



Transport and fate of microplastic particles in wastewater treatment plants



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ABSTRACT

Municipal wastewater treatment plants (WWTPs) are frequently suspected as *significant* point sources or conduits of microplastics to the environment. To directly investigate these suspicions, effluent discharges from seven tertiary plants and one secondary plant in Southern California were studied. The study also looked at influent loads, particle size/type, conveyance, and removal at these wastewater treatment facilities. Over 0.189 million liters of effluent at each of the seven tertiary plants were filtered using an assembled stack of sieves with mesh sizes between 400 and 45 μm . Additionally, the surface of 28.4 million liters of final effluent at three tertiary plants was skimmed using a 125 μm filtering assembly. The results suggest that tertiary effluent is not a significant source of microplastics and that these plastic pollutants are effectively removed during the skimming and settling treatment processes. However, at a downstream secondary plant, an average of one micro-particle in every 1.14 thousand liters of final effluent was counted. The majority of microplastics identified in this study had a profile (color, shape, and size) similar to the blue polyethylene particles present in toothpaste formulations. Existing treatment processes were determined to be very effective for removal of microplastic contaminants entering typical municipal WWTPs.

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1. Introduction

Microplastic particles, often smaller than 5 mm, are primarily made of polyethylene, polypropylene and other polymers. As the production and utility of plastic steadily increased over the decades, the occurrence of microplastics in the environment has likewise escalated and these new pollutants are now commonly found in rivers (McCormick et al., 2014; Yonkos et al., 2014), lakes (Eriksen et al., 2013; Free et al., 2014), and shorelines (Thompson et al., 2004; Browne et al., 2011). Microplastics have been shown to have negative impacts on aquatic organisms in our environment. von Moos et al. (2012) reported microplastics were taken up by cells of the blue mussel *Mytilus edulis*, where experimental exposures induced adverse effects on the tissue of the mussel. Cole et al. (2013) found microplastics were ingested by zooplankton, commonly drifting in salt and fresh water. Polybrominated diphenyl ethers (PBDEs), a group of flame retardants widely applied in electronics, were shown to be assimilated from microplastics by

a marine amphipod, *Allorchestes Compressa* (Chua et al., 2014). Because of their hydrophobic nature (Cole et al., 2013), microplastics tend to absorb PBDEs, endocrine-disrupting compounds (EDCs), pharmaceuticals and personal care products (PPCPs), along with other persistent organic pollutants in aqueous media. Concentrations of PBDEs, EDCs and PPCPs, which are detected at parts per trillion levels in many effluent samples (Nelson et al., 2011; Liu and Carr, 2013), could be adsorbed and enriched on the surfaces of microplastic particles (MPPs). These toxic pollutants may eventually enter into an ecosystem's food chain if the contaminated plastic residues are ingested by fish, aquatic invertebrates, and other wildlife (Ivar do Sul and Costa, 2014).

Microplastic particles (MPPs) are present in numerous personal care and cosmetic products such as lotions, soaps, facial and body scrubs and toothpaste. Many of these products are used daily in the United States and around the world. When used, the microplastics in cosmetics are rinsed directly down household drains; these MPPs and other plastic debris end up at municipal wastewater treatment plants (WWTPs). In some published reports (McCormick et al., 2014; Browne et al., 2011), WWTPs were mentioned as potential sources of microplastics in aquatic systems. However, other researchers were unable to confirm a direct link between

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microplastic pollution in rivers and WWTPs (Kein et al., 2015). The debate over whether discharged effluents contribute significantly to the accumulation of microplastics in our environment has widened. Moreover, at this time, it is unknown how these pollutants behave during transport through wastewater treatment facilities. Understanding the fate and transport pathways of microplastics in wastewater treatment processes is of great interest to plant design engineers and environmental scientists alike. New findings could help us to refine and improve existing treatment plant processes to manage or eliminate this new class of pollutants. Here, we report the first complete survey on the presence of microplastic particles in wastewater treatment systems as well as their transport and removal during typical wastewater treatment.

2. Material and methods

2.1. Microplastics

Five sizes of fluorescent polyethylene microbeads, red (10–45 μm), blue (53–63 μm), green (90–106 μm), violet (125–150 μm), and yellow (250–300 μm), were purchased from *Cospheric Innovations in Microtechnology* (Santa Barbara, CA 93160, USA). Additionally, microplastics in a dozen randomly chosen commercial products such as toothpaste, facial washes, body scrubs, and hand soaps were isolated (Fig. S1). In general, ~5 g of these products were placed into 8"-diameter sieves (mesh size: 45 μm), and washed thoroughly with deionized water (DI) to remove gels and other formulation additives. The micro-solids retained on the sieve were then placed on a 10- μm filter paper (S&S filter paper, USA), and washed exhaustively with DI water, and methanol, using a glass vacuum filtration apparatus. Isolated particles were air dried then examined under a microscope (Model 570, 0.7 to 4.2 $\times$ American Optical Corporation, Buffalo, NY 14215, USA) to observe the colors, shapes and sizes.

2.2. Bench-scale studies

To evaluate buoyancy and settling properties of microbeads in mixed liquor (a mixture of raw wastewater and activated sludge) and effluent, 10 mg each of the fluorescent microbeads were mixed together then spiked into 1-L of mixed liquor or effluent. After manually shaking for 2 min, the solution was poured into a 1-L Imhoff cone. The distribution of microbeads was examined, after settling for 10 min.

To simulate the partitioning behaviors of microbeads in raw high-solids influent, ~1.7 g of toilet paper was blended in 300 mL of effluent using a heavy duty blender (Waring® Commercial, Torrington, CT 06790, USA) for 5 min. 5–6 mg blue fluorescent microbeads (53–63 μm) was added to the paper slurry and shaken vigorously. The distribution of microbeads in the settled solution was then observed using a UV hand lamp.

To examine other possible removal modes of microbeads in tertiary plants, a 3"-diameter by 2'-tall bench-scale column was constructed to simulate gravity filters at tertiary plants which typically consist of ~24" anthracite, ~12" sand, ~54" gravel. Our bench filter was assembled to approximate these ratios using the same media: 5"-anthracite, 2"-sand, 2"-small gravel, 3.5"-large gravel, and 5"-stone from column top to bottom (Fig. S4), respectively. The column was first conditioned with DI water, then flushed with 2 L of unfiltered secondary effluent. The flow was maintained at 4 mL/s. One liter of effluent was then spiked with 1 mg each of standard microbead particles (5 mg total), the effluent-bead slurry was then poured into the column. The microbead-spiked mix was filtered and the post-column filtrate collected. A second liter of effluent was used to rinse the spiked microbead vessel. The entire

1-L rinse was then poured into the bench filter to maintain head volume and column flow. 2.2 L of collected filtrate was then re-filtered through a 10- μm filter paper to isolate any microbeads that broke through the bench filter. The column was then back-flushed with DI water and air sparged for ~15 min. 2 L of back-wash water sample was collected.

To study the impact of biofilm on MPPs, two vials containing 20 mL of final effluent were dosed with 5 mL of mixed liquor. One of the vials was autoclaved at 121 °C for 34 min. After cooling, both the sterilized and non-sterilized vials were spiked with MPPs (~1.5 mg) extracted from toothpaste. The vials were capped and tumbled on a Dynabeads® rotary mixer at 20 revolutions per minute (RPM) for >48 h (Dynal Biotech, INC., Lake Success, NY 11042, USA).

2.3. Field sampling

The Sanitation Districts of Los Angeles County, one of the largest wastewater treatment utilities in the United States, operates twelve wastewater treatment facilities (Fig. 1). Four of these facilities have solids handling capabilities. Ten wastewater reclamation plants (WRPs) in this system provide tertiary treatment for approximately 681 million liters per day (MLD) of wastewater. Two sites discharge only secondary effluent. The smaller of the two secondary plants processes 0.3 MLD, while the larger, a combined wastewater/solids handling facility, currently treats 1.06 billion liters per day. Tertiary effluents were collected at seven WRPs (1–7). Secondary effluent was collected from the larger secondary plant (WWTP). All sampling events were conducted between June 2014 and January 2015.

2.4. Sampling methods

Two different sieving methods were used for filtering tertiary effluents at the location shown (Fig. 2). The first method employed a stack of 8"-diameter stainless steel sieve pans with mesh sizes ranging from 400 to 20 μm (Cole-Parmer, Vernon Hills, IL 60061, USA). Whenever possible, existing plumbing and flows from sampling boxes used for plant compliance samples were utilized. At other locations, plumbed final effluent streams were intercepted using PVC line splices. Calibrated effluent flows were filtered through a stack of sieves assembled from coarse to fine (Fig. S5). Flows were set 11.4–22.7 L per minute and were checked daily and adjusted if needed. After calibration, constant flows were maintained for the duration of filtration, in order to accurately determine the volumes of effluent filtered. Volumes were calculated using ($\text{flow rate} \times \text{time}$). Sieve stacks were protected from direct sunlight and fugitive atmospheric debris by wrapping the filtration assemblies in aluminum/plastic shrouds.

The second method used a surface filtering assembly (Figs. S6 and 7) designed for skimming the water surface at the final outfall location. The filtering assembly was deployed at the effluent discharge outfall. Deployment times varied with flows and water quality. Surface skimming was closely monitored by checking flows and filtering performance. If clogging (i.e., any flow restrictions) was indicated, the assembly was immediately retrieved, taken to the lab to recover the residues then cleaned. Surface-skimmed volumes were estimated using [$(\text{skimmer assembly length/weir outfall length}) \times \text{discharged volumes}$].

To investigate the transport of microplastics in each stage of tertiary treatment process (Fig. 2), WRP 1 was chosen because of logistical consideration and proximity to technical resources; and samples were taken from primary stage (influent pumps, skimming troughs located right after influent pumps), secondary stage (aeration tanks, return activated sludge (RAS)), and tertiary stage (secondary wastewater, gravity filters). Sampling at treatment stages of secondary WWTP (grit chamber located in the front of

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