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Controlling factors of microplastic riverine flux and implications for reliable monitoring strategy

Mengyu Bai [1,2,3], Yan Lin [4], Rachel Hurley [4], Lixin Zhu[1,2,3], Daoji Li [1,2,3]*

[1] State Key Laboratory of Estuarine and Coastal Research, East China Normal University, 500 Dongchuan Road, Shanghai 200241, China

[2] Plastic Marine Debris Research Center, East China Normal University, 500 Dongchuan Road, Shanghai 200241, China

[3] Regional Training and Research Center on Plastic Marine Debris and Microplastics, IOC-UNESCO, 500 Dongchuan Road, Shanghai 200241, China

[4] Norwegian Institute for Water Research (NIVA), Gaustadelléen 21, 0349 Oslo, Norway

Abstract: A significant proportion of marine plastic debris and microplastics is assumed to be derived from river systems. In order to effectively manage plastic contamination of the marine environment, an accurate quantification of riverine flux of land-based plastics and microplastics is imperative. Rivers not only represent pathways to the ocean, but are also complex ecosystems that support many life processes and ecosystem services. Yet riverine microplastics research is still in its infancy, and many uncertainties still remain. Major barriers exist in two aspects. First, nonharmonized sampling methodologies make it problematic for compiling data across studies to better estimate riverine fluxes of microplastics globally; Second, the significant spatiotemporal variation of microplastics in rivers which was affected by the river characteristics, MPs properties, etc. also have important influence on the estimation of riverine MPs fluxes. In this study, we made a comprehensive review from the above two aspects based on published peer-reviewed studies and provide recommendations and suggestions for a reliable monitoring strategy of riverine MPs, which is beneficial to the further establish sampling methods for rivers in different geographical locations. Besides, methods for achieving a high level of comparability across studies in different geographical contexts are highlighted. Riverine microplastic flux monitoring is another important part of this manuscript. The influential factors and calculation methods of microplastic flux in rivers are also discussed in this paper.

Key words: River water; Microplastic; Monitoring strategy; Sampling methodologies; Flux

1. Introduction

Microplastics (MPs), defined as plastic debris smaller than 5 mm [1], are derived from the breakdown of larger plastic items (e.g., fibrous or fragmented MPs) or are deliberately produced (e.g., microbeads or glitter). The mismanagement of plastic litter worldwide has led to the widespread occurrence of MPs [2]. MPs have similar shapes and sizes to plankton and can be unintentionally ingested by fish, filter feeders and planktivores [3, 4]. After entering organisms, MPs can be transferred through the food web, which has been detected in both invertebrates and zooplankton [5, 6]; although, the true risk associated with this is still yet to be fully articulated. MPs can be transported over long distances to remote islands [7], polar regions [8], Arctic Sea ice [9], and mountain lakes in Mongolia [10] by physical factors (such as wind, ocean currents and river flow) [11-13]. The ocean is viewed as a major MP sink, whereas rivers are viewed as important pathways for plastic litter transport into the oceans. Meijer et al. [14] revealed that 1000 rivers globally transport 0.8-2.7 million metric tonnes of plastic litter each year to the ocean, accounting for 80% of annual global emissions. The Danube River was estimated to transport 1553 tonnes of plastic debris to the Black Sea annually at a rate of 7.5 $mg/m^3•s$ [15]. River basins are the main contributors of plastics to estuaries, where transportation and accumulation patterns are determined by the fluctuation of flow regimes [16]. Other studies have also confirmed river basins as the major sources of inland MPs to estuaries, as well as exporters of MPs to the oceans [17-20].

Yet, river environments represent a crucial component of the global hydrosphere and biosphere, supporting important ecological diversity. Rivers are often described in terms of conduits of plastic to the ocean, but they are complex and dynamic systems that can accumulate, store, and remobilize plastic particles over different spatial and temporal scales. The majority of plastics are produced, consumed, and disposed of on land [21]. Due to this quantity and extent of MP sources, rivers often exhibit elevated MP concentrations compared to the marine environment [22]. High MPs concentrations have been found in both river sediment [23-25] and surface waters [26, 27]. Many riverine ecosystems globally are expected to be exposed to MP in water and sediments, and these organisms also need protection from the potential adverse effects of contamination. Especially given the high degree of complexity in river systems, the fate and transport of MP remains relatively under-researched. The behavior of plastic litter in freshwater, especially in rivers, varies greatly from that in the marine environment, and freshwater-specific studies are required to investigate relevant processes of MP movement and accumulation in rivers. Inconsistent sampling methods hinder the possibility to compare between results, and different sample types reveal different snapshots of riverine MP contamination [28]. Hermsen et al. [29] established a means for assessing methodological and data quality based on 10 criteria, and found that the information integrity of most studies needs to be further improved. Further publications have since proposed reporting guidelines for MP studies, covering topics from field sampling to quantification from laboratory data [30].

River stratification and hydrodynamic action have a great influence on the distribution and transport characteristics of plastic litter [31-33]. Artificial structures, such as dams [34-36], bridges [37] and human-created tributaries [38], hydropower station [39], as well as natural characteristics, such as riparian vegetation [40] and river curvature [41], geographical factors determined by the shape of the river [42] will affect river flow and may lead to the accumulation of plastic litter and MP. Hydrological conditions are also an important control on MP fate and transport, affecting the partitioning between the water and sediment phase [28, 43, 44], influencing the movement of MPs with channel bed sediments [43, 45, 46], and acting as a control on the export of MPs downstream [47].

Currently, the reported values from literatures utilize an array of sampling methods, size limits, processing methods, and different instrumentation under different conditions. There is also variability in the definition of MPs (such as the size categories analyzed) and the degree of quality control implemented into methodologies. Several studies have now reviewed the different sampling methodologies for assessment of MP occurrence in water and sediments [42, 48-50]. This article instead specifically focuses on the emission of MPs from rivers to the ocean on a certain time scale (riverine MP flux) and reviews the approaches to monitoring and establishing flux estimates within this context. This review intends to address the following key issues related to MP monitoring: (I) comparative analysis of the different sampling methods; and (II) review of the the calculation and influential factors of river MP flux, and requirements for improvement of the methods for both short-term field sampling and long-term monitoring to achieve more accurate river MP flux calculations.

2. Materials and methods

An extensive and systematic literature search was conducted in ISI Web of Science (WOS), Scopus, Google Scholar and Elsevier Science Direct for this review. The following key words were used in literature collecting: “microplastic(s)”, “river”, “fresh water”, “riverine”, “plastic flux” and “stream”. A total of 83 scientific articles and reviews about riverine MPs fluxes published before June, 2021 were included and reviewed. Data analysis and visualization were based on Python 3.8.5 and IBM SPSS Statistics 26.

This review includes global (locate in 5 Asian countries, 8 European countries, 3 countries in the Americas, 1 African country and 2 Oceanian countries) rivers with different discharge, and covers multiple types of rivers locating in different climatic zones. The physical geography classifications of rivers include tributaries (e.g., Ottawa River and its tributaries, Keelung River-the tributary of Tamsui River), river estuaries (e.g., Tamar estuary, Yangtze River estuary), and the main stream of larger river catchments (e.g., Beijiang River, Pearl River).

3. Sampling methodologies for MPs in rivers

To link the sources and fate of MPs in freshwater environments, it is necessary to establish a unified approach to enhance data comparability. This review will introduce sampling methodologies for MPs in rivers based on the different equipment and deployment locations.

3.1 Sampling equipment

By reviewing studies about MPs in river water, sampling equipment can be summarized as the following three types: i) Direct sampling-using precleaned stainless buckets, water samplers, glass jars or other containers to directly collect surface water. In some studies, the direct sampling process is accompanied by volume-reduction operations to decrease the volume of water; ii) Pumps - where various types of submersible pumps are used to draw a larger volume of water; and iii) Nets - which include Manta, Neuston and other surface nets that are widely used in water sampling. Some types of nets (e.g., Bongo net) can also acquire the samples from lower layers of the water column. A detailed comparison of the sampling approaches is shown in Table 1.

Table 1 Comparison of the specific sampling methodologies of MP in river water

	Commonly used	Types	Sampling volume	Sampling layer	Water volume calculating method	Advantages	Disadvantages
Direct sampling	Stainless bucket; Water sampler; Glass jar; Niskin bottles; Other containers	Laboratory filtration; Volume reduced (stainless steel sieves / bolting-silk)	Min (less than 100 liters)	Surface water layer; Surface microlayer; Limited water column	Use containers for volume determination	Cheap, easy to operate and quick; Smaller size and fibrous MPs can be captured; Accurate and controllable volume of filtered water; Boat is not necessary	Small sampling volume; Sample is easily contaminated; Possible to transport bulky samples to the lab; Hard to sample water column in deep water; Need to bring bulky samples to the lab (without volume reduced process)

Pump	Teflon pump; Peristaltic pump; Submersible pump; Plankton pump	Laboratory filtration; Volume reduced (stainless steel sieves / bolting-silk)	Medium (several cubic meter)	Full water layer	Flowmeter; Rated power multiply by running time	Relatively cheap, easy to operate and quick; Relatively smaller size and fibrous MPs can be captured; Accurate and controllable volume of filtered water; Possible to sample the water column	Relatively small sampling volume; Need power supply and boat; Relatively higher cost; Possible to transport bulky samples to the lab; Need to bring bulky samples to the lab (without volume reduced process)
Net	Manta net; Neuston net; Plankton net; Bongo net and other nets	Volume reduced	Max (up to a few hundred cubic meters)	Full water layer	Flowmeter; Towing length multiply by net opening size	Large volume sampling; Net aperture can be changed according to the actual situation; Volume reduced on site; Possible to sample the water column(plankton net)	Net and manual sample transfer may bring into contamination; Risk of clogging; Expensive and not easy operation; Mesh sizes impose a relatively large lower size limit, potentially underestimating MP concentrations

The different approaches to sampling MPs in river water can be methodologically divided into bulk water sampling and volume-reduced sampling [51]. The primary difference is that the former method collects the entire volume of the sample without performing volume reduction (e.g., use of filters or sieves) in situ. Bulk sampling has advantages including: (i) non-in-situ filtration, which may reduce potential contamination; (ii) samples with low concentrations or small particle sizes can be obtained by collecting large volume samples; and (iii) reducing subjective errors by quantitative sample collection. Direct sampling and pumping can be all categorized together as bulk sampling.

Volume-reduced sampling usually employs nets or sieves [52] to reduce the sample volume and concentrate the particulate load for MP analysis. Net sampling represents a commonly used volume-reduced method. Manta and Neuston nets are often used to sample surface waters [53], whilst stationary conical driftnets and hand nets are also used in some studies [15, 32, 54]. Nets can be deployed at the river surface to capture the floating MP components, as well as in the middle and lowermost sections of the water column with the use of Bongo nets, for example [50]. All the nets should be equipped with flow metres whilst in operation to calculate the volume of water that has been sampled. Nash et al. [55] advised using two flowmeters simultaneously, with one equipped inside the net and another equipped outside the net to better understand how the clogging of the net may affect filtration efficiency. Figure 1 depicts three typical nets used in studies: (a) Manta net, (b) Bongo net and (c) Plankton net.

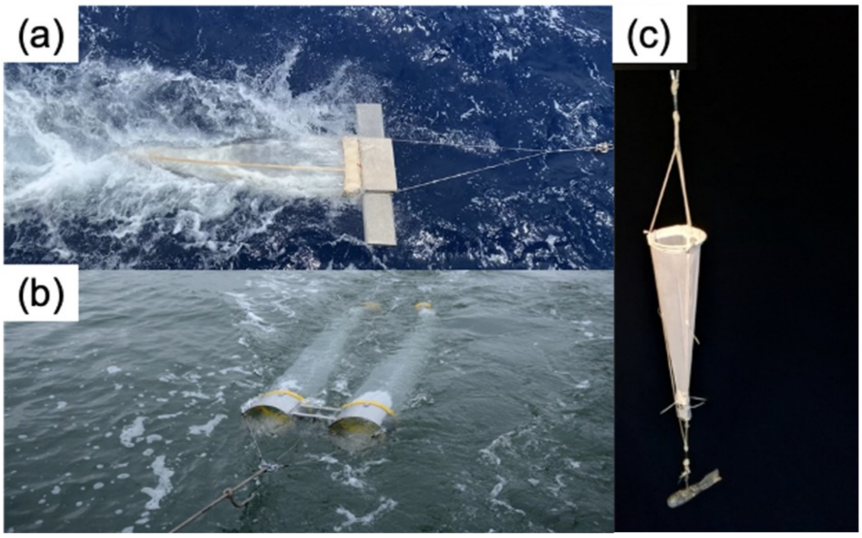


Figure 1 Three normal nets in practical application (a)Manta net,(b)Bongo net,(c)Plankton net.

By reviewing the sampling equipment for MPs in river water, it was found that net sampling with mesh sizes between 100-335 μm accounted for more than 50% of studies, which is represented in Figure 2(a). Clogging risks (which may lead to the sample loss or backflow) may occur when the mesh sizes are smaller than the given range, while more particles will escape from the net if the mesh sizes are too large. It also indicates that net sampling is more widely used and therefore offers greater potential for data comparability. Figure 2(b) shows the geographical distribution of river MPs studies, which refers to 5 Asian countries, 8 European countries, 3 countries in the Americas, 1 African country and 2 Oceanian countries) rivers with different flow regimes (monthly discharge ranges between 4.15×10^5 to 7.41×10^{10} m^3). Kataoka et al.[56] conducted surveys in 29 rivers in Japan, which is the country with the most rivers involved in sampling. The U.S.A has the second largest number of sampled rivers. The third largest is China, which has 13 investigated rivers in this review.

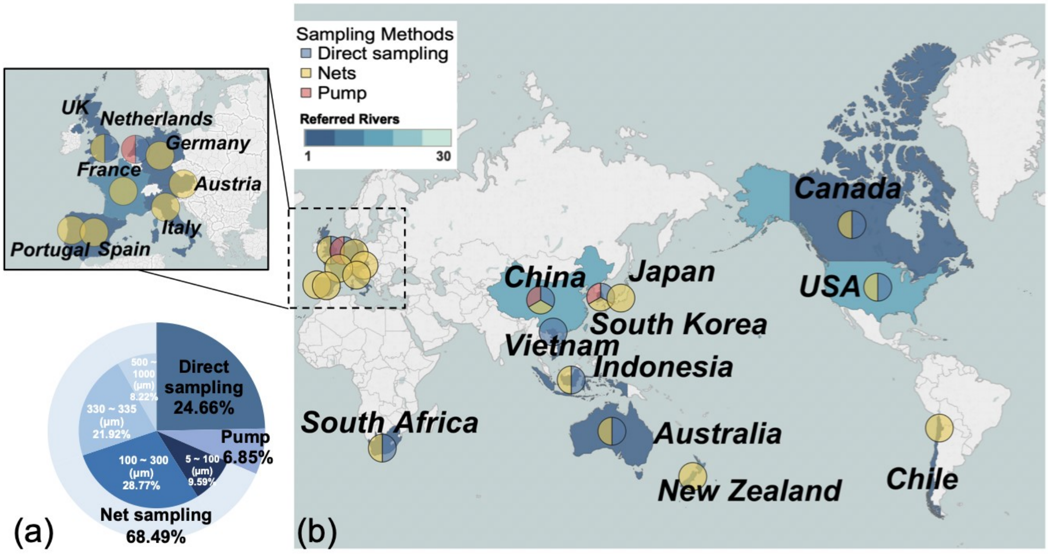


Figure 2 (a) The proportions of three sampling methods in reviewing studies. (b)Global rivers discussed in this review.

Small samples volumes and use of different (coarser) sieve or mesh sizes can lead to inaccurate determination of MPs concentrations [57-60]. Several studies [61, 62] used small volume sampling methods (e.g., 1 L surface water in rivers) and the MPs were found to be fibrous or mostly fibrous (80%). By comparing the results from a 1 L surface grab sampling method with a 335 μm Neuston tow, Barrows et al. [57] found that the concentration of MPs (n/L) of the former is 3 orders of magnitude higher than the latter. Furthermore, Norén et al. [63] reported that plastic fibers filtered using an 80 μm mesh size net were up to 5 orders of magnitude more abundant than those obtained using a 450 μm mesh size net. Dris et al. [58] reported that a plankton net with 100 μm mesh size collected 100 times the amount of MPs compared with sampling by 330 μm Manta net. Studies using nets for sampling must consider the minimum mesh size that can be feasibly used in order to sample small microplastic particles and report more accurate MP concentrations, whilst balancing trade offs related to clogging and the flow conditions of the river.

MP fibers are typically long and thin and so the portion captured by the net may depend on several conditions such as the orientation and curvature of the particle and the flow conditions. This represents an additional important consideration when using nets to sample river water: fibers observed in net samples may represent an unknown proportion of the total fiber contamination at that sampling location. A field survey conducted with 100 L water pumped and a 60 μm sieve, the overall mean MP concentrations in the Yangtze River and East China Sea were respectively 157.2 ± 75.8 n/m^3 and 112.8 ± 51.1 n/m^3, with fibers accounting for more than 80% of the total [64]. Zhao et al.[65] showed that a 32 μm sieve can retain abundant fibers by filtering the surface water in the Yangtze River Estuary and East China Sea, which contained 79.1% and 83.2% fibrous MPs, respectively. Studies using sieves or nets with small mesh sizes will collect large quantities of fibrous MPs, although data comparability is poor compared with using Manta trawls with mesh sizes between 100-335 μm as they are less often used in sampling activities.

3.2 Sampling locations

Flux estimates normally utilize measured plastic data and extrapolate to the total river channel or river catchment based on factors of time or discharge [64, 66]. However, several factors may influence the movement of plastic particles from initial input to eventual release to the ocean, such as deposition, trapping, and remobilisation. Besides, the spatial context of the sampling locations also introduces additional uncertainty in the flux estimates. Exploring sampling locations is conducive to establishing scientific monitoring methods according to the specific conditions of river. It is also important to consider the deployment location for different sampling methods, as this may also determine appropriate sampling locations. This section will review aspects related to sampling locations.

Firstly, from the perspective of deployment location, sampling techniques can be deployed from bridges or from the riverbanks, for example with buckets or nets attached to retractable rods. Also, sampling activities can be conducted from the research vessels in the rivers. Figure 3 presents a schematic diagram showing specific sampling measures.

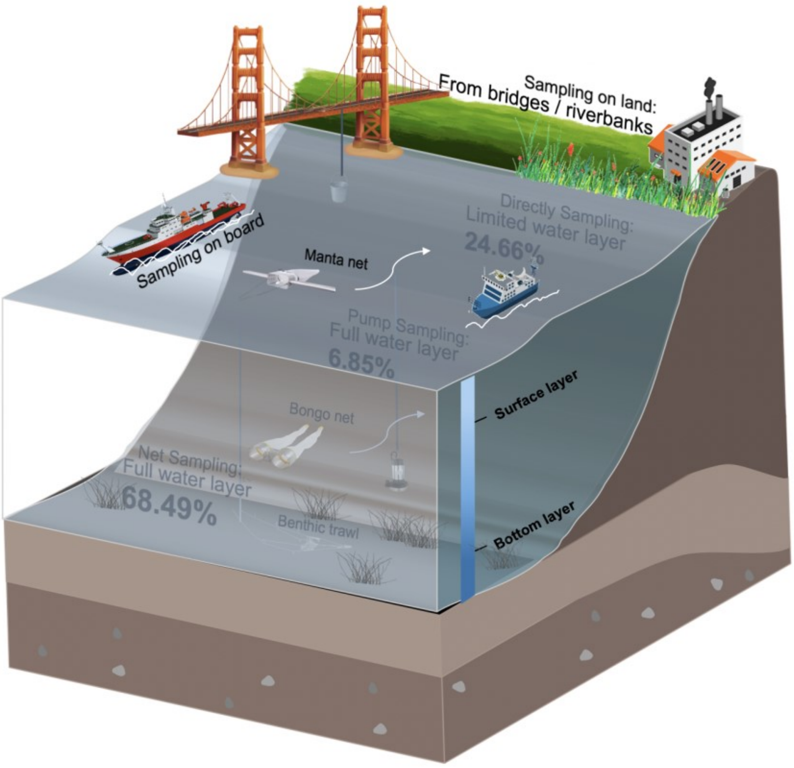


Figure 2 Schematic diagram of sampling measures with the proportion of each method in the reviewed papers

Research vessels may hinder the flow of water and interfere with the stability of the water body, which may influence the effectiveness or representativeness of sampling. Sampling from bridges or riverbanks is less heavily affected by this factor, although introducing the net into the river may still interrupt flows and generate additional water turbulence. The mode of deployment will depend on the geographical context of the site. For example, deploying from bridges necessitates the occurrence of this form of infrastructure that also offers safe access. The use of research vessels requires that rivers are over a certain depth, and that vessels are available in the vicinity or can be entered into the water. Sampling from river banks requires access and stability, and is only recommend for narrower channels where it is possible to take representative samples from within reach of the bank.

When selecting riverine MP monitoring sites, the following aspects should be considered: representativeness, accessibility, hydrology, and stability (e.g., potential for long-term monitoring) [67]. Replicate samples, or simultaneous multiple samples from different points across the river cross-section can help to account for potential variability in MP loads. For example, small scale spatial variability across a river cross-section or temporal variability across a short (e.g., 1 hour) sampling period. Wong et al. [68] emphasized the importance of sampling locations to the MPs flux estimation. Complex hydrodynamic processes in estuaries may alter the transport and distribution of MPs (e.g., sedimentation, aggregation, resuspension, hyporheic exchange and biological effects including biofouling, ingestion and excretion [46, 69, 70]). Xiong et al.[71] concluded that the plastic debris might accumulate in the river estuary area due to tidal activities, and emphasized that MPs flux calculations that are only based on the data obtained from the river estuary will lead to an overestimation. González et al.[67] recommended to sampling upstream for reference to avoid the influence of estuaries, and the selection of exact monitoring sites depending on the actual situation on site (e.g., population density, waste discharge source, possibility of sampling location implementation). To minimize these effects, multimedium, multi-layer sampling, and long-time scale monitoring are effective methods. It is recommended to collect samples from multiple reaches of the river (e.g., covering the upper reaches of the river, river outlets and the place where the river meets the sea). Cowger et al. [72] also advised for the adoption of depth-integrated sampling.

Understanding river characteristics is essential for sampling. Sampling strategies should be adjusted accordingly, such as with respect to suspended sediment concentration, flow velocity/discharge, water depth, functional zoning of river basins and tidal influence. Studying the transport patterns at different time scales can help to better understand the fate of MPs, as rivers are globally highly diverse and have various features [48]. A sampling site with features that reduce turbulence complexity (e.g., homogeneous bed characteristics, gentle curve of the riverbank, stable with little interference) need to be identified and included in monitoring. Site metadata should also be recorded to allow for effective interpretation of observed MP concentrations, such as flow velocity, water level, suspended sediment concentrations, meteorological conditions (including antecedent conditions), hydrogeomorpholocial context and bed substrate type. To investigate the impact of point and regional pollution sources, for example, artificial facilities, sewage treatment plants and population density (e.g., tourist areas, rural and densely populated areas), sampling activities should cover the upper and lower reaches of the sources [22, 73].

4. Riverine MPs distribution, transport and flux

4.1 Characteristics of MPs in rivers

The occurrence of chemical and colour compositions of MPs in river water according to reviewed papers were analyzed and sorted, and the proportion of occurrence frequency was calculated (Figure 4(a), (b)) (See Table S1 Microplastic composition in river water in Supporting Information). After analyzing, it was found that the most common polymer type detected in rivers is polyethylene (PE) (42%), followed by polypropylene (PP) (30%) and polystyrene (PS) (11%) (Figure 4a). Other common polymer types like PET, polyamide and polyester are also often observed. PE and PP comprised the majority of polymers in the studied rivers [64, 66, 71, 74-80], which represent low-density plastic types. Besides, PE and PP are commonly used in disposable plastic products. While PS foam (normally used in food packaging and shockproof container) was the most abundant MP category by number in Hongkong waters [81]. In a study of river in Saigon River, Vietnam, the percentage of polyolefin and PS accounted for the most while PS foam food box scraps accounted for a very large proportion [37].

Colourless MPs (including transparent and white were dominant in the articles included in this review (Figure 4 (b)). This phenomenon has been reported in many previous studies about MPs in river water [24, 68, 82]. The colourless aspect may be specifically engineered (e.g., from white textiles) or caused by fading due to photodegradation. Wong et al. [68] sampled plastic particles in both river water and sediments from the beach, and found that the colourless plastic particles in river water are less abundant than that in sediment. The higher incidence of colourless particles found on river beaches could be interpreted as resulting from greater exposure to ultraviolet light in this context, in comparison to particles moving in the water.

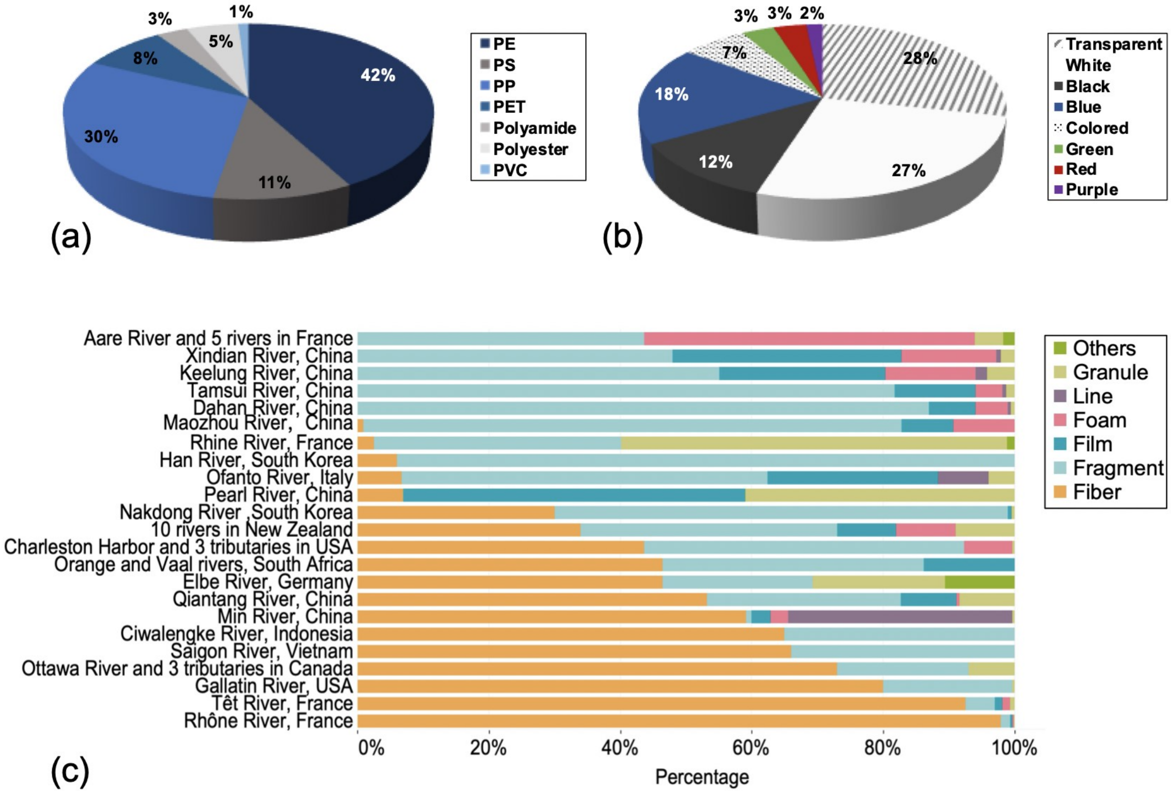


Figure 4 (a) chemical, (b) colour and (c) shape compositions of riverine MPs referenced by the articles discussed in this review. The ratios in (a) and (b) refer to the occurrence frequencies in all reviewed papers, while the ratios in (c) are original data from each independent studies.

MPs from different release pathways partly have their own characteristics, for example, fibrous and small size of MPs are typical features of MPs from sewage treatment plants, which can be used to partly explain the origin of some of the numerous MPs in the shoreline and river shore sediments with size ranges from 63 – 200 μm [83, 84]. MPs derived from tire and road marking found in river water can help to reveal contributions from runoff [85, 86], while the MPs formed from the fragmentation of fish lines and floating rafts also reveal the source of directly discarded plastic waste [79]. It is helpful for the consideration of influencing factors and development of monitoring strategy of MPs flux in rivers by reviewing the major release pathways. Considering the shape information is important for the identification of source, fate and transport for riverine MPs, the original shape compositions data from independent researches was assessed (Figure 4 (c)). At present, the definition of MP shape has a unification of broad categories, but there are still differences in the subdivision process. The shapes were unified here and fall into the following categories: fiber, line, fragment (which includes hard fragment and film-type fragment [87]), film (which includes flake [88]), foam, granule (which includes sphere, microbead and pellet [54, 62, 68, 88-90], opaque and transparent spherules [87, 91]), others (which includes combined [87]).

In general, fibers represent the dominant shape of MPs in rivers [54, 62, 64, 65, 75-77, 79, 80, 92-94]. Several studies emphasized the use of synthetic textiles as a source of riverine MPs pollution, also pointing towards sewage treatment plants as a potentially important pathway [77, 95-97]. Fibers are more easily entrained and maintained in suspension by river flows [43], potentially leading to higher proportions observed in the water than in sediments. In some studies, fragments account for more than 50 % of the MP concentrations [24, 68, 78, 82, 87, 88, 98, 99], which may be derived from the fragmentation of plastic products [66, 100-102]. Lahens et al. [93] also advised to investigate the role of in-situ macroplastic fragmentation as a source of MPs to rivers. Thompson et al.[103] found 9 types of MPs, including fibers and fragments typically derived from synthetic fabric, packages and rope in 23 out of 30 sedimentary samples around Plymouth, UK.

MPs concentration in river water is an important data source which can be used to extrapolate riverine MPs flux. Yet, an important problem arises in data extrapolation, related to MPs sizes. Several studies have shown that the lower limit of the mesh size is not equal to the detection limit of MP samples [79, 93, 98, 104]. Recently, Koelmans et al. [105] provide a method to mitigate size range differences: correction factors can be used to convert MPs sizes into three default size ranges. Figure 5 shows the MPs density in global rivers in this review. To reduce the variation of different sampling size ranges, the data shown in Figure 5 has been standardized with the correction factors [105] (See Table S2 Correction factors used for mitigating size range differences in Supporting Information). MP densities in rivers show great differences, which may be attributable to the individual or combined effects of the influencing factors mentioned above, such as sampling methods, river morphology, watershed conditions, abnormal weather conditions, and so on. It is likely that many rivers exhibit a unique microplastic assemblage based on the quantity and diversity of sources and the hydrogeomorphological conditions of the river, which influence MP fate and the concentrations and particle types observed in a given sample.

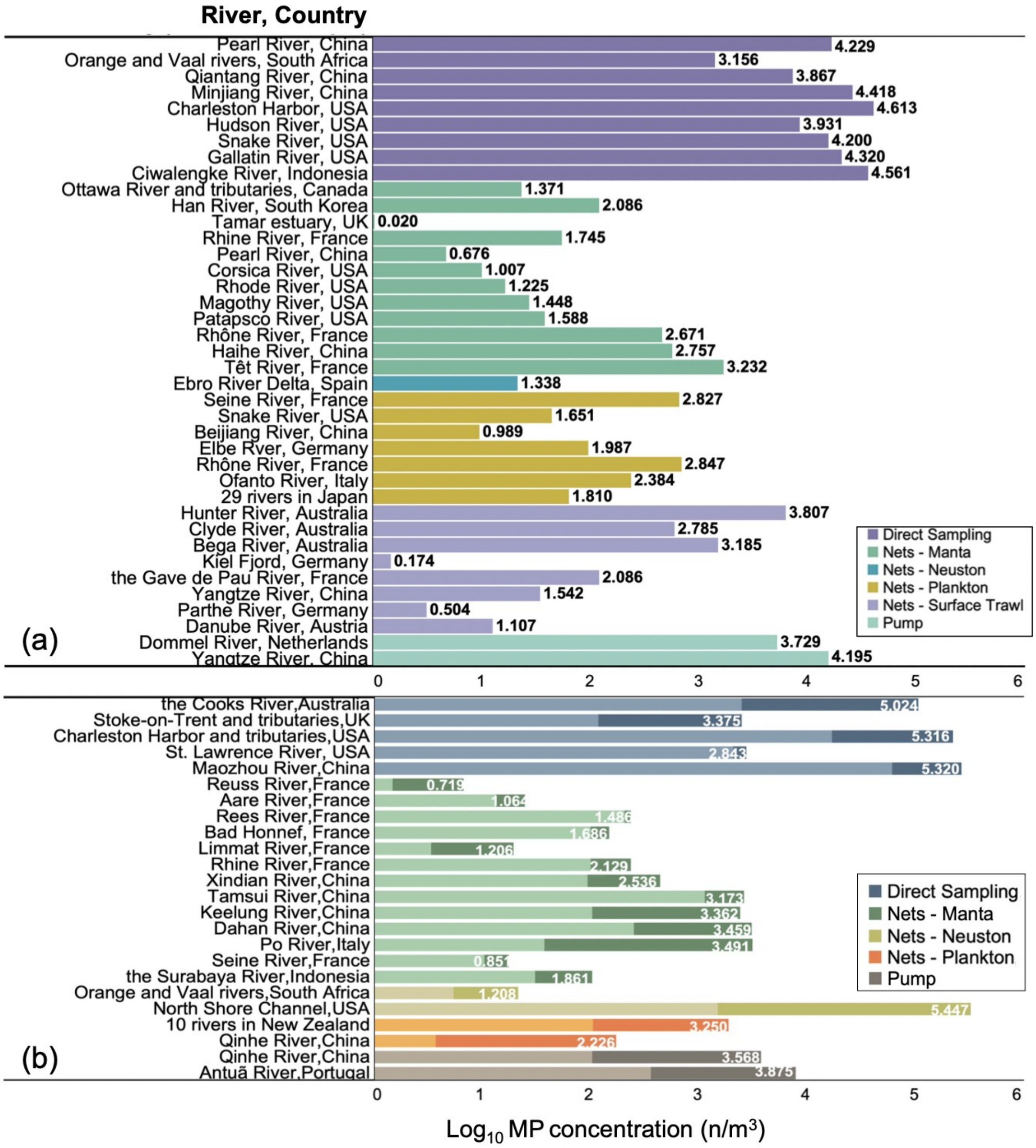


Figure 5 MP concentration (n/m^3) in global rivers (data shown in logarithmic form); (a) numbers represent the mean values; (b) numbers represent the quantitative difference between the max and min values, lighter colored columns represent the minimum MP concentration and darker colored columns represent the maximum MP concentration.

4.2 Fate and transport processes

Hydrology (e.g., channel morphology, turbulence and tidal influence), spatiotemporal variability in sources and environmental processes, artificial factors (e.g., basin population and area, watershed function zoning), and the characteristics of MPs (small size thus easily widely dispersed with currents and hydrodynamic processes) will influence the transport behaviors of

MPs in the freshwater environment [42, 106-109]. This is relevant for sampling MP in rivers and understanding what monitoring data conveys, which can also influence the accuracy of flux estimates. Currently, many fate and transport processes governing MP distributions in rivers are poorly understood and more research is needed to identify and unpick the dominant mechanisms. Sampled MP from rivers represent a snapshot of concentrations and information about possible fate and transport processes should be collected to aid in the interpretation of this data to evaluate what it means in the context of riverine flux.

In rivers, particles may partition between the water and sediment phase based upon numerous factors, such as particle density, flocculation and flow velocity. In the water column, the movement of particles is not uniform and MPs are not evenly distributed in the water layer. The density of a type of plastic imparts an important control over the depth of occurrence in the water [110, 111], whilst the biodegradability, shape, oxidation resistance, flocculation / aggregation, surface properties and degree of biofilm formation are also influencing factors [112]. Low-density MPs mostly float close to the surface of rivers and denser MPs could be expected to accumulate at the bottom of rivers or buried in the sediments [33, 35, 36, 113]. A study conducted in the Nakdong River showed the MP concentration in surface river water was 3 times higher than that in bottom water [78]. Yet, changes in MP particle density may occur as a result of biochemical processes, including surface biofilm generation [114], ageing and leaching of additive chemicals [115, 116], and lead to the change of settling rates [117]. Biofilms can be easily formed on the surface of plastics in the marine environment and attract adherence by alages and invertebrates, increasing the sinking speed [118]. MPs may sink to the bottom of the water layer through biofouling by organisms and accumulate in sediments [118-120]. Scherer et al. [121] found that the MP abundance in bottom sediment is 6×10^6 times higher than that in the overlying water, and the studied results in the Nakdong River [78] showed that the MP content in sediment is 2827 times higher than that in the water column. However, several studies identify high density polymer types in water samples and low density – and theoretically buoyant – polymer types in the sediments [78, 93, 113, 122]. This is partly due to turbulent flow, which is likely to lead to entrainment and mixing of particles within the water column. An uneven distribution of MPs in the vertical profile of rivers has been reported [123], which was mainly affected by fluvial hydrodynamics. Water turbulence below the surface may mix particles with a density close to that of the surrounding water, and the density and shape of small items and particles will also affect their rising or sinking speed [67]. Drummond et al. [46] found that, for MPs smaller than 100 μm, retention in river sediments can be substantially increased with the influence of the hyporheic exchange. Besides, hyporheic abrasion may decrease particle size, thus influencing other variable such as surface area or propensity for biofilm formation, which could further influence fate and transport processes.

The morphological characteristic of a river is a key factor, and the morphology of riverbed forms may also impact the plastic debris travel distance [124]. For example, a sharp drop in MP concentrations was found by Mani et al.[91] in the section of the Rhine River with the lowest bed slope. Concentrations in the river decreased in the water column, which may be attributed to the lowest bed slope and low flow velocity in the river bed [91]. Also in the Rhine river, Klein et al. [84] found a dramatical increase in MPs concentration in sediments near to the confluence of tributaries and the main stream. Similarly, the concentration of MPs in the sediment of the Elbe River has been shown to decrease in the lower part of the river [121].

Natural meteorological events, such as storms and heavy rainfall, are factors that influence the instantaneous concentrations and spatial accumulation of MPs. Barnes et al.[11] found that the wind action of a typhoon and heavy rainfall would increase the speed of MP migration from land into the aquatic environment. According to a study in the Yangtze River Estuary, typhoons are an influencing factor for MPs accumulation in the water environment [80]. Moore et al., [125] showed that surface plastic debris on the California coast near the Los Angeles stormwater conveying system increased from 10 n/m^3 to 60 n/m^3 after a heavy storm, indicating that the storm increased the export of MP from the catchment. A study in southern California coastal water also found that MPs accumulate in coastal areas from less than 1 n/m^3 to 18 n/m^3 after a typhoon and heavy rain event [50]. And the MP abundance in Venoge river water in Switzerland increased 150 times after a rainfall event [126]. Flooding can also lead to a flushing of MPs stored in river sediments and may export MP from catchments or redistribute particles, for example through overbank deposition [43, 45]. The thresholds for MP deposition, remobilization and entrainment have not yet been established for a representative range of MP particle types (sizes, shapes, polymer types), so the hydrological conditions under which sedimentation or transport of particles occurs is still poorly understood.

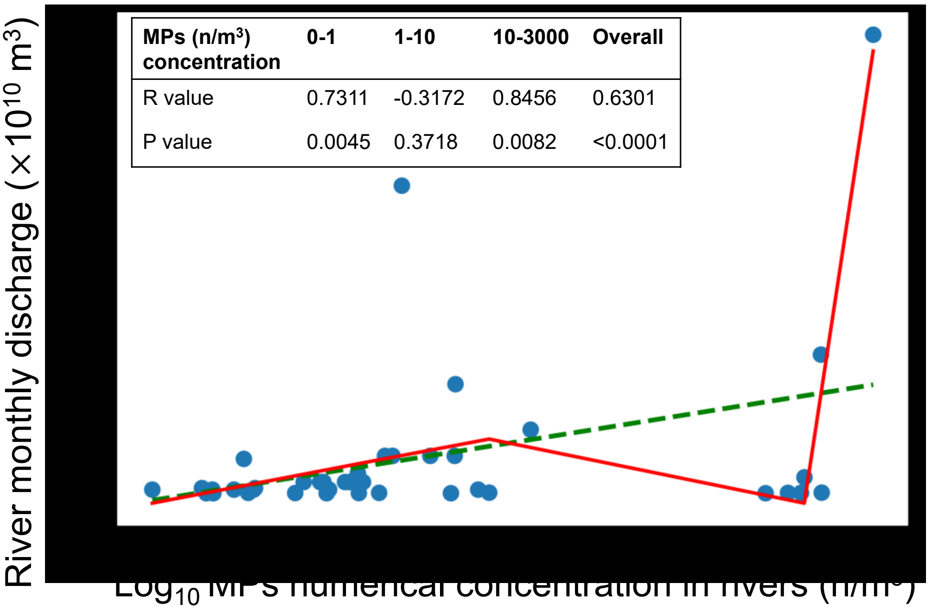
Some MPs may deposit in riverbank or floodplain sediments, due to overbank deposition during flood events. These sediments are subject to less erosion than channel bed sediments, and thus the river corridors can be seen as a possible storage and release component to MPs movement. Scheurer et al. [127] and Christensen et al. [44] all show MPs can accumulate in floodplains adjacent to rivers. The timescales over which particles will be remobilized from these sedimentary archives remains poorly understood, but they could constitute potential long-term legacy sources of MP contamination to the active channel into the future. The geomorphological context will present a dominant control in this case, where channel and floodplain morphology differ significantly, globally. Further research is needed to constrain this potential source and how it contributes to present and future MP fluxes in a variety of rivers.

Artificial facilities, such as dams, bridges and human-made tributaries, may introduce additional turbulence which is more likely to entrain MPs. Xiong et al. [71] found that the MP concentrations downstream of the Three Gorges Reservoir is an order of magnitude lower than that upstream. Lisa et al. [35] reported more MPs in the reservoir water and sediment than in the upstream water and sediment. According to a study combining manual visual and static trawl sampling on the Thu Thiem Bridge in the Saigon River, a high concentration area of large plastic debris was observed near the bridge column, which may be caused by the eddy currents created by the bridge column that carry the plastic debris [37]. MPs may show a similar response, which should be further tested to better understand the fate and transport of MPs in the context of sampling and interpreting monitoring data.

4.3 Spatiotemporal variability and relationship with discharge

MPs are not evenly distributed in different river sections. The complex hydrological conditions near the estuary will affect the spatial distribution of MPs [71]. For example, MP abundance in the surface water of Qinhe River increased from upstream to downstream and reached its highest level in the estuary where MP accumulation zones had formed [79]. Human activities may lead to spatial variations in MPs. A high density of MPs was detected at stations near densely populated areas [66, 80, 81, 83, 128, 129], and the lowest abundance was found at sites located far from urban centres [77, 91]. In a study conducted in the coastal water of South Korea [32], the urban areas had a MP abundance of approximately two times that in rural areas, and there was a strong correlation between the population of the river and coastal basins and the mean MP abundances. In contrast, Kapp et al.[130] found high MP concentrations in a rural site impacted by agriculture where plastic film was widely used. These spatial patterns in potential MP sources can lead to a heterogeneous distribution of MP in the catchment.

As mentioned above, precipitation and storm events can cause large shifts in MPs concentration on short time scales. MPs abundance in rivers shows seasonal variation, which is manifested in the difference of MPs concentration in river water between rainy and dry season. The dominant shape of MPs also has differences between the dry and rainy seasons. In one study, fibers were most abundant in the dry season, while fragments were most abundant in the rainy season [81]. There was a significant difference in MPs concentration in Hongkong waters near the Pearl River Estuary between the rainy (median = 2.657 n/m^3, 0.227 mg/m^3) and dry seasons (median = 0.183 n/m^3, 0.023 mg/m^3) [81]. According to Lebreton et al. [18], 74.5% of the total plastic load emitted from rivers to the ocean occurs between May and October, with a peaks in August and minimal release in January. Soeun et al. [78] estimated that 70-80% of the annual MP load by to the ocean occurred in the wet season. A similar situation also occurred in the sediment of the Brisbane River; that is, the MP concentration in the wet season was higher than that in the dry season [131]. These findings may relate to the greater number of sources that are accessed by precipitation through the increase in connectivity between land and the river. However, in contrast, Fan et al.[74] has found the MP abundances in the river water were notably lower during the wet season, which they attributed to the dilution effect of the precipitation and subsequent increase in discharge. These effect was also reported in the Gallatin River [62] and the Yangtze Estuary [71], where the abundance of MPs is inversely proportional to river discharge. To further discuss the relationship between the MPs concentration and river discharge, the correlation analysis was carried out with the database used in this article and collected river monthly discharge data.



MPs (n/m^3) concentration	0-1	1-10	10-3000	Overall
R value	0.7311	-0.3172	0.8456	0.6301
P value	0.0045	0.3718	0.0082	<0.0001

Figure 6 Correlation between the MPs numerical concentration with river monthly discharge. The red lines represent the fitting curves after subsection regression, respectively for MPs concentration between 0-1,1-10,10-3000 n/m^3; and the green line represents the overall regression analysis of all MPs concentration.

The correlation coefficients between MPs numerical concentration and river monthly discharge is shown in Figure 6. River monthly discharge data were obtained from original articles or U.S.Geological Survey (*usgs.gov*), and the monthly discharge of sampling time is selected. The average MP numerical concentration ranges (n/m^3) were divided into 0-1; 1-10 and 10-3000 (n/m^3), that can be simply described as low, medium and high MPs numerical concentrations. It shows that both low and high MPs numerical concentration have a positive correlation with river monthly discharge (Pearson test, $P<0.05$), while medium concentration are not correlated with river discharge. In general, the MPs numerical concentrations are positively correlated with river discharge (Pearson test, $P<0.0001$). This finding suggests that the flux calculation should not simply multiply MP density by river discharge in a single snapshot in time and instead the measurements need to be integrated over a range of flows, while considering more practical situations (e.g., the influence of hyporheic exchange, biological effects, interception of plants). To obtain a more accurate river plastic flux model, it is necessary to couple the model with hydrodynamic simulations. When calculating for rivers that lack actual measured data, it is advised to add a relationship coefficient between the river discharges and MPs concentration in riverine MPs flux equations according to specific river.

4.4 *Riverine MPs flux*

There is no single method for assessing MP or total plastic flux in rivers, which reflects the different hydrogeomorphic settings of rivers globally, as well as the different data used to generate such estimates. As stated above, this relates to the difficulties associated with sampling rivers, especially those which are large, fast, or have a high suspended load. Different sampling methods are capable of capturing a different proportion of the total MPs load in a river, and the way that these data are interpreted will influence the accuracy of calculated flux estimates. The selection of sampling methods or geographical factors can lead to the differences in units or data expressions. For example, specific measurement of MPs and larger plastic flows with nets [66] and visual observation [37] reported either the numerical or mass flux. Conversely, some waste management infrastructures such as manual waste collection activities [93] and booms[132] were used in rivers, which are tend to report a total mass of plastics that is intercepted. Some estimates incorporate multiple measurements which have been conducted under different hydrological conditions or sampling over a longer time scale to consider the temporal variability in MPs and plastic flows [66, 78, 133, 134].

For MPs flux calculation, a common method is to build a model and combine it with field data. Based on this, Moore et al. [135] estimated that two rivers in Los Angeles, the U.S.A, can transfer 2 billion pieces of MPs into Californian coastal waters in three days. Zhao et al. [64] adopted the mean MP concentration from field data to calculate the annual plastic flux, and the Yangtze River was estimated to have transported 16-20 trillion MPs through the top layer of water (approximately 30 cm depth), a total weight of 537.6-905.9 tonnes, into the East China Sea annually. The river discharge from the Nakdong River, South Korea, was calculated by dividing the river into two vertical portions at a downstream site: surface (from the surface to 0.2 m) and subsurface (from 0.2 m to the bottom) [78]. The estimated total annual load in the Nakdong River reflecting the abundance of MPs in both surface and subsurface waters at the estuary across four seasons was 5.4 trillion particles, or 53.3 tonnes, in 2017. Mai et al. [66] used Manta trawls (330 μm) to sample the MPs in the surface water of the Pearl River Delta and calculated the riverine MP inputs by multiplying the concentrations of MPs and river discharge. The annual transport number of MPs in the Pearl River is 390 billion, which weighs 66 tonnes, and can be converted into an average plastic debris mass of 2900 tonnes year^{-1}. By comparing the MP concentrations in the surface waters of 22 global rivers, the MP concentration in the Pearl River was at the lower middle level. Max et al. [85] modelled MPs loads in rivers. The model had three input factors: the density of the population connected to sewage systems, per capita input of MPs, and sewage treatment efficiency. Approximately 14400 tonnes of MPs from point sources were calculated to enter the North Sea, Baltic Sea, Black Sea, Mediterranean Sea and European River basins and then flow into the Atlantic Ocean in 2000. In addition, these numbers differed by sea. The MP load amount to the Mediterranean Sea was 5600 tonnes, the load to the Black Sea was 4100 tonnes, the load to the European part of the Atlantic Ocean was 2700 tonnes, the load to the North Sea was 1100 tonnes, and the load to the Baltic Sea was 900 tonnes.

MP fluxes may be inaccurately estimated if the MPs data used are not comprehensive enough. Zhao et al.[64] has found an overestimation of more than 50% in the Yangtze River Estuary and East China Sea may occur if only use the data in July. Soeun et al. [78] reported the influence of small size MPs (< 300 μm), water layer transportation and seasonal variation to the estimation of riverine MPs load. Small sampling volume also may lead to the error estimation of riverine MPs flux which has been demonstrated in the study of Park et al.[99].

5. Perspective and remaining knowledge gaps

The non-uniform sampling locations and methods of riverine MPs may lead to the underestimation or overestimation of the riverine MPs flux calculation, which has been demonstrated in 5.4. Considering that the mesh size is negatively correlated with the number of filtered MPs, a unified approach to minimizing the disparities must be identified. As a widely used method for sampling MPs in surface water, the net sampling with mesh size varies from 100-300 μm has great data comparability (Figure 2). However, this only from the data comparability of the dimension of analysis, the selection of specific sampling methods should be targeted according to different rivers. To study the variation characteristics of riverine MPs and plastic debris loads, physical hydrological data along with monitored MP data are essential. Real-time data during sampling, including river flow, salinity, velocity, turbidity, sediment concentration and temperature, can be analysed with MP concentrations to research the correlations among data to gain further understanding.

A long term monitoring strategy of riverine MP should consider to establish the MP particle size distribution curves for the monitored river under different representative flow regimes. And the monitoring results are recommended to report a power law based distribution curve since plastic particles tend to break down over time to ever smaller pieces. Kooi et al.[136] also has suggested a universal equation for this purpose. A scientific monitoring strategy should also consider the seasonality which needs to set the sampling intervals according to the river flow regime. As indicated above, the river flow may greatly affect the trend of particle numbers in the river.

One can use the sampling equipment which are already available, however, monitoring results should be reported with necessary auxiliary information including the flow regime. The sampling methods described in detail in chapter 4. The monitoring results can then be comparable by extrapolating the monitoring results based on MP particle size distribution curve under the specific flow regime. The flow regime is primarily controlled by the climatic conditions and may also be subject to considerable modification by natural impoundments, dams, or water storage. Flow characteristics may also be changed by water uses, such as withdrawal for irrigation. The discharge of a river (e.g., in m^3/s) is the most important measurement that indicates the river's flow condition. When possible, people should report the hydrograph based on measurements of daily river discharges for the whole monitoring period, this is extremely important in determining the flow regimes of the microplastic sampling dates. A comprehensive monitoring strategy should cover both base flow regimes and high flow regimes.

MP abundance distribution in different particle size ranges can be derived based on the above steps, the riverine MPs flux can then be estimated by using the mass curves corresponding to different MP particle sizes [105], rather than using a single reported value which may greatly sacrifice the accuracy of estimation.

Further research is required to evaluate the effect of sampling methodology on observed MPs concentrations and compositions, as different morphologies may dominate within different size classes of MPs [137], and different sampling methodologies may be more effective at capturing different particle types [79, 138]. In order to make the monitoring results consistent and comparable, it is important to establish a monitoring strategy for riverine MPs flux considering spatial and temporal variations, one has also to acknowledge that the selection of sampling equipment in different locations depend on the availability and tradition, it is therefore not realistic to require people to use samplers of the same size. The following considerations are therefore recommended:

1) Determine how the estuarine processes affects the riverine MPs flux estimation, for example, the riverine MP flux in downstream sections may be unidirectional flows, but the influence mechanism of tidal current action is not clear enough;

2) The sampling methods have differences, it is important to calculate the proportion of the total MP load captured by each method, and how representative are different sampling campaigns in terms of the full flux across a given cross-section;
3) Normalize the riverine MP flux measurement in different section of the same catchment or different catchment for data comparability. Usually, MP sampler cannot cover the whole cross-section of a river, due to the velocity difference, the microplastic flux measured at different location along the cross section will be different. It is therefore suggested to develop a microplastic cross-sectional profile for the monitored location. For example, the profile can be developed by measurements at each quartile along the vertical and horizontal directions of the cross-section of a river.

Corresponding Author

Daoji Li – State Key Laboratory of Estuarine and Coastal Research, East China Normal University,500 Dongchuan Road, Shanghai 200241, China; orcid.org/0000-0002-3447-3485; E-mail: daojili@sklec.ecnu.edu.cn

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supporting Information Available

This information is available free of charge via the Internet at http://pubs.acs.org.

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