N. R. Do polyethylene microplastic beads alter the intestinal uptake of Ag in rainbow trout (*Oncorhynchus mykiss*)? Analysis of the MP vector effect using in vitro gut sacs. *Environ. Pollut.* **2017**, *231*, 200−206.
(21) PlasticsEurope, An analysis of European plastics production, demand and waste data. In *Plastics - the facts 2016*; PlasticsEurope, 2016.
(22) Fok, L.; Cheung, P. K. Hong Kong at the Pearl River Estuary: A hotspot of microplastic pollution. *Mar. Pollut. Bull* **2015**, *99*, 112−118.
(23) Masura, J.; Baker, J. E.; Foster, G. D.; Arthur, C.; Herring, C. *Laboratory methods for the analysis of microplastics in the marine environment : recommendations for quantifying synthetic particles in waters and sediments*; NOAA, 2015.
(24) 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*, 10377−10385.
(25) Chen, Q.; Reisser, J.; Cunsolo, S.; Kwadijk, C.; Kotterman, M.; Proietti, M.; Slat, B.; Ferrari, F. F.; Schwarz, A.; Levivier, A.; Yin, D.; Hollert, H.; Koelmans, A. A. Pollutants in plastics within the North Pacific Subtropical Gyre. *Environ. Sci. Technol.* **2018**, *52*, 446−456.
(26) Norén, F. *Small plastic particles in Coastal Swedish waters*; KIMO, 2007.
(27) Dris, R.; Gasperi, J.; Saad, M.; Mirande, C.; Tassin, B. Synthetic fibers in atmospheric fallout: A source of microplastics in the environment? *Mar. Pollut. Bull.* **2016**, *104*, 290−293.
(28) Guan, Y.-F.; Wang, J.-Z.; Ni, H.-G.; Luo, X. J.; Mai, B. X.; Zeng, E. Y. Riverine inputs of polybrominated diphenyl ethers from the Pearl River Delta (China) to the coastal ocean. *Environ. Sci. Technol.* **2007**, *41*, 6007−6013.
(29) Cheung, P. K.; Fok, L.; Hung, P. L.; Cheung, L. T. O. Spatio-temporal comparison of neustonic microplastic density in Hong Kong waters under the influence of the Pearl River Estuary. *Sci. Total Environ.* **2018**, *628-629*, 731−739.
(30) Lin, L.; Zuo, L.-Z.; Peng, J.-P.; Cai, L.-Q.; Fok, L.; Yan, Y.; Li, H.-X.; Xu, X.-R. Occurrence and distribution of microplastics in an urban river: A case study in the Pearl River along Guangzhou City, China. *Sci. Total Environ* **2018**, *644*, 375−381.
(31) Lusher, A. L.; Burke, A.; O'Connor, I.; Officer, R. Microplastic pollution in the Northeast Atlantic Ocean: Validated and opportunistic sampling. *Mar. Pollut. Bull.* **2014**, *88*, 325−333.
(32) Mai, L.; Bao, L.-J.; Shi, L.; Wong, C. S.; Zeng, E. Y. A review of methods for measuring microplastics in aquatic environments. *Environ. Sci. Pollut. Res.* **2018**, *25*, 11319−11332.
(33) Bureau, G.S. *Guang Dong Statistical Yearbook*; China Statistics Press: Beijing, 2001.
(34) Yonkos, L. T.; Friedel, E. A.; Perez-Reyes, A. C.; Ghosal, S.; Arthur, C. D. Microplastics in four estuarine rivers in the Chesapeake Bay, U.S.A. *Environ. Sci. Technol.* **2014**, *48*, 14195−14202.
(35) 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*, 582−591.
(36) Lima, A. R. A.; Costa, M. F.; Barletta, M. Distribution patterns of microplastics within the plankton of a tropical estuary. *Environ. Res.* **2014**, *132*, 146−155.
(37) Veerasingam, S.; Mugilarasan, M.; Venkatachalapathy, R.; Vethamony, P. Influence of 2015 flood on the distribution and occurrence of microplastic pellets along the Chennai coast, India. *Mar. Pollut. Bull.* **2016**, *109*, 196−204.
(38) Barnes, D. K. A.; Galgani, F.; Thompson, R. C.; Barlaz, M. Accumulation and fragmentation of plastic debris in global environments. *Phil. Trans. R. Soc. B* **2009**, *364*, 1985−1998.

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817

(39) Lima, A. R. A.; Barletta, M.; Costa, M. F. Seasonal distribution and interactions between plankton and microplastics in a tropical estuary. *Estuarine, Coastal Shelf Sci.* **2015**, *165*, 213−225.
(40) Hurley, R.; Woodward, J.; Rothwell, J. J. Microplastic contamination of river beds significantly reduced by catchment-wide flooding. *Nat. Geosci.* **2018**, *11*, 251−257.
(41) Zhang, K.; Xiong, X.; Hu, H.; Wu, C.; Bi, Y.; Wu, Y.; Zhou, B.; Lam, P. K. S.; Liu, J. Occurrence and characteristics of microplastic pollution in Xiangxi Bay of Three Gorges Reservoir, China. *Environ. Sci. Technol.* **2017**, *51*, 3794−3801.
(42) Vianello, A.; Boldrin, A.; Guerriero, P.; Moschino, V.; Rella, R.; Sturaro, A.; Da Ros, L. Microplastic particles in sediments of lagoon of Venice, Italy: First observations on occurrence, spatial patterns and identification. *Estuarine Coastal Shelf Sci.* **2013**, *130*, 54−61.
(43) Pazos, R. S.; Bauer, D. E.; Gómez, N. Microplastics integrating the coastal planktonic community in the inner zone of the Río de la Plata estuary (South America). *Environ. Pollut.* **2018**, *243*, 134−142.
(44) Anderson, P. J.; Warrack, S.; Langen, V.; Challis, J. K.; Hanson, M. L.; Rennie, M. D. Microplastic contamination in Lake Winnipeg, Canada. *Environ. Pollut.* **2017**, *225*, 223−231.
(45) Kapp, K. J.; Yeatman, E. Microplastic hotspots in the Snake and Lower Columbia rivers: A journey from the Greater Yellowstone ecosystem to the Pacific Ocean. *Environ. Pollut.* **2018**, *241*, 1082−1090.
(46) Sadri, S. S.; Thompson, R. C. On the quantity and composition of floating plastic debris entering and leaving the Tamar Estuary, Southwest England. *Mar. Pollut. Bull.* **2014**, *81*, 55−60.
(47) 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*, 11863−11871.
(48) Naidoo, T.; Glassom, D.; Smit, A. J. Plastic pollution in five urban estuaries of KwaZulu-Natal, South Africa. *Mar. Pollut. Bull.* **2015**, *101*, 473−480.
(49) 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.
(50) Cole, M.; Lindeque, P.; Halsband, C.; Galloway, T. S. Microplastics as contaminants in the marine environment: A review. *Mar. Pollut. Bull.* **2011**, *62*, 2588−2597.
(51) Arthur, C.; Baker, J.; Bamford, H. *Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris*, September 9−11, 2008, University of Washington Tacoma: Tacoma, WA, USA, 2009.
(52) Reisser, J. W.; Slat, B.; Noble, K. D.; Plessis, K. D.; Epp, M.; Proietti, M. C.; De Sonneville, J.; Becker, T.; Pattiaratchi, C. The vertical distribution of buoyant plastics at sea: an observational study in the North Atlantic Gyre. *Biogeosciences* **2015**, *12*, 1249−1256.
(53) Eo, S.; Hong, S. H.; Song, Y. K.; Han, G. M.; Shim, W. J. Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. *Water Res.* **2019**, 228.
(54) Siegfried, M.; Koelmans, A. A.; Besseling, E.; Kroeze, C. Export of microplastics from land to sea. A modelling approach. *Water Res.* **2017**, *127*, 249−257.
(55) Hoornweg, D.; Bhada-Tata, P. *What a waste: A global review of solid waste management*; The World Bank: Washington, DC, 2012.
(56) *Annual Data, China Statistical Yearbook, 2010−2018*; National Bureau of Statistics of China Ed.; China Statistics Press Beijing.
(57) Dris, R.; Imhof, H.; Sanchez, W.; Gasperi, J.; Galgani, F.; Tassin, B.; Laforsch, C. Beyond the ocean: Contamination of freshwater ecosystems with (micro-) plastic particles. *Environ. Chem.* **2015**, *12*, 539−550.

DOI: 10.1021/acs.est.9b04838
Environ. Sci. Technol. 2019, 53, 11810−11817