Chemosphere 162 (2016) 277–284

Contents lists available at ScienceDirect

Chemosphere

journal homepage: www.elsevier.com/locate/chemosphere

Influence of wastewater treatment plant discharges on microplastic concentrations in surface water

Shirin Estahbanati, N.L. Fahrenfeld*

Department of Civil and Environmental Engineering, Rutgers, The State University of New Jersey, 96 Frelinghuysen Rd., Piscataway, NJ 08854, United States

HIGHLIGHTS

- Microplastic particles are emerging contaminant in the freshwater environment.
- Distribution of microplastic particles in four size categories up to 2 mm was determined.
- Microplastic in 125 μm and 250 μm size classes increased downstream of several WWTP.
- Secondary were more abundant than primary microplastics across the study area.

GRAPHICAL ABSTRACT

ARTICLE INFO

Article history:
Received 3 April 2016
Received in revised form
23 July 2016
Accepted 25 July 2016
Available online 7 August 2016

Handling Editor: Tamara S. Galloway

Keywords:
Microplastics
Wastewater
Emerging contaminants

ABSTRACT

The abundance of microplastic particles in the marine environment is well documented, but less is known about microplastics in the freshwater environment. Wastewater treatment plants (WWTPs) may not effectively remove microplastics allowing for their release to the freshwater environment. To investigate concentration of microplastic in fresh water and the impact of WWTP effluent, samples were collected upstream and downstream of four major municipal WWTPs on the Raritan River, NJ. Microplastics were categorized into three quantitative categories (500–2000 μm, 250–500 μm, 125–250 μm), and one semi-quantitative category (63–125 μm). Then, microplastics were classified as primary (manufactured in small size) or secondary (derived from larger plastics) based on morphology. The concentration of microplastics in the 125–250 and 250–500 μm size categories significantly increased downstream of WWTP. The smaller size classes, often not quantified in microplastic studies, were in high relative abundance across sampling sites. While primary microplastics significantly increased downstream of WWTP, secondary microplastic was the dominant type in the quantitative size categories (66–88%). A moderate correlation between microplastic and distance downstream was observed. These results have implications for understanding the fate and transport of microplastics in the freshwater environment.

1. Introduction

Worldwide plastic production has been growing since 1950 (Plastics Europe, 2013). Consequently, millions of tons of plastics enter oceans and landfills each year (Gourmelon, 2015). All oceans have been affected by plastic pollution (Wright et al., 2013). Plastics entering aquatic environments have a wide size distribution, ranging from micrometers to meters (Hidalgo-Ruz et al., 2012). Microplastics are defined as plastic particles smaller than 5 mm

* Corresponding author.
E-mail address: nfahrenf@rutgers.edu (N.L. Fahrenfeld).

http://dx.doi.org/10.1016/j.chemosphere.2016.07.083
0045-6535/

(Arthur et al., 2008) derived from larger particles (secondary microplastics) or manufactured in small size (primary microplastics) (Hidalgo-Ruz et al., 2012). In the United States it is estimated that 8 trillion microplastic beads enter the aquatic environment daily (Rochman et al., 2015). Due to slow rates of plastic degradation, microplastics persist in the environment (Eerkes-Medrano et al., 2015). The presence and consequences of microplastics in the marine environment have been studied since 1970 (Carpenter et al., 1972). However, less is known about microplastic abundance in the freshwater environment (Eerkes-Medrano et al., 2015).

Accumulation of microplastic in lakes (Faure et al., 2012; Eriksen et al., 2013), estuaries (Sadri and Thompson, 2014), and rivers (Lechner et al., 2014; McCormick et al., 2014) has been reported. There is a high concentration of microplastics in WWTP influent (10^4 to 10^5microplastic/m^3) and incomplete removal during the treatment process (70–100%) can result in microplastic pollution in the receiving water (Magnusson and Wahlberg, 2014). WWTP effluent has been identified as one of the sources of microplastics in the freshwater environment (Magnusson and Norén, 2014). WWTP effluent resulted in an increase in the concentration of microplastic in Chicago River (McCormick et al., 2014). However, the cumulative impact of WWTP effluents along a river has not been demonstrated. In addition, insufficient removal of microplastics <300 μm in WWTPs has been reported (Magnusson and Wahlberg, 2014). Further, most microplastic studies focus on plastics larger than 330 μm, overlooking the smaller size classes which are potentially important sources of microplastic pollution in the freshwater environment.

The objective of this study was to investigate the abundance of microplastic and the impact of municipal WWTP effluents on the microplastic concentration in the Raritan River. In this study, the presence of microplastics in a wider size range (125 μm-2mm) is reported. Based on morphology, microplastics were categorized into primary and secondary groups to aid in identification of the sources of microplastic contamination. Moreover, correlations between distances downstream and microplastic concentration were tested to provide insight into the fate and transport of microplastic in the river environment.

2. Materials and method

2.1. Sampling

Sampling was performed on the Raritan River, located in central New Jersey (NJ), US (Fig. 1). The river basin covers 2850 km^2 and provides water for drinking, irrigation, agriculture, recreation, and industry. The River has two branches, north and south, that meet then flow into the Raritan Bay. The primary land use of the river main stem is urban and suburban, (51.3%) and the primary land use of the south and north branches are agricultural and forest (61.3%) (Newcomb et al., 2000). More than 10 municipal WWTPs discharge into the Raritan River, five of which are major [>1 million gallons per day (MGD)].

Samples were collected upstream and downstream of three major municipal WWTPs located upstream of head-of-tide (selected based on ease of access for sampling). Sites included two discharging into south branch (WWTP-A1 with design flow 2.3 MGD and A2 with design flow 3.8 MGD) and one into the north branch (WWTP-B, design flow 5 MGD). Samples were also collected upstream of the confluence with the Millstone River (with WWTP-D, design flow 9.2 MGD and WWTP-E, design flow 4.45 MGD) and downstream of a major WWTP (WWTP-C, design flow 23 MGD and) discharging into the main branch of the Raritan. These locations will be referred to as WWTP-M/C and were also upstream of head of tide. A background site was selected on the south branch (Background) as a control without WWTP discharge upstream.

Samples were collected during baseflow with plankton nets (0.2 m diameter, 0.51 m long) with 153 μm mesh size (Fieldmaster, Lenexa, Kansas) in duplicate in October–November 2015. The nets were fixed perpendicular to flow on the river surface, with half of net opening submerged to collect floating particles. The water velocity was estimated at the sampling locations by the float method and verified using a pygmy meter (USGS Model 6200 AA, Columbus, Ohio). Samples were collected for 1 h. Sampling was performed downstream first, then upstream of a given WWTP, with paired samples collected within 3–72 h of one another. The volume of sample collected was calculated by taking the product of river surface velocity, cross sectional area of the submerged portion of the net opening, and sample collection time. Nets were transferred to the lab for analysis. Field blanks were performed by pouring Deionized (DI) water (5×10^{-3} m^3) through the net in the field, then leaving the net open and exposed to air for 1 h. Matrix spike duplicates were performed in the field by adding 1 g of personal care product containing polyethylene to the net after sampling but prior to microplastic extraction. The size distribution of the microplastic in four personal care products was determined by dissolving the product in the hot water (100 °C) and categorizing the particle size by wet sieving (63–125 μm, 125–250 μm, 250–500 μm, and 500–2000 μm classes).

2.2. Extraction of microplastics

The contents of each net were rinsed with DI water (0.25 m^3) three times into a series of sieves (4000, 2000, 500, 250, 125, and 63 μm aperture size). Material captured on the largest two size categories of sieves was discarded. Then, the contents of each sieve were rinsed with DI water, transferred to a 200 mL beaker, and dried overnight at 90 °C. The organic content of each sample was oxidized by hydrogen peroxide catalyzed by iron (II) (Baker et al., 2015). Iron (II) solution (20 mL, 0.05 M) was added to each beaker, following by 20 mL hydrogen peroxide. The solutions were heated to 75 °C for 30 min after which sodium chloride was added to increase the mixture density. Then, the solutions were transferred to a funnel to facilitate density separation, covered with foil, and left overnight for settling. In the density separation step, dense particles settled and buoyant particles, including microplastics, floated on the surface. Settled materials were discarded and floating particles were rinsed with DI water and transferred to a glass petri dish.

Recovered particles were visualized under a reflected microscope (Stereo Zoom Microscope, Olympus, Japan). For the 500 μm size category, plastics were counted directly. Due to the high abundance of particles in the 63, 125, and 250 μm size categories, the area of each petri dish was divided into an 80 block grid (29.3 mm^2), and random grid blocks (20–30 blocks) were counted per sample. The total number of microplastics was calculated by scaling up the number of counted microplastics based on the surface area of grids counted. This method was found to be accurate within 1.7–9.6% compared to counting the total number of microplastics directly (Fig. A1, A2). During visualization, the microplastic particles were categorized as primary and secondary microplastic based on visual inspection of particle morphology. Morphology of the plastic particles collected in the field was compared to plastic particles extracted from a variety of personal care products containing polyethylene (Fig. 2). The microplastics were classified as primary or secondary based on shape and surface texture (i.e., smooth edges/texture, symmetrical shape classified as primary). The concentration of microplastics in field samples was determined by dividing the number of microplastics counted by the

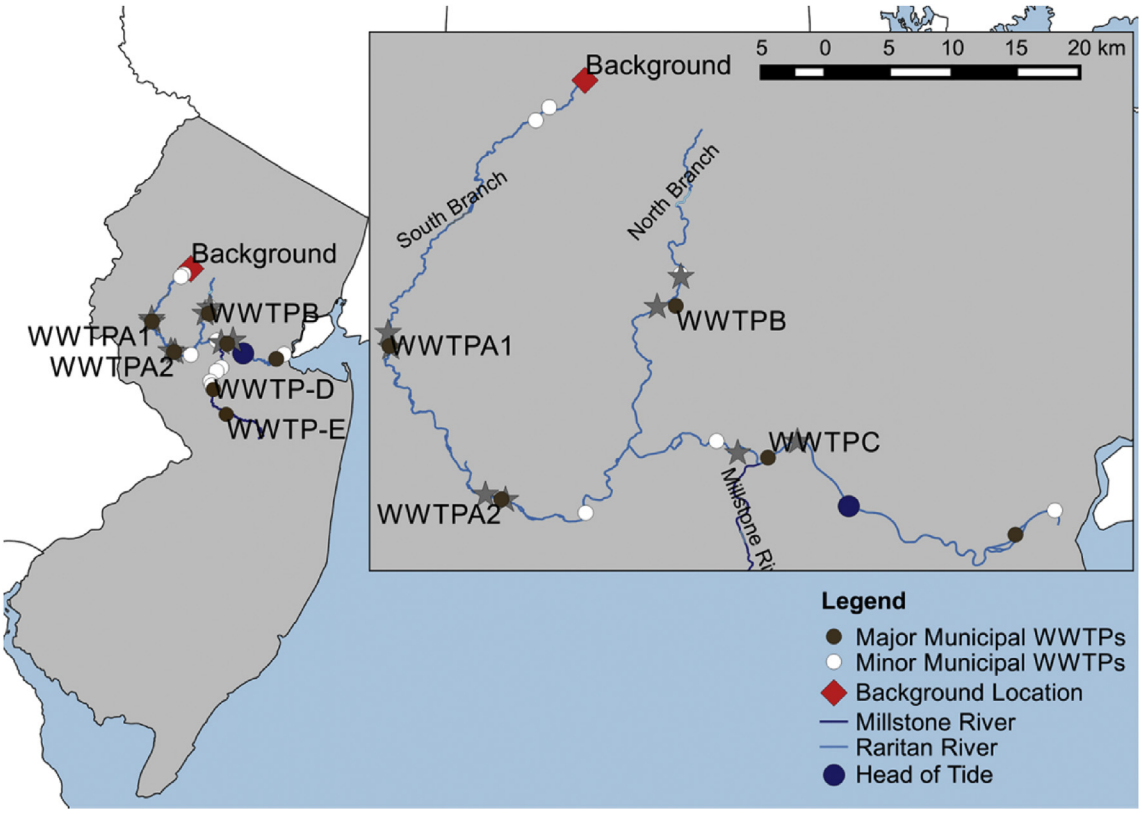

Fig. 1. Map of sampling locations. Sampling was performed up and downstream of four major municipal WWTP [>1 million gallons/day (MGD)] and at a Background location. Minor WWTP (<1MGD) are present on the river.

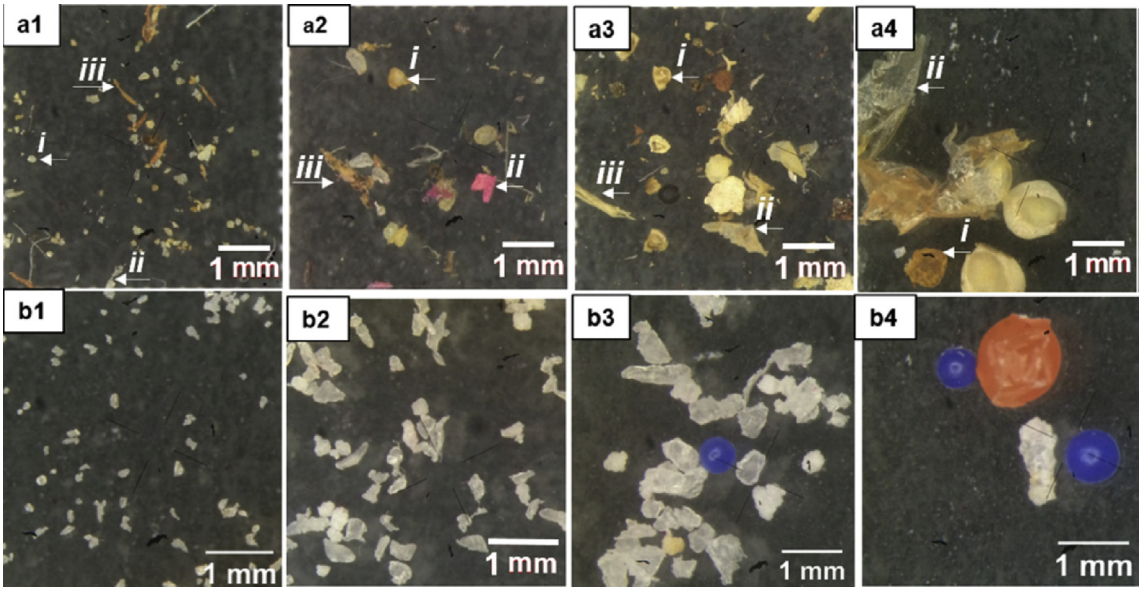

Fig. 2. (A) Microplastics recovered in samples in the 1) 63–125 μm size category, 2) 125–250 μm size category, 3) 250–500 μm size category, and 4) 500–2000 μm size category. (B) Microplastics recovered from personal care products in the 1) 63–125 μm size category, 2) 125–250 μm size category, 3) 250–500 μm size category, and4) 500–2000 μm size category. Examples of different particles classifications are labeled i) primary microplastic, ii) secondary microplastic, and iii) non-microplastic particles excluded during the counting step.

volume of sample collected (the product of the cross sectional area of the submerged net opening, river velocity, and length in time of sample collection).

A Student's t-test was applied for normal and a Wilcox Rank Sum test for non-parametric data to compare (a) the total concentration of microplastics upstream and downstream of each WWTP for a given size category, (b) the concentration of primary and secondary microplastics for a given size category, and (c) the

upstream to downstream concentration of primary microplastics for a given size category across all WWTPs. Correlation between microplastic concentration and distance downstream was tested for the South and Main Branch of the River using linear regression in Excel (Microsoft, Silicon Valley, California). A Kruskal-Wallis test followed by a post-hoc pairwise *t*-test with a Bonferroni correction for multiple comparisons was performed to compare the concentration of microplastics in the four size categories across all sampling sites.

3. Results

Microplastics of all size categories were observed in all samples, including those collected at the background site (Fig. 3). Of the four major WWTPs sampled, microplastic concentrations increased downstream of WWTP-A1, A2, and M/C in the 125 μm size category ($p = 0.024–0.048$), and for WWTP-A2 and M/C in the 250 μm size category ($p = 0.0012–0.0052$) compared to upstream samples. No differences were observed in concentration of microplastics in samples collected downstream of the WWTP discharge compared to samples collected upstream for the 63 and 500 μm size categories ($p = 0.075–0.67$). The 500 μm size category was the least abundant size class across all sampling sites ($p = 0.0014–0.013$). There was significantly more microplastics observed in the 125 μm size category than the 250 μm size category across all sampling sites ($p = 0.0088$).

The average relative percent differences for field replicates were 44.9% for the 63 μm, 19.8% for the 125 μm, 26.2% for 250 μm, and 27.4% for 500 μm size categories. The average recovery of microplastics in matrix spikes was 45.1% for 63 μm, 75.1% for 125 μm, 97.5% for 250 μm, and 54.4% for 500 μm size category. Concentrations of microplastic by size class in the matrix spike and in three other personal care products are shown as Fig. A3. Microplastics observed in the blank samples were 19.4 times lower than observed in any river sample at upstream of the WWTP –A1 and 3 times lower than observed in at the background location.

Microplastics were categorized as primary and secondary plastics based on visual inspection of morphology (Figs. 2 and 4, Fig. A3). In a pair-wise comparison across all sampling sites, secondary microplastics were more abundant than primary microplastics in the 125, 250 and 500 μm categories ($p = 0.038$, 0.0039, and 0.0038, respectively). The average percent of secondary microplastics was 42.4% for the 63 μm, 73.1% for the 125 μm, 66.9% for the 250 μm, and 87.9% for the 500 μm size category. Comparing upstream and downstream samples for all WWTPs, the concentrations of primary microplastics increased downstream of WWTPs in the 63, 125, and 250 μm size categories ($p = 0.0078$, 0.0078, and 0.023, respectively).

A moderate correlation was observed between distance from Raritan Bay and the total concentration of microplastics (slope = 1.82, $R^2 = 0.42$, Fig. A4) and for the concentration of microplastics in the 125–2000 μm size categories (slope = 1.14, $R^2 = 0.46$, Fig. A5). Two out of seven of the locations for all size categories including semi-quantitative and four of seven sampling locations for the quantitative size categories were within the 95% confidence interval for the regressions. The pattern of regression residuals indicates that concentrations decreased between the sample collected downstream of a given WWTP and the next upstream sampling location. For example, downstream of WWTP-A1 microplastic concentration was 43.9 microplastics/m^3 and 19.3 km downstream (at the upstream sampling location for WWTP-A2) microplastic concentration was 27.8 microplastics/m^3.

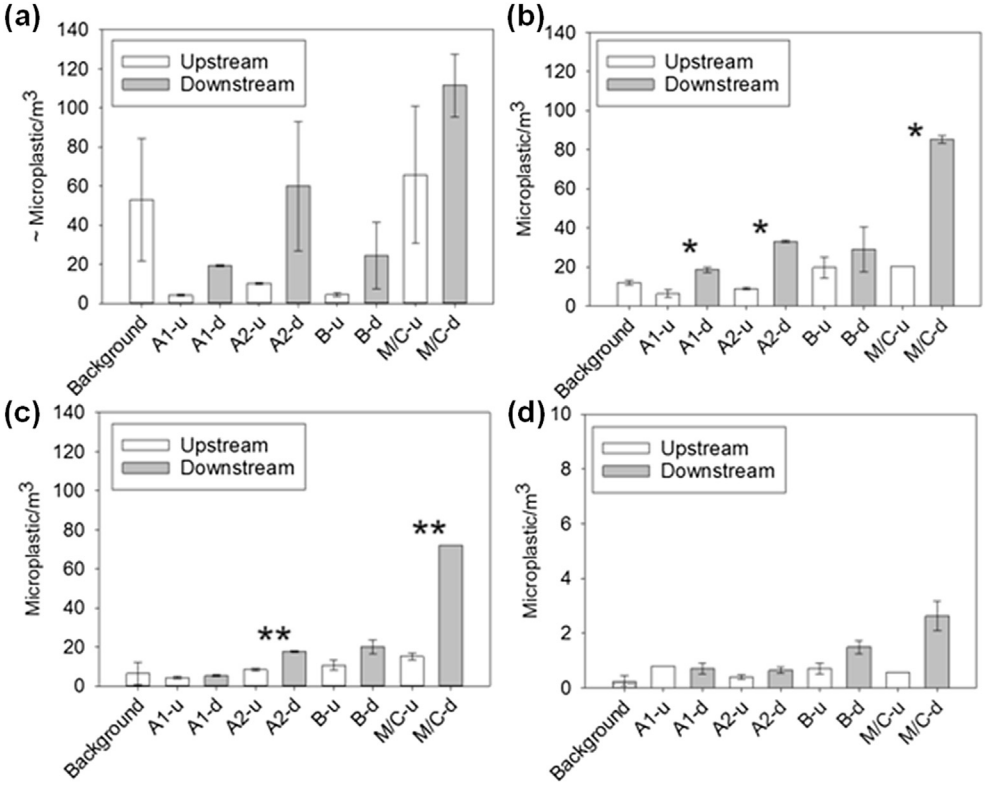

Fig. 3. Concentration of microplastics in a. 63–125 μm, b. 125–250 μm, c. 250–500 μm, and d. 500–2000 μm size categories upstream (white) and downstream (gray) of WWTPs. Concentrations for 63 μm size category is semi-quantitative because the net used for sampling had an aperture of 153 μm. Error bars represent standard deviation of field duplicates. P value for significant differences observed comparing upstream and downstream concentrations for a given WWTP and size category are for $p < 0.05$ with * and $p < 0.01$ with. **.

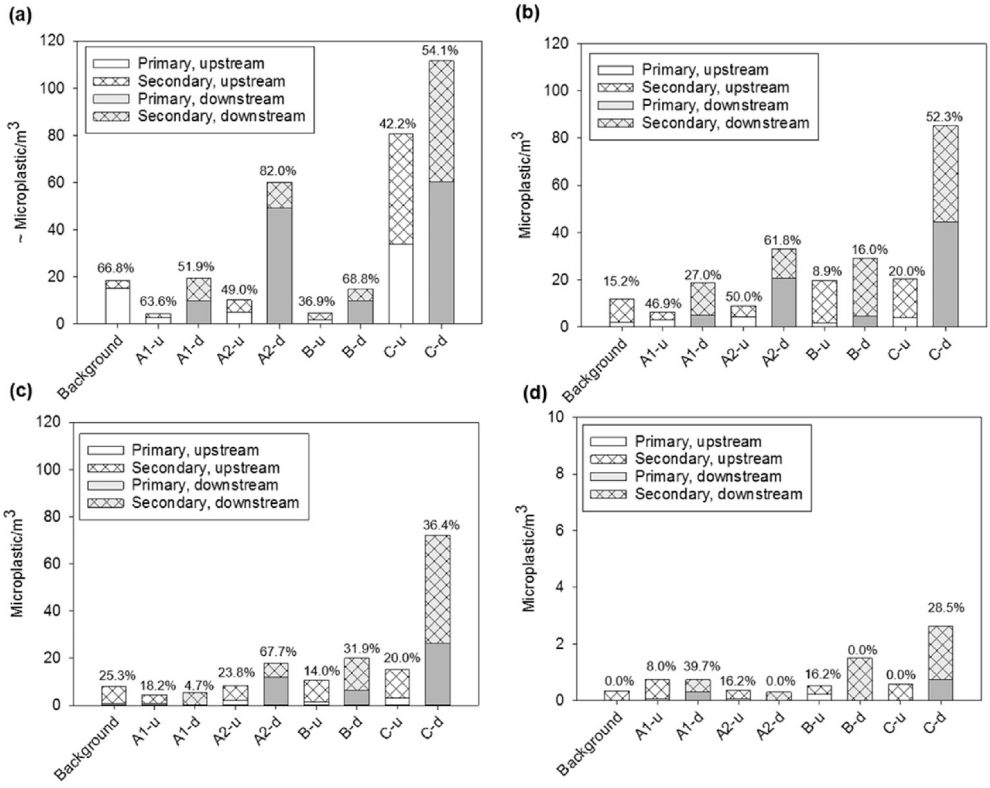

Fig. 4. Concentration of primary (solid) and secondary (cross hatched) microplastics in a. 63–125 μm, b. 125–250 μm, c. 250 μm-500 μm, and d. 500–2000 μm size categories upstream (white) and downstream (gray) of WWTPs. Concentrations for 63 μm size category is semi-quantitative because the net used for sampling had an aperture of 153 μm. Average percent of primary microplastics at a given location are listed above the bars.

4. Discussion

This study provides insight into the impact of wastewater treatment plant discharges on the prevalence of a wide size distribution of microplastics in the freshwater environment. Increases in microplastic concentrations were observed for the 125–250 μm and 250–500 μm size categories downstream of select wastewater treatment plants on the Raritan River. This result is consistent with results of McCormick et al. (2014) who observed microplastic concentrations (including fibers, which were not quantified in this study) increased downstream of a WWTP on the Chicago River. WWTPs are known sources of microplastics in the freshwater environment given their incomplete removal during wastewater treatment (Magnusson and Norén, 2014). Interestingly, the microbeads in the 500 μm category, which have received considerable attention (Eriksen et al., 2013; Lechner et al., 2014), were in low abundance compared to other microplastic types. At the background location on the Raritan River microplastic was observed with concentrations 3.8 times less than the most contaminated sampling location (downstream of WWTP-C) but 4.6 times greater than the site with the lowest plastic concentration, upstream of WWTP-A1. This indicates sources of microplastic other than WWTP are present in the river. While fibers were observed in many samples, quantitative analysis for fibers was not performed due to the difficulty in accurately counting these plastics which were often present in knotty masses.

Given that several major WWTP are located along the Raritan River, the concentration of microplastics as a function of downstream distance was investigated. While microplastic concentrations were greatest at the furthest downstream sampling point (WWTP-C), a strong correlation between distance downriver and microplastic concentration was not observed. Microplastic concentrations decreased between any given WWTP downstream sampling site and the next closest upstream sampling site (e.g., WWTP-A1 downstream and WWTP-A2 upstream). Uptake by biota (Barnes and Milner, 2005; McCormick et al., 2014), dilution, settling (Castañeda et al., 2014; Wagner et al., 2014), and/or skimming of microplastic particles during transport could account for the decrease in concentration of microplastics between the sampling locations downstream of a WWTP to the upstream of the next WWTP. it is also possible that temporal variations in microplastic concentration obscure the trend given that samples for different locations were collected up to one week apart. Daily and seasonal temporal variation was observed by other researchers (Magnusson and Norén, 2014). Given that all sampling locations were upstream of head of tide, plastic pollution in Raritan Bay should not have influenced these observations.

The presence of microplastics at the furthest downstream sampling location indicates that the Raritan River is likely a source of microplastics in the receiving estuary. Widespread observation of microplastics in the New York/New Jersey Harbor estuary was recently reported (New York New Jersey Baykeeper, 2016). Therefore, our results indicate that rivers serve as a source of the microplastics in the marine environment as suggested by Eerkes-Medrano et al. (2015).

Comparison of results among microplastic studies is complicated due to the variations in sampling techniques (i.e., size classes targeted), plastic categories quantified (e.g., primary, secondary, beads, and/or fibers), and units used in reporting results (Hoellein et al., 2014). Notably, some researchers report microplastic abundance based on number of microplastics per volume of collected samples [e.g., (Lechner et al., 2014)], while others report the

Table 1
Comparison of microplastics observed in this study to select other studies.

Location	Freshwater upstream of WWTP (No./m^3)	Freshwater downstream of WWTP (No./m^3)	Freshwater, unspecified (No./m^3)	Marine water (No./m^3)	Study units	Net mesh size (μm)	Sampled size range	Volume/Distance sampled	Reference
Raritan River*	24 ± 11.4*	71.7 ± 60.2*			No./m^3	153	125–2000 μm	1.3–13.5 m^3	This study
Chicago River	1.9 ± 0.8	17.9 ± 11.0			No./m^3	333	333–2000 μm	Not Reported	(McCormick et al., 2014)
Danube River			$3.2 \times 10^{-2} \pm 4.7$		Items 1000 m^3	500	<2 mm, 2–20 mm	Maximum 1 km	(Lechner et al., 2014)
Swedish west coast waters				102,550	No./m^3	80	500–2000 μm	5–19 km	(Norén, 2007)
South California Shore				3.9	No./m^3	333	300–500 μm 500–700 μm 700–1000 μm 1–2.8 mm 2.8–4.7 mm	0.5–1 km	(Lattin et al., 2004)
Long Beach California				7.3	No./m^3	333	300–500 μm 500–700 μm 700–1000 μm 1–2.8 mm 2.8–4.7 mm	0.5–1 km	(Moore et al., 2002)
North Pacific Ocean				2.2	No./m^3	333	300–500 μm 500–700 μm 700–1000 μm 1–2.8 mm 2.8–4.7 mm	5–19 km	(Moore et al., 2001)
Hovsgol Lake**			12.7×10^{-7}**		Items km^{-2}	333	355–999 μm 1–4.7 mm >4.7 mm	0.4–8.5 km	(Free et al., 2014)
Laurentian Great Lakes**			27.0×10^{-7}**		Particle km^{-2}	333	355–999 μm 1–4.7 mm >4.7 mm	1.94–4 km	(Eriksen et al., 2013)
Tamar estuary			0.028		Items/m^3	300	<1 mm 1–3 mm 3–5 mm >5 mm	3.7 km	(Sadri and Thompson, 2014)

*63–125 μm were counted in samples, but not included here because net mesh size was greater than size class, ** Concentration of particles per area was converted to NO./m^3 by dividing reported numbers by the height of the net used for sampling.

microplastic concentration based on surface area of field sampling [e.g., (Eriksen et al., 2013)]. Nonetheless, microplastic concentrations observed in this study were generally greater than other freshwater studies and within the range reported for marine studies (Table 1). For all comparisons of the concentration of microplastic across studies discussed here, only particles in the 125–2000 μm size class for the Raritan River are considered so that comparisons are quantitative. Further, particles per unit area reported by other researchers were converted to particles per unit volume for consistency of units. The concentration of microplastic in Raritan River was higher than other studies of microplastic in Chicago river (McCormick et al., 2014) and Danube River (Lechner et al., 2014). The concentrations of microplastic observed in this study were less than the maximum value in the marine environment (Norén, 2007), but microplastic concentrations were higher than concentrations for ocean studies along the Southern California shore, Southern California's coastal water, and North Pacific Ocean (Lattin et al., 2004). The microplastic concentration in the river was higher than the concentration in the Hovsgol and Laurentian Great lakes (Eriksen et al., 2013; Free et al., 2014) and the Tamar estuary (Sadri and Thompson, 2014). The higher concentrations observed in this study are likely to due to the smaller net mesh sized used here than in most other studies given that the smaller size classes were in higher relative abundance in our study. It is also possible that converting from particles per area to particles per volume for other studies under estimated the concentrations reported given that it was assumed the nets were fully submerged. Because many studies rely on visual identification including this one, there is potential to over or under count plastic particles and follow-up studies including chemical identification like FTIR are recommended.

To the authors' knowledge, this is the first study of the distribution of microplastic smaller than 355 μm in the fresh water environment. Other researchers concentrated on quantifying larger microplastics: 355–900 μm in Hovsgol Lake (Free et al., 2014), 500–2000 μm in Danube River (Lechner et al., 2014), and 300–1000 μm in Tamar Estuary (Sadri and Thompson, 2014). Microplastics in 63–125 μm and 125–250 μm size categories dominated the personal care products tested in the lab (constituting 3.5–34.4% and 45–71% of the microplastics in the tested cosmetic products, Fig. A6). Because the plankton net used in this study had a mesh size of 153 μm, the particles collected on the 63 μm sieve should be considered semi-quantitative as particles of in this size category can pass through the net. Despite this, the 63 μm size category was the most abundant in 6 out of 9 of our sampling locations, and in the remaining 3 locations 125 μm size category was the dominant category. Lower removal rates have been reported for smaller microplastics (<300 μm) inWWTPs (Magnusson and Wahlberg, 2014). Sampling with nets with smaller mesh size is recommended to quantify this size category given results for the extraction of personal care products and the semi-quantitative field data presented here indicate this size class may be the dominant size category. Visually differentiating and chemically confirming the composition of these particles presents challenges but may be worthwhile nonetheless because these smaller microplastics provide more specific area for adsorption of contaminants and growth of biofilm. Both sorption of hazardous organic contaminants (Moore, 2008) and biofilm growth (Hoellein et al., 2014) have been reported on microplastic surfaces.

Microplastics measured in the Raritan River were classified as primary or secondary to provide insight into the potential source. Secondary microplastics result from the fragmentation of larger pieces of plastic as opposed to from the use of small plastic particles in personal care products. The increasing concentration of primary microplastics downstream of the all WWTPs in >63, 125, and 250 μm categories highlights the role of WWTPs as a source of primary microplastics. The presence of primary microplastics at the background location indicates that WWTP effluent was not the only source and/or may be indicative of inaccuracies in this visual class categorization. However, across all sampling sites in the >125, 250, and 500 μm size classes, secondary microplastics were present at higher concentrations. Secondary microplastics are formed by biological degradation, photodegradation, chemical deposition, and physical breakdown of larger pieces of plastics (Andrady, 2011). The sources of secondary microplastics are varied and may include cigarettes, plastic bags, and tires (Hoellein et al., 2014). Other studies have categorized microplastics based on different characteristics such as chemical composition (e.g. polypropylene, polystyrene) (Morét-Ferguson et al., 2010), shape (e.g. color, texture) (Corcoran et al., 2015), size (Eriksen et al., 2013), and type (e.g. food wrappers, tobacco packing) (Morritt et al., 2014), therefore direct comparisons are not currently feasible.

Current legislation in the US focuses on phasing out the production of personal care products containing non-degradable microplastics less than 5 mm in size (Energy Commerce Committee, 2015) by July 2018. In addition, the Netherlands, Austria, Belgium and Sweden have supported bans on the production of microplastics used in detergents and cosmetic products (Simon, 2014). This action would be expected to decrease the release of primary microplastics in WWTP impacted waters and may decrease the levels of secondary microplastics formed from the primary microplastic particles in personal care products.

5. Conclusions

This study demonstrated the spatial pattern of primary and secondary microplastics in the Raritan River. The results of this study indicated that microplastic concentration in select size categories, particularly primary microplastics, increased downstream of several WWTP outfalls. Additionally, the presence of microplastics at the background location showed that WWTPs are not the only source of microplastic contamination in the river. Correlations between distance from the Raritan Bay and concentration of microplastic indicated other fate and transport processes may be driving microplastic concentrations between WWTP outfalls including potentially microplastic digestion by biota, settling, skimming, and dilution. To the author's knowledge, this is the first report of the abundance of microplastic in small size categories (125–2000 μm quantitatively, and 63–125 μm semi-quantitatively). The high concentrations observed for the smaller size categories indicate these are potentially important in the freshwater environment, especially given their high surface areas, and should be confirmed by chemical identification.

Acknowledgements

Thanks to Alessia Eramo and Johnny Quispe for sampling assistance. This research was supported by university start-up funds to NLF, a Presidential fellowship to SE, and a NJ Water Resources Research Institute Graduate Student Grant-in aid.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.chemosphere.2016.07.083.

References

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596–1605.
Arthur, C., Baker, J., Bamford, H., 2008. International Research Workshop on the

Occurrence, Effects, and Fate of Microplastic Marine Debris.
Baker, J., Foster, G., Masura, J., Arthur, C., 2015. Laboratory Methods for the Analysis of Microplastics in the Marine Environment: Recommendations for Quantifying Synthetic Particles in Waters and Sediments. NOAA Technical Memorandum.
Barnes, D., Milner, P., 2005. Drifting plastic and its consequences for sessile organism dispersal in the Atlantic Ocean. Mar. Biol. 146, 815–825.
Carpenter, E.J., Anderson, S.J., Harvey, G.R., Miklas, H.P., Peck, B.B., 1972. Polystyrene spherules in coastal waters. Science 178, 749–750.
Castañeda, R.A., Avlijas, S., Simard, M.A., Ricciardi, A., Smith, R., 2014. Microplastic pollution in St. Lawrence river sediments. Can. J. Fish. Aquatic Sci. 71, 1767–1771.
Corcoran, P.L., Norris, T., Ceccanese, T., Walzak, M.J., Helm, P.A., Marvin, C.H., 2015. Hidden plastics of Lake Ontario, Canada and their potential preservation in the sediment record. Environ. Pollut. 204, 17–25.
Eerkes-Medrano, D., Thompson, R.C., Aldridge, D.C., 2015. Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res. 75, 63–82.
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.
Faure, F., Corbaz, M., Baecher, H., de Alencastro, L., 2012. Pollution due to plastics and microplastics in lake Geneva and in the Mediterranean sea. Archives Sci. 65, 157–164.
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.
Gourmelon, G., 2015. Global Plastic Production Rises, Recycling Lags. Worldwatch Institute (Online). Available: http://vitalsigns.worldwatch.org/sites/default/files/vital_signs_trend_plastic_full_pdf.pdf.
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.
Hoellein, T., Rojas, M., Pink, A., Gasior, J., Kelly, J., 2014. Anthropogenic litter in urban freshwater ecosystems: distribution and microbial interactions. Plos One 9.
Lattin, G.L., Moore, C.J., Zellers, A.F., Moore, S.L., Weisberg, S.B., 2004. A comparison of neustonic plastic and zooplankton at different depths near the southern California shore. Mar. Pollut. Bull. 49, 291–294.
Lechner, A., Keckeis, H., Lumesberger-Loisl, F., Zens, B., Krusch, R., Tritthart, M., Glas, M., Schludermann, E., 2014. The Danube so colourful: a potpourri of plastic litter outnumbers fish larvae in Europe's second largest river. Environ. Pollut. 188, 177–181.
Magnusson, K., Norén, F., 2014. Screening of Microplastic Particles in and Downstream a Wastewater Treatment Plant. IVL Swedish Environmental Research Institute.
Magnusson, K., Wahlberg, C., 2014. Mikroskopiska Skräppartiklar I Vatten Från Avloppsreningsverk. Rapport NR B 2208, p. 33.
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.
Moore, C.J., 2008. Synthetic polymers in the marine environment: a rapidly increasing, long-term threat. Environ. Res. 108, 131–139.
Moore, C.J., Moore, S.L., Leecaster, M.K., Weisberg, S.B., 2001. A comparison of plastic and plankton in the North Pacific central gyre. Mar. Pollut. Bull. 42, 1297–1300.
Moore, C.J., Moore, S.L., Weisberg, S.B., Lattin, G.L., Zellers, A.F., 2002. A comparison of neustonic plastic and zooplankton abundance in southern California's coastal waters. Mar. Pollut. Bull. 44, 1035–1038.
Morét-Ferguson, S., Law, K.L., Proskurowski, G., Murphy, E.K., Peacock, E.E., Reddy, C.M., 2010. The size, mass, and composition of plastic debris in the western North Atlantic Ocean. Mar. Pollut. Bull. 60, 1873–1878.
Morritt, D., Stefanoudis, P.V., Pearce, D., Crimmen, O.A., Clark, P.F., 2014. Plastic in the Thames: a river runs through it. Mar. Pollut. Bull. 78, 196–200.
New York New Jersey Baykeeper, 2016. NY-NJ Harbor Estuary Plastic Collection Report (Online) Available: http://nynjbaykeeper.org/wp-content/uploads/2016/02/NYNJBaykeeper-Plastics-Report-February-2016.pdf.
Newcomb, D.J., Stanuikynas, T.J., Van Abs, D.J., 2000. Setting of the Raritan River Basin: a Technical Report for the Raritan Basin Watershed Management Project. NJ Water Supply Authority.
Norén, F., 2007. Small Plastic Particles in Coastal Swedish Waters. KIMO Sweden.
Plastics Europe, 2013. Plastics-the Facts 2013: An Analysis of European Latest Plastics Production, Demand and Waste Data. (Online) Available: http://www.plasticseurope.org/documents/document/20131014095824-final_plastics_the_facts_2013_published_october2013.pdf.
Rochman, C.M., Kross, S.M., Armstrong, J.B., Bogan, M.T., Darling, E.S., Green, S.J., Smyth, A.R., Veríssimo, D., 2015. Scientific evidence supports a ban on microbeads. Environ. Sci. Technol. 49, 10759–10761.
Sadri, S.S., Thompson, R.C., 2014. On the quantity and composition of floating plastic debris entering and leaving the Tamar Estuary, Southwest England. Mar. Pollut. Bull. 81, 55–60.
Simon, F., 2014. Dutch Rally Support for Microplastic Ban to Safeguard Their Mussels. EurActiv (Online). Available: https://www.euractiv.com/section/science-policymaking/news/dutch-rally-support-for-microplastic-ban-to-safeguard-their-mussels/.
U.S. Energy Commerce Committee, 2015. Microbead-free Waters Act of 2015. HR 1321. 114th Congress.
Wagner, M., Scherer, C., Alvarez-Muñoz, D., Brennholt, N., Bourrain, X., Buchinger, S., Fries, E., Grosbois, C., Klasmeier, J., Marti, T., 2014. Microplastics in freshwater ecosystems: what we know and what we need to know. Environ. Sci. Eur. 26, 12.
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.