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# Superimposed microplastic pollution in a coastal metropolis

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ABSTRACT

The mitigation of microplastic pollution in the environment calls for a better understanding of the sources and transportation, especially from land sources to the open ocean. We conducted a large-scale investigation of microplastic pollution across the Greater Melbourne Area and the Western Port area, Australia, spanning gradients of land-use from un-developed catchments in conservation areas to more heavily-developed areas. Microplastics were detected in 94% of water samples and 96% of sediment samples, with abundances ranging from 0.06 to 2.5 items/L in water and 0.9 to 298.1 items/kg in sediment. The variation of microplastic abundance in sediments was closely related to that of the overlying waters. Fiber was the most abundant (89.1% and 68.6% of microplastics in water and sediment respectively), and polyester was the dominant polymer in water and sediment. The size of more than 40% of all total microplastics observed was less than 1 mm. Both light and dense polymers of different shapes were more abundant in sediments than those in water, indicating that there is microplastic accumulation in sediments. The abundance of microplastics was higher near coastal cities than at less densely-populated inland areas. A spatial analysis of the data suggests that the abundance of microplastics increases downstream in rivers and accumulates in estuaries and the lentic reaches of these rivers. Correlation and redundancy analysis were used to explore the associations between microplastic pollution and different land-use types. More microplastics and polymer types were found at areas with large amounts of commercial, industrial and transport activities. Microplastic abundances were also correlated with mean particle size. Microplastic hotspots within a coastal metropolis might be caused by a combination of natural accumulation via hydrological dynamics and contribution from increasing anthropogenic influences. Our results strongly suggest that coastal metropolis superimposed on increasing microplastic levels in waterbodies from inland areas to the estuaries and open oceans.



## 1. Introduction

The research into microplastics has increased exponentially since that term was initially proposed, as it has had widespread scientific and public media salience for over a decade (Sutherland et al., 2019; Thompson et al., 2004). Although there is yet no internationally agreed-upon definition for the cut-off size for microplastics, it is generally accepted to be those <5 mm (Frias and Nash, 2019; Law, 2017). There are indications that microplastics pose risks to organisms across the full spectrum of biological organization, from cellular to population level (Wright et al., 2013). Microplastics can also contaminate a wide range of species that are consumed by humans (Seltenrich, 2015; Wright and Kelly, 2017). What is clear is that more scientific field-based evidence is needed to steer current debates over the real risks of microplastic pollution (Burton, 2017; Kramm et al., 2018; Sedlak, 2017). The rates at which plastics are accumulating in urban and peri-urban waste streams have sparked research efforts to better to understand the major sources of microplastics in the environment and help improve management of this pollution.

For over half a century, impacts of plastic pollution to marine environments have been of great public concern (Carpenter and Smith, 1972). Large-scale efforts have been made to map microplastic distributions in global oceans, shorelines and marine

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organisms (Browne et al., 2011; Eriksen et al., 2014; Law et al., 2010). Marine microplastic dynamics can shed light upon various sources which are predominantly from land-based inputs (Jambeck et al., 2015; Lebreton et al., 2017). A world-scale modeling approach predicted that the amount of plastic waste entering oceans is estimated to be close to three orders of magnitude greater than the current monitoring results of floating marine plastic debris would suggest (Jambeck et al., 2015). Many sources of microplastic pollution in land-based watersheds have been identified; such as discharges of treated wastewater, industrial and commercial activities, municipal solid waste collection and agricultural activities. (Andrady, 2017; He et al., 2019; Law, 2017; McCormick et al., 2014). However, the relative contribution of these land-use activities to microplastic pollution is unknown.

The spatial distribution of microplastics in water and sediment can help predict their movement and deposition within local aquatic habitats. For example, vertical profiles of microplastics in waterbodies are greatly influenced by water flow and sediment dynamics (Nizzetto et al., 2016). For sediments, some case studies found extremely high concentrations of microplastics (74,800 items/kg) in freshwater sediment, which acted as a temporary or permanent sink (Wang et al., 2018). Particle sizes and densities were suggested as key factors governing with microplastic deposition process (Wagner et al., 2019). Nevertheless, the detailed mechanisms involved in microplastic transportation between sediment and water are not fully understood.

Reportedly high loads of microplastics in freshwaters has sparked interest in better understanding their sources. The spatial distribution of microplastics at a catchment scale has been described of some rivers, lakes and estuaries (Kapp and Yeatman, 2018; Schmidt et al., 2017; Siegfried et al., 2017). Plot and field studies involving direct monitoring suggest that microplastic pollution is closely related to anthropogenic factors such as population density, land-use and point source pollution (Barrows et al., 2018; Hendrickson et al., 2018; Klein et al., 2015). River systems were suggested as major transport pathways for plastic debris, especially in urban catchments (Atwood et al., 2019; Mani et al., 2015, 2019). The dominant role of natural forces like weather and hydrological conditions in microplastic transport dynamics has also been modeled (Hurley et al., 2018; Siegfried et al., 2017). However, it is not clear to what extent and how those factors ultimately determine the fate of microplastic transport from land to the ocean.

Snapshot sampling for microplastic distribution is still an economic way to help gauge microplastic transport pathways from source to sink, especially for places where baseline data are relatively insufficient. Although microplastics have been found on all continents, limited reports on their distribution exist for Australia's inland water bodies (Lebreton et al., 2017; Schmidt et al., 2017). Such a knowledge gap limits global estimations of microplastic emissions from land to ocean. A better understanding of microplastic sources is required to best address the elimination of the principal sources. Herein we selected waterbodies in the Port Phillip and Western Port catchments which includes the Greater Melbourne Area (GMA) and one of the most populated cities in Australia. We aim to determine: (i) the spatial distribution of microplastics from inland water bodies to estuaries at a catchment scale, (ii) the environmental factors affecting the variation of microplastic pollution, and (iii) the anthropogenic impacts on the distribution of microplastics in sediment and water.

## 2. Materials and methods

### 2.1. Survey area and sample collection

The GMA supports a population of approximately 4.3 million people, covering a watershed area >10,000 km$^2$ (Victoria, 2017). There are numerous streams, wetlands and estuaries in the GMA that have a variety of land-use activities in their catchments (Sharley et al., 2016). The urban sprawl of Melbourne stretches around a large area of Port Phillip Bay and a small area of Western Port Bay. The Yarra, Maribyrnong and Werribee Rivers and Dandenong Creek are the major streams in the Port Phillip Bay catchment (Fig. 1). These rivers and many other smaller streams in this catchment support a diverse community of birds, fishes and benthic invertebrates (Mehler et al., 2018). Being the largest marine embayment in Victoria, Port Phillip Bay is surrounded by the extensive coastal communities including the GMA and the city of Geelong. In comparison, Western Port has a more rural catchment, although the GMA extends into the north-western and western areas of the catchment, and Phillip Island, located at the mouth of Western Port, is also largely urbanized. Sediments and surface waters were collected from 54 monitoring sites in the study area between April and May 2018 (Fig. 1). The survey sites were located in estuaries (n = 15), streams (n = 24) and wetlands (n = 15) (for details see Supplementary Material Table 1).

Prior to use, all sampling tools were rinsed with ethanol and *in situ* water. Water samples were collected first, then sediment to avoid collecting suspended solids due to sediment disturbance during sampling. A 5-L container was submerged and filled with surface water (0−5 cm in depth) at each site. In comparison to net and trawl sampling, this bottle sampling approach can collect microplastics from a small volume of water (Kapp and Yeatman, 2018). Sediments were collected close to the shoreline from a maximum water depth of approximately 1 m. The top layer (top 2 cm) was withdrawn using a handheld shovel from three separate and undisturbed surfaces no closer than 5 m apart. Approximately 500 g of the sediments were collected from each shovel and transferred into an unopened glass jar, which was promptly closed once full.

### 2.2. Determination of catchments and land use level

Topographic vector data, including elevation as contour lines, major catchment polygons, and waterway and water body polygon layers were supplied by Melbourne Water and the land-use data was acquired from the Australian Bureau of Statistics (ABS, 2016). Sub-catchments for all sites were determined topographically using spatial analyst in ArcGIS 10.3 based on detailed methods outlined in Kunapo et al. (2009). Briefly, 10 m flow-weighted digital elevation model (DEM)s were derived from 5 m contour layers with flow direction ingrained into the DEM by stream layers and wetlands. Sink-holes were removed, then flow direction and accumulation drainage layers were created and catchments were derived using the hydrology extension toolbar. Where existing high-resolution stormwater drainage layers were available from local government jurisdictions, these were compared against watershed layers with some minor adjustments made ensure alignment with stormwater drains.

Catchment land-use was extracted from census mesh block counts from the 2016 Australian census (ABS, 2016). Land-use polygon categories were summed for each catchment, then divided by the area in hectares in grouped land-uses (Residential, Parkland, Education, Commercial, Industrial, Hospital/Medical, Transport, Primary Production, Other and Water) for subsequent analysis (for details see Supplementary Material Table 2).

### 2.3. Isolation of microplastics

Extraction of microplastics in water and sediment followed our established two-step filtration process (Su et al., 2016, 2018). For

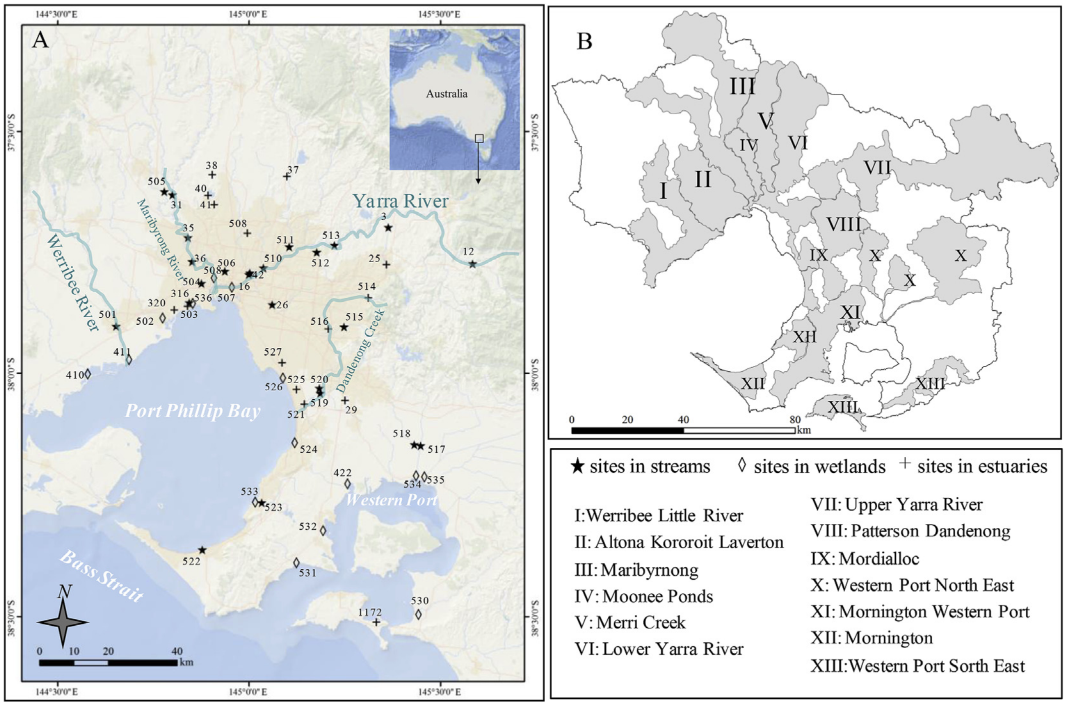


**Fig. 1.** Locations of sampling sites (A) and catchments (B) within the watersheds of Port Phillip and Western Port Bays.

water samples, volume was recorded and all material >20 μm in the water was collected on a filter (Millipore Nylon NY2004700). Any substances on the filter were washed into a glass jar using 100 mL of NaOH solution (2M) to digest the biological material (Cole et al., 2014). The jar was then covered and placed into a water bath at 55 °C for 48 h. Once digested, the liquid in these jars were filtered and the filter papers were then covered and placed in Petri dishes for microplastic inspection. Before extraction, sediments were placed in glass jars and dried in an oven at 55 °C for 4 d to reduce the water content. Approximately 80 g of dry sediment was weighed and mixed with saturated NaCl solution (1.2 g/mL) at a ratio of 1:2 (V/V) in a 600-mL glass beaker. The mixture was stirred with a steel spoon and allowed to settle for 24 h. The supernatant was collected and filtered, and the process was repeated twice. All substances on filters were transported into glass jars and digested using sodium hydrate solution (2M) following the same method as the water samples.

### 2.4. Observation and validation of microplastics

A visual inspection was first carried out to quantify and sort the suspected microplastics based on their properties. All items suspected to be microplastics (<5 mm) on the filters were inspected and photographed using a microscope (Leica M125) with 25-100x magnification. Microplastics were catalogued as either being fragment (small irregular pieces), pellet (spherical items), film (thin and small layers), or fiber (elongated) (Yang et al., 2015). The sizes of microplastics was also measured and recorded. Then these items were examined using micro-Fourier Transform Infrared Spectroscopy (Nicolet iN10 MX, Thermo-Fisher Scientific). Data were collected from a resolution of 4 $cm^{-1}$ with a 32-s scan time and spectra were compared with a database from Thermo-Fisher to verify the polymers. The spectra matching with a quality index >70 were accepted (Yang et al., 2015). We tried to analyze all suspected items, but some particles could not be identified due to poor quality spectra, small size or being lost and damaged. Finally, an average percentage of 42.4% and 57.9% of suspected items from water and sediment, respectively, were successfully verified for the sites, which agreed with the recommended quality criteria (Hermsen et al., 2018). Non-plastic items were removed from the microplastic counts. Given that a set of visually identified microplastics was not instrumentally confirmed, our results may potentially be overestimated.

### 2.5. Quality control

In order to reduce the risk of microplastic aerial deposition in the field, containers were only open once samples were ready to be deposited into them and closed immediately afterwards. Cotton clothing was worn during field sampling and a cotton laboratory coat was worn during the analytical procedures and preparation of all liquid solutions including tap water, sodium hydrate solution (2M) and sodium chloride solution (1.2 g/ml) were filtered prior to use (filter pore size = 20 μm). Before use, all containers and apparatus were washed thoroughly by filtered tap water. To account for process contaminations in experiments, procedural blanks (consisting of distilled water) were run without field water or sediment with every three replicates in the laboratory. Overall, 108 blanks were performed for water and sediment. Procedural contamination ranged from 0.22 to 0.38 items per filter for water and sediment samples. These procedural blanks were at a similar level to our previous work (Hu et al., 2018). All the contamination in blank samples was fiber, with mean concentrations of 0.078 items/L for water and 3.7 items/kg for sediment. We subtracted the background from our final results.

### 2.6. Data analysis

Non-parametric tests were applied because the data sets are not normally distributed. Spearman's rank correlation was used to describe the relationships among data sets and the Kruskal-Wallis test was used to compare among groups. These non-parametric statistical analyses were performed using SPSS 22. Redundancy analysis is a useful tool for the ordination of multiple variance and has been widely used in ecology and environmental sciences (Legendre and Anderson, 1999; McArdle and Anderson, 2001). It was used to explore the correlations between microplastic variance and land-use influences. The microplastic properties (abundance, size, shape etc.) were considered as "response variables", while land-use and elevation were considered as "explanatory variables". A linear ordination method was selected according to detrended correspondence analysis (length of the gradient = 1.32). All the values were standardized by Z-scoring before carrying out the analysis. Redundancy analysis was performed and plotted with R 3.22.

## 3. Results

### 3.1. Abundance and spatial distribution of microplastics

Microplastics were detected in 94% of water samples and 96% sediment samples from the entire sampling area, ranging from 0.06 to 2.5 items/L in water and 0.9 to 298.1 items/kg in sediments (Fig. 2 A and C). The mean abundance of microplastics varied from 0.03 to 1.7 items/L in water and 4.5 to 172.7 items/kg in sediments (Fig. 2 A and C). Wetlands, estuaries and streams respectively contained average microplastic concentrations of 0.8, 0.9 and 1.0 items/L from water samples. Sediment samples from the same water bodies contained average microplastic concentrations of 75.5, 79.1 and 87.4 items/kg, respectively. No significant difference was found between the three water body types (Supplementary Material Fig. S1). Microplastic abundance variation in sediment and overlying water was closely related ($p < 0.05$). The abundance of microplastics was higher near coastal cities than at less densely-populated inland areas (Fig. 2 B and D).

Microplastic spatial distribution was highly variable within sampling areas and its concentrations in water and sediment have large coefficients of variation (CV) up to 82.3% and 88.4%. The range of microplastic abundance level spanned one to two orders of magnitudes. In contrast, microplastic abundance exhibited minor variation between different waterway types (CV = 7.6%−11.1%). The elevation of sampling sites ranged from 1 to 213 m above sea level and was negatively correlated with microplastic abundance ($p < 0.05$) (Fig. 3).

### 3.2. Morphology characteristics and chemical composition

Microplastic size density was similar between samples from water and sediments (Fig. 4A). The mean size of microplastics in water (1.26 ± 0.93 mm) was also close to those found in sediments (1.24 ± 0.84 mm). There was no significant difference in size distribution of microplastics in water or sediment between different waterway types (Supplementary materials Fig. S2). Items <1 mm contributed more than 40% of total microplastic counts in water and sediment (Supplementary materials Fig. S2). Fibers were the most common shape accounting for 89.1% and 68.6% in water and sediment, respectively, while the percentage of fragments in sediment was three times higher than those in water (Fig. 4B; Supplementary materials Fig. S3). In addition, microplastic shape varied significantly between the three waterway types, where wetland samples contained the highest amount of fibers in sediment (Supplementary materials Fig. S4).

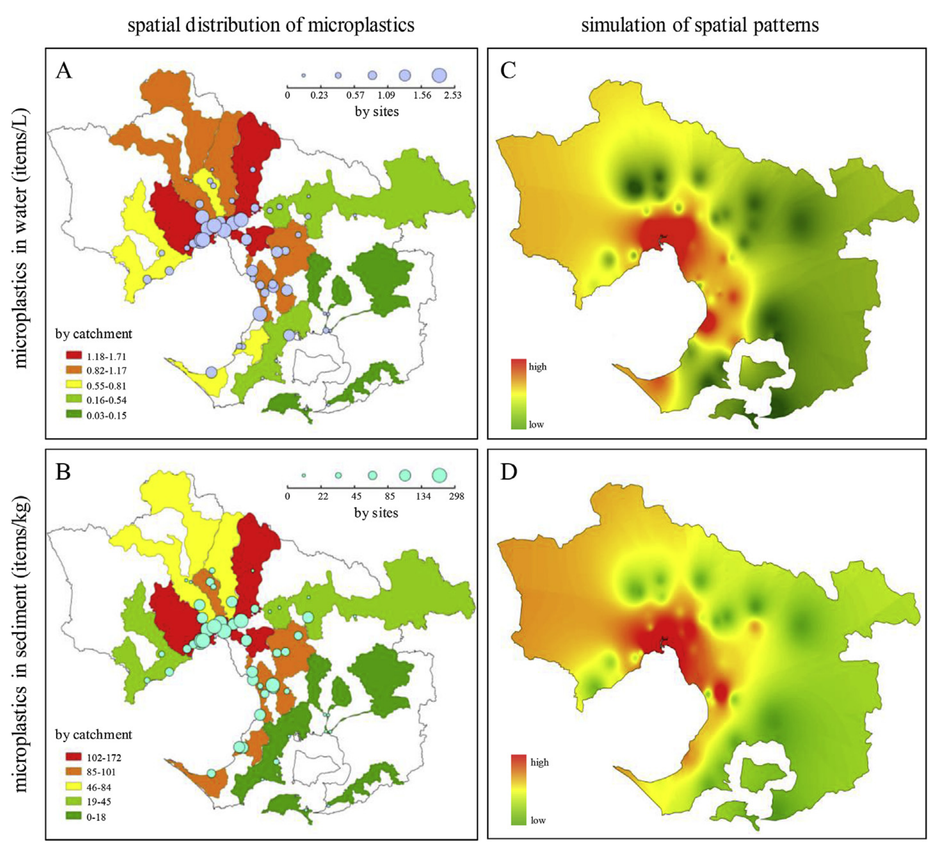


**Fig. 2.** Spatial distribution of microplastics in sampling areas in terms of sites and catchments (A–B) and the simulation of spatial patterns based on inverse distance weighted interpolation (C–D). The data intervals in A and C were optimized via Jenks natural breaks classification method.

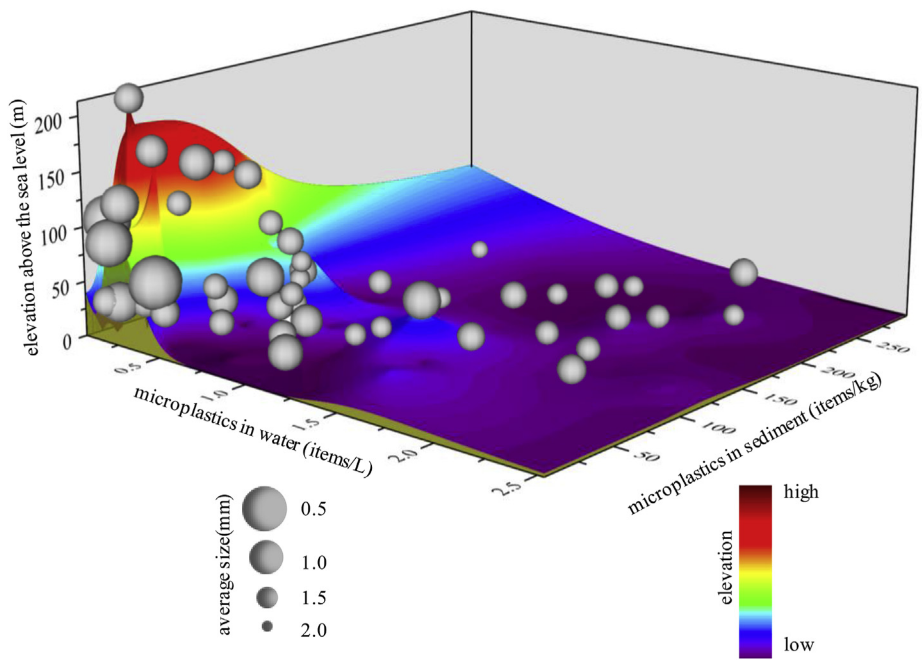


**Fig. 3.** Microplastic abundance increased downstream; Kriging process was applied to generate and smooth surfaces and the size of ball represented microplastic mean size variation (The color depth represented elevation from low to high). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

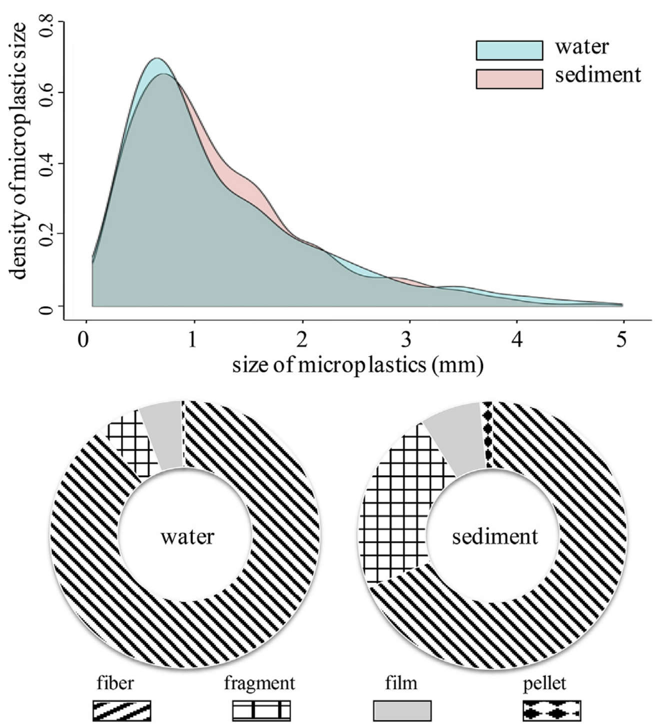


**Fig. 4.** Kernel density estimation of microplastic size (A) and the shape distribution (B) in water and sediment.

In all items verified (n = 828), synthetic polymers represented 72.4% and 69.8% of items in water and sediment, respectively (Supplementary materials Tables S3 and 4). There were 14 kinds of polymer types in water and 19 types in sediment (Fig. 5). Polyester was the most common type; being present in more than 60% of sampling sites and comprising more than 30% of all synthetic polymers confirmed in both water and sediment (Fig. 5A and B). In addition, polypropylene, polyethylene and polyamide were also commonly found and together comprise >30% abundance of all polymer types found. Although most polymer distributions were similar between water and sediment, polyvinyl chloride was eighteen times more abundant with a frequency of detection four times higher in sediment than in water.

### 3.3. Land-use in relation to microplastic characteristics

Land-uses were categorized as 10 individual types associated with the catchment for each sampling site (Supplementary materials Table S2). Residential (32.5%) land-use was the most common category found in all sampling catchment followed by primary production (32.0%) and parkland (17.4%). Urban-related categories such as transport, industrial and commercial land-uses are positively correlated ($p < 0.05$) (Fig. 6 red text). Regarding characteristics (Fig. 6 purple text); abundance, size and polymer diversity were also correlated with transport, industrial and commercial land-use categories ($p < 0.05$).

Redundancy analysis was used to further understand the role of land-use in microplastic spatial patterns and characteristics. Land-use was selected as an environmental variable to explain the variation of microplastic abundance, shape distribution, mean size and polymer diversity (Fig. 7A). Thirty six percent of the variance of microplastic features can be explained by land-use, the first and second axis represented 91.1% together. Commercial, industrial and transport land-uses were significantly associated with an increase in the microplastic abundance and polymer diversity a decrease in the mean particle size. The sites which explained the bulk of this variation were located within urbanized areas of Port Phillip bay (red spots in Fig. 7B). Interestingly, although fiber accounted for more than 70% of microplastics in water and sediments, they only correlated with domestic influences such as parkland, residential and education land-uses.

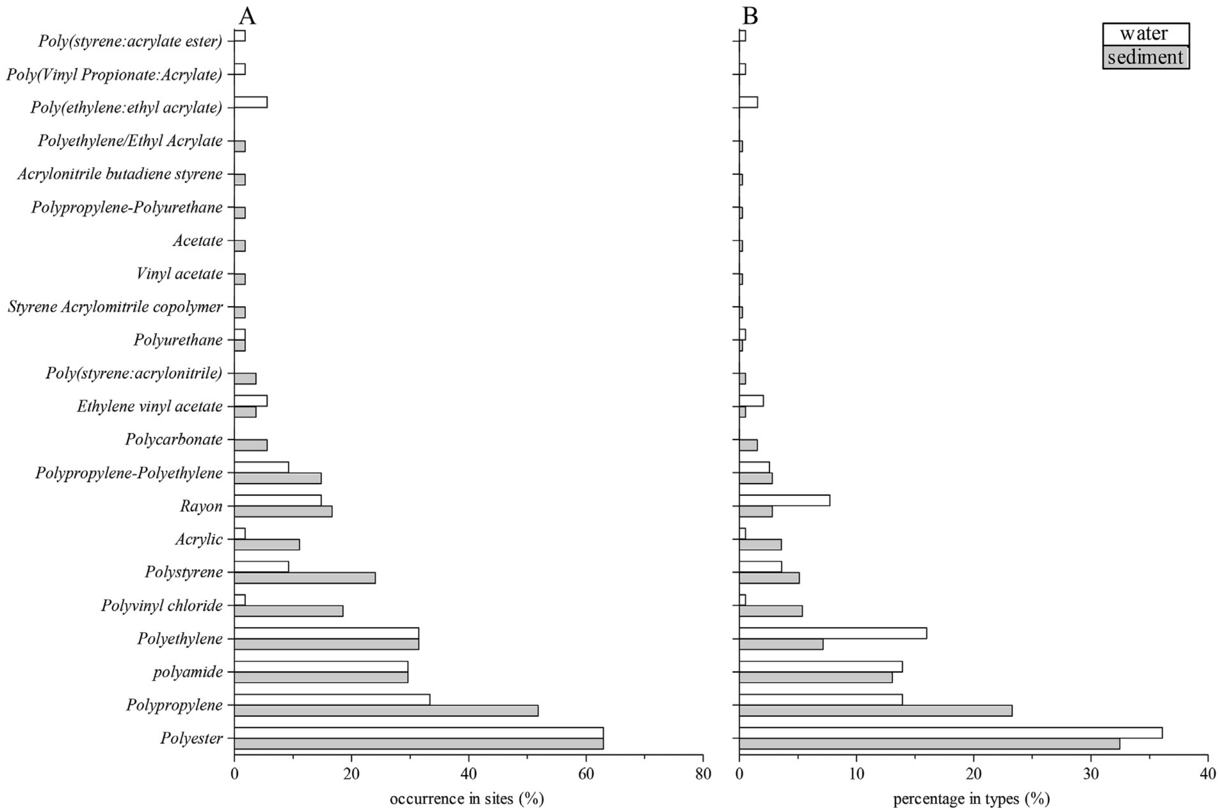


**Fig. 5.** The occurrence rate (%) of different polymer in sampling sites (A) and the composition of polymer in water and sediment (B).

## 4. Discussion

### 4.1. Microplastic pollution in GMA and Western Port catchments

Concerns are rising about the ubiquitous presence of microplastics in the environment. This is the first time data have been reported from Australian inland waters and estuaries. This work enriches the world microplastic pollution map and provide spatial features spanning gradients of land-use from un-developed catchments in conservation areas, to more urbanized areas.

Our studies of microplastic concentrations in the GMA and Western Port catchments show that the abundance of microplastic pollution in water and sediment from these areas is low by global comparison; being below 50% and 70% respectively of the mean abundances reported in 55 independent studies measured in terms of microplastic items per liter or kilogram (Supplementary Material Fig. S5). From the compiled results, different methods involved in microplastic sampling and measurement complicate efforts to make rigorous global comparisons (Hidalgo-Ruz et al., 2012). However, the ease to make such comparisons is gradually improving as more studies approach a uniform methodology.

It is hard to conclude general trends of microplastic shape or size in different studies because the field results are largely determined by regional background. Pellets can be extremely abundant in specific industrial areas (Lechner and Ramler, 2015), while they may be absent from remote waterbodies (Castaneda et al., 2014; Free et al., 2014). Regarding microplastic size, the concentrations of small particles may be positively correlated with overall abundances but sometimes samples with high abundance can predominantly contain the larger particles (>1 mm) (Zhang et al., 2017). In our work, there are no spatial patterns of microplastics' morphological characteristics except for decreasing sizes, suggesting that indicated that diffuse plastic pollution is more predominant in our study area than point source plastic pollution.

Some studies have compared different variations in abundances in different waterway types, suggesting lentic waterbodies could be a sink for microplastics due to low flow rates and wave energy (Hu et al., 2018; Wang et al., 2018; Zhang et al., 2017). However, we found no difference in the amount and type of microplastic pollution between streams, wetlands and estuaries (Supplementary Material Fig. S5). Microplastic monitoring for environmental management should ensure site selection does not create bias towards particular water body types or spatial features. An unbiased approach will improve the representativeness of natural habitat and the strength of any observations and conclusions.

### 4.2. Environmental factors affecting spatial patterns of microplastic

Buoyant microplastics will accumulate in surface waters but eventually deposit in sediments due to bio-fouling and other processes reducing their buoyancy (Galloway et al., 2017). Before settling down, the dispersion of microplastics in waterbodies is inevitably impacted by natural forces such as current, wave and weather conditions (Browne et al., 2010; Law et al., 2010). In our case, the most obvious spatial trend was that microplastic concentrations increased downstream (Fig. 5). Current and water flow play a major role in the inventory and transport of microplastics downstream as microplastics behave similarly to particles that are neutrally buoyant in water, such as plankton and suspended solids (Di Mauro et al., 2017; Nizzetto et al., 2016). Extreme hydrological changes from weather events such as floods, can strongly alter the microplastic budget in local habitats (Hurley et al., 2018). Estuaries and river mouths receive significant proportions of materials, including microplastics, sourced from inland parts of catchments

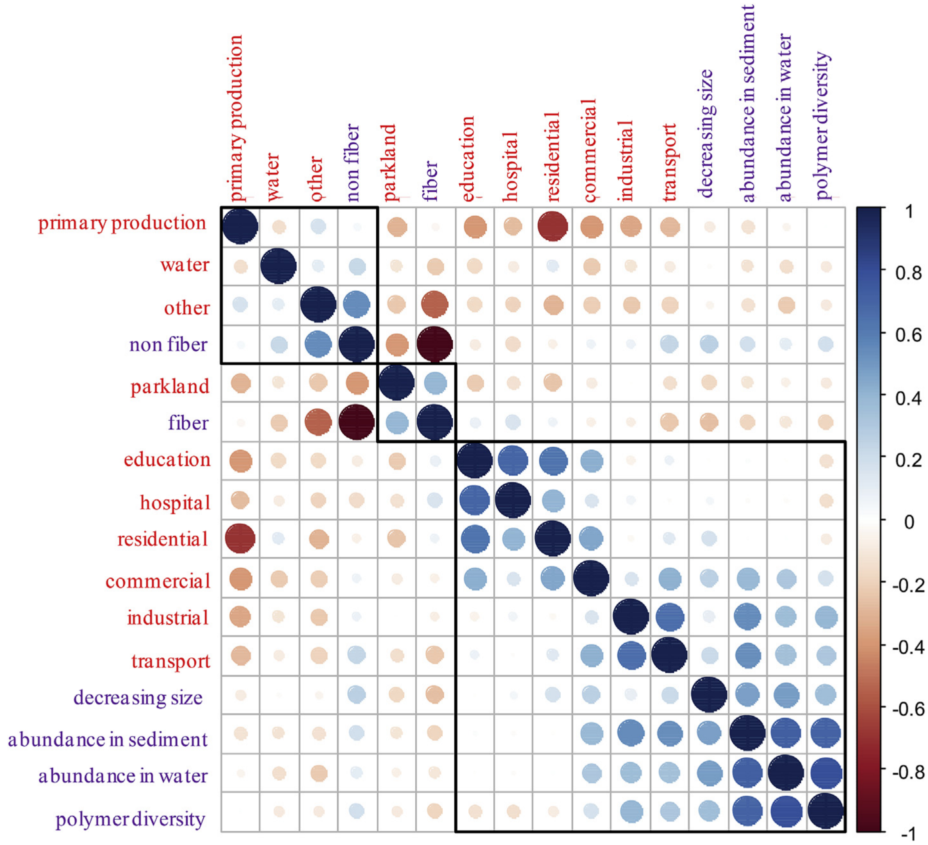


**Fig. 6.** Spearman's rank correlation coefficient between land use data and microplastic characters. The bold frames grouped similar index was produced by analytic hierarchy process.

and may therefore be considered as "natural sinks" of microplastic pollution (Leslie et al., 2017; Luo et al., 2019).

Microplastics are prone to sedimentation, as evidenced by differences in polymer types and shapes shown between sediments and water (Figs. 4 and 5). The differences in microplastic concentrations between sediment and water spanned two orders of magnitudes in our study and can reach over six orders of magnitude in a study on lake (Fischer et al., 2016). It was noteworthy that microplastic size decreased with increasing microplastic concentrations in water and sediments (Fig. 3). According to Stokes' law, small neutral or negatively-buoyant particles will take more time to settle than larger ones (Richardson and Zaki, 1954; Zwanzig, 1964). If ideal conditions for this law are met, large amounts of small microplastics may be transported far away with current and accumulate downstream into the estuaries and oceans. On the other hand, small microplastics may be directly generated by the fragmentation of large plastic debris and used as indicators for large plastic waste. Such particles likely account for the "missing microplastic fraction" in open oceans once deposited in freshwater sediments (Wang et al., 2018). More efforts should be made to track smaller microplastic particles from freshwaters to oceans in future monitoring schemes.

### *4.3. Anthropogenic influences on the spatial patterns of microplastics*

Being anthropogenic in origin, microplastic footprints in environments are largely determined by anthropogenic influences. For example, effluents without appropriate treatments from wastewater treatment plants significantly increased microplastics abundances in water and sediment downstream (Estahbanati and Fahrenfeld, 2016; McCormick et al., 2016; Vermaire et al., 2017). Plastic industrial activities directly contributed to the microplastic load downstream and in proximal water bodies (Castaneda et al., 2014; Mani et al., 2015; Wagner et al., 2019). Recreational activity along urban rivers contributes a considerable amount of plastic litter which is a direct source of microplastic (Kiessling et al., 2019). Microplastic shape characteristics, size distribution and polymer composition have been suggested as associative links for source identification (Auta et al., 2017). The presence of fibers and pellets are considered as indicators of domestic effluents and plastic industry activities, respectively (Lechner and Ramler, 2015; Mintenig et al., 2017). The prevalence of microfibers in our water samples suggested that runoff from urban areas is a major source of microfibers.

Land-use analysis provides more specific information regarding source identification and has been commonly used as a predictor variable of microplastic pollution (Peters and Bratton, 2016; Yonkos et al., 2014). In the current study, microplastic pollution in water and sediment was closely related with the transport, industrial and commercial land-use. Those influences were even stronger than the residential activities (Fig. 6), although regional population size has often been proposed as an indicator of microplastic emission level (Eriksen et al., 2013; Yonkos et al., 2014).

Efforts have been made to prioritize major pollution sources and trace possible pathways. Some studies of urban catchments have demonstrated the role of rivers in microplastic transport from land sources to estuaries and open oceans, which include the influences from river water discharge, sewer and catchment urbanization (Barrows et al., 2018; Eo et al., 2019; Wagner et al., 2019). It

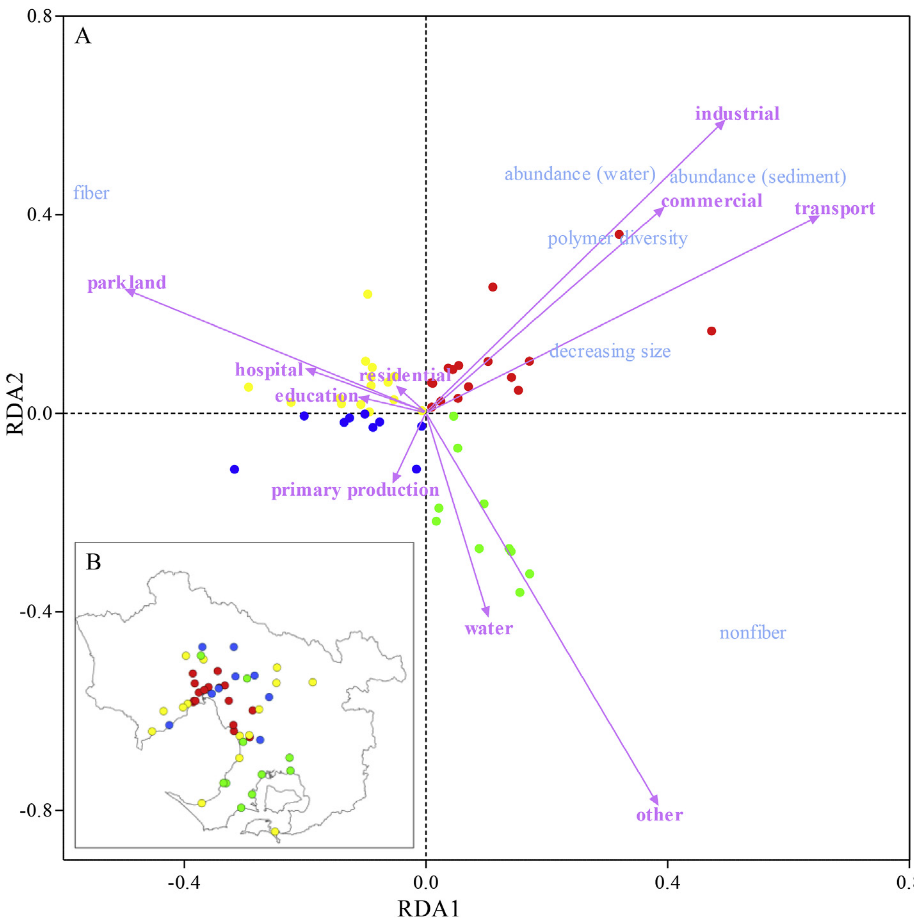


**Fig. 7.** A redundancy analysis of variation in microplastic features (blue characters) and land use influences (purple characters with arrows) in monitoring sites (scatter spots); B: Spatial distribution of monitoring sites by quadrant according to redundancy analysis. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

remained difficult to pinpoint the particular sources of microplastic pollution in estuaries and large lakes given numerous, often interacting, factors acting on transportation, retention and accumulation.

### 4.4. Superimposed microplastic pollution in a coastal metropolis

Of all the plastics ever created only a small faction has been recycled. Managing plastic pollution sources might be the only tangible solution to reduce microplastic emissions (Jambeck et al., 2015). It is important to identify and address microplastic pollution hotspots. Our study suggests that coastal population centers with intensive urban land-uses are expected to contain higher levels of microplastic pollution, especially at regions located downstream of major waterways. Several suspected local hotspots of microplastic pollution exist, such as Shanghai and Saigon in Asian (Lahens et al., 2018; Luo et al., 2019), Amsterdam and the Mediterranean region in Europe (Leslie et al., 2017; Vianello et al., 2013), Chicago metropolitan area in North America (McCormick et al., 2014), South Africa's coastline in Africa (de Villiers, 2019). Estuaries and river deltas can provide buffer zones for microplastics received upstream and adjacent urban areas (Atwood et al., 2019; Simon-Sánchez et al., 2019). Therefore, microplastic hotspots within those areas might be caused by the combination between natural accumulation via hydrological dynamics and contribution from increasing anthropogenic influences.

In the GMA and Western Port area, high abundances of microplastics in the surface waters around cities are related to either great urbanization or treated wastewater discharges. While Australian urban cities have separate storm water and sewerage systems, sewage is not common in our urban waterways except where there treated wastewater is discharged into streams. In contrast, microplastics were most likely carried by surface runoff and associated with urban non-point pollution sources. For example, traffic emission and road pollution account for a large portion of land-based microplastic transport to estuaries (Unice et al., 2019). One preliminary study recovered up to 160,000 microplastics per kg soil from roadside dust (Dehghani et al., 2017), which is almost three order of magnitude higher than what we found in GMA's sediments. Urban soils had been also suggested as suspected sinks for microplastics but more field-based evidence is needed (Rillig, 2012). Unfortunately, current technologies cannot provide a full spectrum of microplastic presence in ecosystems due to the challenges in verifying small size microplastics. Little is known about those plastic particles that are consumed or destroyed by organisms (Dawson et al., 2018; Gigault et al., 2018). Regardless if non-point source pollution is the main source of microplastic in aquatic environment or not, the pathways involved in microplastic transportation in coastal cities deserve a better understanding. Fifty percent of the global population is expected to live within 100 km of coastline by 2030 (Adger et al., 2005). The development of coastal urban communities is driven by human actions that directly increase the local environmental stress (Adger, 1999). As shown in our work, Melbourne waterways received and accumulated a lot of microplastics from upstream and the city itself was also a major source of this pollution. Recent large-scale efforts have been undertaken on global shorelines and river-nets to estimate the mass of microplastics entering the ocean (Eo et al., 2019; van Wijnen et al., 2019). However, before we can fill the knowledge gap between sources and sinks, we may need to step back further and take a look at our own coastal cities.

## 5. Conclusion

We have shown spatial pollution patterns of microplastics in water and sediment in the range of Greater Melbourne Area and Western Port area. The catchments are an important source of microplastics entering the oceans. Urbanized water bodies in this study were more polluted with microplastics compared to other areas. When comparing the results with similar international studies, the level of microplastic pollution in these urbanized areas is regarded as low. The spatial analysis associated with catchment land-use levels suggests that microplastic pollution was "strengthened" by both hydrological dynamics and anthropogenic influences. A coastal metropolis first receives microplastic inputs from upstream areas, then magnifies its abundance and transportation via urban activities, although the pollution sources are still poorly understood. A long-term and large-scale monitoring of microplastics in coastal metropolitan areas should be considered in further global schemes, which are important in understanding microplastics fate during the journey from land to sea.

## 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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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.watres.2019.115140.

## References

ABS, 2016. (Australian Bureau of Statistics) census of population and housing: mesh block counts 2074.0. Available online at:http://www.abs.gov.au/AUSSTATS/abs@.nsf/DetailsPage/2074.02011?OpenDocument. . (Accessed 26 July 2016).
Adger, W.N., 1999. Social vulnerability to climate change and extremes in coastal Vietnam. World Dev. 27, 249–269.
Adger, W.N., Hughes, T.P., Folke, C., Carpenter, S.R., Rockström, J., 2005. Social-ecological resilience to coastal disasters. Science 309, 1036–1039.
Andrady, A.L., 2017. The plastic in microplastics: a review. Mar. Pollut. Bull. 119, 12–22.
Atwood, E.C., Falcieri, F.M., Piehl, S., Bochow, M., Matthies, M., Franke, J., Carniel, S., Sclavo, M., Laforsch, C., Siegert, F., 2019. Coastal accumulation of microplastic particles emitted from the Po River, Northern Italy: comparing remote sensing and hydrodynamic modelling with in situ sample collections. Mar. Pollut. Bull. 138, 561–574.
Auta, H.S., Emenike, C.U., Fauziah, S.H., 2017. Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ. Int. 102, 165–176.
Barrows, A.P.W., Christiansen, K.S., Bode, E.T., Hoellein, T.J., 2018. A watershed-scale, citizen science approach to quantifying microplastic concentration in a mixed land-use river. Water Res. 147, 382–392.
Browne, M.A., Crump, P., Niven, S.J., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines woldwide: sources and sinks. Environ. Sci. Technol. 45, 9175–9179.
Browne, M.A., Galloway, T.S., Thompson, R.C., 2010. Spatial patterns of plastic debris along estuarine shorelines. Environ. Sci. Technol. 44, 3404–3409.
Burton, G.A., 2017. Stressor exposures determine risk: so, why do fellow scientists continue to focus on superficial microplastics risk? Environ. Sci. Technol.
Carpenter, E.J., Smith Jr., K.L., 1972. Plastics on the Sargasso sea surface. Science 175, 1240–1241.
Castaneda, R.A., Avlijas, S., Simard, M.A., Ricciardi, A., 2014. Microplastic pollution in St. Lawrence river sediments. Can. J. Fish. Aquat. Sci. 71, 1767–1771.
Cole, M., Webb, H., Lindeque, P.K., Fileman, E.S., Halsband, C., Galloway, T.S., 2014. Isolation of microplastics in biota-rich seawater samples and marine organisms. Sci. Rep. 4, 2231–2236.
Dawson, A.L., Kawaguchi, S., King, C.K., Townsend, K.A., King, R., Huston, W.M., Nash, S.M.B., 2018. Turning microplastics into nanoplastics through digestive fragmentation by Antarctic krill. Nat. Commun. 9, 1001.
de Villiers, S., 2019. Microfibre pollution hotspots in river sediments adjacent to South Africa's coastline. Water S.A. 45, 97–102.
Dehghani, S., Moore, F., Akhbarizadeh, R., 2017. Microplastic pollution in deposited urban dust, Tehran metropolis, Iran. Environ. Sci. Pollut. Res. 24, 20360–20371.
Di Mauro, R., Kupchik, M.J., Benfield, M.C., 2017. Abundant plankton-sized microplastic particles in shelf waters of the northern Gulf of Mexico. Environ. Pollut. 230, 798–809.
Eo, S., Hong, S.H., Song, Y.K., Han, G.M., Shim, W.J., 2019. Spatiotemporal distribution and annual load of microplastics in the Nakdong River, South Korea. Water Res. 160, 228–237.
Eriksen, M., Lebreton, L.C., Carson, H.S., Thiel, M., Moore, C.J., Borerro, J.C., Galgani, F., Ryan, P.G., Reisser, J., 2014. Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea. PLoS One 9, e111913.
Eriksen, M., Mason, S., Wilson, S., Box, C., Zellers, A., Edwards, W., Farley, H., Amato, S., 2013. Microplastic pollution in the surface waters of the Laurentian great lakes. Mar. Pollut. Bull. 77, 177–182.
Estahbanati, S., Fahrenfeld, N.L., 2016. Influence of wastewater treatment plant discharges on microplastic concentrations in surface water. Chemosphere 162, 277–284.
Fischer, E.K., Paglialonga, L., Czech, E., Tamminga, M., 2016. Microplastic pollution in lakes and lake shoreline sediments – a case study on Lake Bolsena and Lake Chiusi (central Italy). Environ. Pollut. 213, 648–657.
Free, C.M., Jensen, O.P., Mason, S.A., Eriksen, M., Williamson, N.J., Boldgiv, B., 2014. High-levels of microplastic pollution in a large, remote, mountain lake. Mar. Pollut. Bull. 85, 156–163.
Frias, J.P.G.L., Nash, R., 2019. Microplastics: finding a consensus on the definition. Mar. Pollut. Bull. 138, 145–147.
Galloway, T.S., Cole, M., Lewis, C., 2017. Interactions of microplastic debris throughout the marine ecosystem. Nat. Ecol. Evol. 1, 1–8.
Gigault, J., Ter Halle, A., Baudrimont, M., Pascal, P.-Y., Gauffre, F., Phi, T.-L., El Hadri, H., Grassl, B., Reynaud, S., 2018. Current opinion: what is a nanoplastic? Environ. Pollut. 235, 1030–1034.
He, P., Chen, L., Shao, L., Zhang, H., Lü, F., 2019. Municipal solid waste (MSW) landfill: a source of microplastics? -Evidence of microplastics in landfill leachate. Water Res. 159, 38–45.
Hendrickson, E., Minor, E.C., Schreiner, K., 2018. Microplastic abundance and composition in western Lake Superior as determined via microscopy, Pyr-GC/MS, and FTIR. Environ. Sci. Technol. 52, 1787–1796.
Hermsen, E., Mintenig, S.M., Besseling, E., Koelmans, A.A., 2018. Quality criteria for the analysis of microplastic in biota samples: a critical review. Environ. Sci. Technol. 52, 10230–10240.
Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel, M., 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ. Sci. Technol. 46, 3060–3075.
Hu, L., Chernick, M., Hinton, D.E., Shi, H., 2018. Microplastics in small waterbodies and tadpoles from Yangtze river delta, China. Environ. Sci. Technol. 52, 8885–8893.
Hurley, R., Woodward, J., Rothwell, J.J., 2018. Microplastic contamination of river beds significantly reduced by catchment-wide flooding. Nat. Geosci. 11, 251.
Jambeck, J.R., Geyer, R., Wilcox, C., Siegler, T.R., Perryman, M., Andrady, A., Narayan, R., Law, K.L., 2015. Plastic waste inputs from land into the ocean. Science 347, 768–771.
Kapp, K.J., Yeatman, E., 2018. Microplastic hotspots in the snake and lower Columbia rivers: a journey from the greater yellowstone ecosystem to the Pacific ocean. Environ. Pollut. 241, 1082–1090.
Kiessling, T., Knickmeier, K., Kruse, K., Brennecke, D., Nauendorf, A., Thiel, M., 2019. Plastic Pirates sample litter at rivers in Germany – Riverside litter and litter sources estimated by schoolchildren. Environ. Pollut. 245, 545–557.
Klein, S., Worch, E., Knepper, T.P., 2015. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main Area in Germany. Environ. Sci. Technol. 49.
Kramm, J., Völker, C., Wagner, M., 2018. Superficial or substantial: why care about microplastics in the anthropocene? Environ. Sci. Technol. 52, 3336–3337.
Kunapo, J., Chandra, S., Peterson, J., 2009. Drainage network modelling for water-sensitive urban design. Trans. GIS 13, 167–178.
Lahens, L., Strady, E., Kieu-Le, T.-C., Dris, R., Boukerma, K., Rinnert, E., Gasperi, J., Tassin, B., 2018. Macroplastic and microplastic contamination assessment of a tropical river (Saigon River, Vietnam) transversed by a developing megacity.

Environ. Pollut. 236, 661–671.

Law, K.L., 2017. Plastics in the marine environment. Annu. Rev. Mar. Sci. 9, 205–229.

Law, K.L., Morét-Ferguson, S., Maximenko, N.A., Proskurowski, G., Peacock, E.E., Hafner, J., Reddy, C.M., 2010. Plastic accumulation in the North Atlantic subtropical gyre. Science 329, 1185–1188.

Lebreton, L.C., Van der Zwet, J., Damsteeg, J.-W., Slat, B., Andrady, A., Reisser, J., 2017. River plastic emissions to the world's oceans. Nat. Commun. 8, 15611.

Lechner, A., Ramler, D., 2015. The discharge of certain amounts of industrial microplastic from a production plant into the River Danube is permitted by the Austrian legislation. Environ. Pollut. 200, 159–160.

Legendre, P., Anderson, M.J., 1999. Distance-based redundancy analysis: testing multispecies responses in multifactorial ecological experiments. Ecol. Monogr. 69, 1–24.

Leslie, H.A., Brandsma, S.H., van Velzen, M.J.M., Vethaak, A.D., 2017. Microplastics en route: field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. Environ. Int. 101, 133–142.

Luo, W., Su, L., Craig, N.J., Du, F., Wu, C., Shi, H., 2019. Comparison of microplastic pollution in different water bodies from urban creeks to coastal waters. Environ. Pollut. 246, 174–182.

Mani, T., Hauk, A., Walter, U., Burkhardt-Holm, P., 2015. Microplastics profile along the Rhine river. Sci. Rep. 5, 7.

Mani, T., Primpke, S., Lorenz, C., Gerdts, G., Burkhardt-Holm, P., 2019. Microplastic pollution in benthic mid-stream sediments of the Rhine River. Environ. Sci. Technol.

McArdle, B.H., Anderson, M.J., 2001. Fitting multivariate models to community data: a comment on distance-based redundancy analysis. Ecology 82, 290–297.

McCormick, A., Hoellein, T.J., Mason, S.A., Schluep, J., Kelly, J.J., 2014. Microplastic is an abundant and distinct microbial habitat in an urban river. Environ. Sci. Technol. 48, 11863–11871.

McCormick, A.R., Hoellein, T.J., London, M.G., Hittie, J., Scott, J.W., Kelly, J.J., 2016. Microplastic in surface waters of urban rivers: concentration, sources, and associated bacterial assemblages. Ecosphere 7.

Mehler, W.T., You, J., Keough, M.J., Lydy, M.J., Pettigrove, V., 2018. Improvements and cost-effective measures to the automated intermittent water renewal system for toxicity testing with sediments. Ecotoxicol. Environ. Saf. 151, 62–67.

Mintenig, S.M., Int-Veen, I., Löder, M.G.J., Primpke, S., Gerdts, G., 2017. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res. 108, 365–372.

Nizzetto, L., Bussi, G., Futter, M.N., Butterfield, D., Whitehead, P.G., 2016. A theoretical assessment of microplastic transport in river catchments and their retention by soils and river sediments. Environ. Sci. Process Impacts 18, 1050–1059.

Peters, C.A., Bratton, S.P., 2016. Urbanization is a major influence on microplastic ingestion by sunfish in the Brazos River Basin, Central Texas, USA. Environ. Pollut. 210, 380–387.

Richardson, J., Zaki, W., 1954. The sedimentation of a suspension of uniform spheres under conditions of viscous flow. Chem. Eng. Sci. 3, 65–73.

Rillig, M.C., 2012. Microplastic in terrestrial ecosystems and the soil? Environ. Sci. Technol. 46, 6453–6454.

Schmidt, C., Krauth, T., Wagner, S., 2017. Export of plastic debris by rivers into the sea. Environ. Sci. Technol. 51, 12246–12253.

Sedlak, D., 2017. Three lessons for the microplastics voyage. Environ. Sci. Technol. 51, 7747–7748.

Seltenrich, N., 2015. New link in the food chain? marine plastic pollution and seafood safety. Environ. Health Perspect. 123, A34–A41.

Sharley, D.J., Sharp, S.M., Bourgues, S., Pettigrove, V.J., 2016. Detecting long-term temporal trends in sediment-bound trace metals from urbanised catchments. Environ. Pollut. 219, 705–713.

Siegfried, M., Koelmans, A.A., Besseling, E., Kroeze, C., 2017. Export of microplastics from land to sea. A modelling approach. Water. Res. 127, 249–257.

Simon-Sánchez, L., Grelaud, M., Garcia-Orellana, J., Ziveri, P., 2019. River Deltas as hotspots of microplastic accumulation: the case study of the Ebro River (NW Mediterranean). Sci. Total Environ. 687, 1186–1196.

Su, L., Cai, H., Kolandhasamy, P., Wu, C., Rochman, C.M., Shi, H., 2018. Using the Asian clam as an indicator of microplastic pollution in freshwater ecosystems. Environ. Pollut. 234, 347–355.

Su, L., Xue, Y., Li, L., Yang, D., Kolandhasamy, P., Li, D., Shi, H., 2016. Microplastics in Taihu lake, China. Environ. Pollut. 216, 711–719.

Sutherland, W.J., Fleishman, E., Clout, M., Gibbons, D.W., Lickorish, F., Peck, L.S., Pretty, J., Spalding, M., Ockendon, N., 2019. Ten years on: a review of the first global conservation horizon scan. Trends Ecol. Evol. 34, 139–153.

Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W.G., McGonigle, D., Russell, A.E., 2004. Lost at sea: where is all the plastic? Science 304, 838-838.

Unice, K.M., Weeber, M.P., Abramson, M.M., Reid, R.C.D., van Gils, J.A.G., Markus, A.A., Vethaak, A.D., Panko, J.M., 2019. Characterizing export of land-based microplastics to the estuary - Part I: Application of integrated geospatial microplastic transport models to assess tire and road wear particles in the Seine watershed. Sci. Total Environ. 646, 1639–1649.

van Wijnen, J., Ragas, A.M.J., Kroeze, C., 2019. Modelling global river export of microplastics to the marine environment: sources and future trends. Sci. Total Environ. 673, 392–401.

Vermaire, J.C., Pomeroy, C., Herczegh, S.M., Haggart, O., Murphy, M., 2017. Microplastic abundance and distribution in the open water and sediment of the Ottawa River, Canada, and its tributaries. Facets 2, 301–314.

Vianello, A., Boldrin, A., Guerriero, P., Moschino, V., Rella, R., Sturaro, A., Da Ros, L., 2013. Microplastic particles in sediments of Lagoon of Venice, Italy: first observations on occurrence, spatial patterns and identification. Estuar. Coast Shelf Sci. 130, 54–61.

Victoria, 2017. Port Phillip Bay Environmental Management Plan. https://www.coastsandmarine.vic.gov.au/__data/assets/pdf_file/0024/88710/PPB-EMP-2017-Main-Doc.pdf.

Wagner, S., Klöckner, P., Stier, B., Römer, M., Seiwert, B., Reemtsma, T., Schmidt, C., 2019. Relationship between discharge and river plastic concentrations in a rural and an urban catchment. Environ. Sci. Technol. 53, 10082–10091.

Wang, Z., Su, B., Xu, X., Di, D., Huang, H., Mei, K., Dahlgren, R.A., Zhang, M., Shang, X., 2018. Preferential accumulation of small ($<300\ \mu m$) microplastics in the sediments of a coastal plain river network in eastern China. Water Res. 144, 393–401.

Wright, S.L., Kelly, F.J., 2017. Plastic and human health: a micro issue? Environ. Sci. Technol. 51, 6634–6647.

Wright, S.L., Thompson, R.C., Galloway, T.S., 2013. The physical impacts of microplastics on marine organisms: a review. Environ. Pollut. 178, 483–492.

Yang, D.Q., Shi, H.H., Li, L., Li, J.N., Jabeen, K., Kolandhasamy, P., 2015. Microplastic pollution in table salts from China. Environ. Sci. Technol. 49, 13622–13627.

Yonkos, L.T., Friedel, E.A., Perez-Reyes, A.C., Ghosal, S., Arthur, C.D., 2014. Microplastics in four estuarine rivers in the chesapeake Bay. USA. Environ. Sci. Technol. 48, 14195–14202.

Zhang, K., Xiong, X., Hu, H., Wu, C., Bi, Y., Wu, Y., Zhou, B., Lam, P.K.-S., Liu, J., 2017. Occurrence and characteristics of microplastic pollution in Xiangxi bay of three Gorges reservoir, China. Environ. Sci. Technol.

Zwanzig, R., 1964. Hydrodynamic fluctuations and Stokes' law friction. J. Res. Natl. Bur. Std.(US) B 68, 143–145.