



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



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## 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  $\mu\text{m}$  and 250  $\mu\text{m}$  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

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  $\mu\text{m}$ , 250–500  $\mu\text{m}$ , 125–250  $\mu\text{m}$ ), and one semi-quantitative category (63–125  $\mu\text{m}$ ). 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  $\mu\text{m}$  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.

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## 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

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(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  $10^5$  microplastic/ $m^3$ ) 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  $\mu m$  in WWTPs has been reported (Magnusson and Wahlberg, 2014). Further, most microplastic studies focus on plastics larger than 330  $\mu m$ , 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  $\mu m$ –2 mm) 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  $\mu m$  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 ( $5 \times 10^{-3} m^3$ ) 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  $\mu m$ , 125–250  $\mu m$ , 250–500  $\mu m$ , and 500–2000  $\mu m$  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  $\mu m$  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, followed 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  $\mu m$  size category, plastics were counted directly. Due to the high abundance of particles in the 63, 125, and 250  $\mu m$  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

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