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Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent☆

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ABSTRACT

Municipal wastewater effluent has been proposed as one pathway for microplastics to enter the aquatic environment. Here we present a broad study of municipal wastewater treatment plant effluent as a pathway for microplastic pollution to enter receiving waters. A total of 90 samples were analyzed from 17 different facilities across the United States. Averaging all facilities and sampling dates, 0.05 ± 0.024 microparticles were found per liter of effluent. Though a small value on a per liter basis, even minor municipal wastewater treatment facilities process millions of liters of wastewater each day, yielding daily discharges that ranged from ~50,000 up to nearly 15 million particles. Averaging across the 17 facilities tested, our results indicate that wastewater treatment facilities are releasing over 4 million microparticles per facility per day. Fibers and fragments were found to be the most common type of particle within the effluent; however, some fibers may be derived from non-plastic sources. Considerable inter- and intra-facility variation in discharge concentrations, as well as the relative proportions of particle types, was observed. Statistical analysis suggested facilities serving larger populations discharged more particles. Results did not suggest tertiary filtration treatments were an effective means of reducing discharge. Assuming that fragments and pellets found in the effluent arise from the 'microbeads' found in many cosmetics and personal care products, it is estimated that between 3 and 23 billion (with an average of 13 billion) of these microplastic particles are being released into US waterways every day via municipal wastewater. This estimate can be used to evaluate the contribution of microbeads to microplastic pollution relative to other sources (e.g., plastic litter and debris) and pathways (e.g., stormwater) of discharge.

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1. Introduction

Annual global plastic production now exceeds 300 million tons (Plastics, 2015). As annual production has exponentially increased since the end of World War II, when the infrastructure for the mass production of plastic was established, the presence of plastic as an environmental contaminant has also increased (Plastic Debris, 2006) to the point that it is now considered by the United Nations Environmental Program to be one of the top environmental issues facing our species (Marine Litter, 2005; Eerkes-Medrano et al., 2015). Plastic pollution has now been identified within the worlds' oceans (Cole et al., 2011; Hidalgo-Ruz et al., 2012; Eriksen et al., 2015), seas (Faure et al., 2012; Dubaish and Liebezeit, 2013) and, more recently, freshwater systems (Eriksen et al., 2013; Free et al., 2014; Gasperi et al., 2014; Lechner et al., 2014; McCormick et al., 2014; Morritt et al., 2014; Yonkos et al., 2014; Dris et al., 2015). Plastic pollution may enter these receiving waters through multiple pathways, including stormwater runoff, wind advection and atmospheric fallout, and treated wastewater discharges (Dris et al., 2015).

Microplastics, defined as plastic particles <5 mm in size (Thompson et al., 2009), can be formed from the degradation, largely via mechanical and photo-oxidative pathways (Singh and

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Sharma, 2008), of larger plastic items (Thompson et al., 2004). Such degraded microplastics are considered secondary microplastics. In contrast, a discrete assembly of microparticles are manufactured to be this size and are referred to as primary microplastics (Eerkes-Medrano et al., 2015). Primary microplastics include industrial 'scrubbers' used to blast clean surfaces, plastic powders used in molding, and plastic nanoparticles used in a variety of industrial processes. As some primary microplastics are utilized in consumer products that wash down the drain, there is concern regarding direct emission of microplastics into aquatic environments from municipal wastewater (Fendall and Sewell, 2009; Browne et al., 2011; Eriksen et al., 2013; McCormick et al., 2014). For example, certain personal care products (e.g., facial cleansers, body washes, cosmetics, and toothpastes) contain plastic 'microbeads' as exfoliants or fillers, and, when used as directed, these microbeads are incorporated into municipal wastewater. Given the physical characteristics of these microbeads, it has been suggested that some portion of these particles likely escape the wastewater treatment plant and are discharged into the aquatic environment (Fendall and Sewell, 2009; Eriksen et al., 2013; McCormick et al., 2014).

Municipal wastewater treatment facilities are typically designed based upon a common schematic (Fig. 1), though each facility will differ slightly in the exact configuration of this same basic design. Primary treatment is utilized to remove large debris items with screen mesh sizes of 6 mm or larger. Secondary treatment is used to remove suspended and dissolved organic material and nutrients, largely through the incorporation of microorganisms within large aeration tanks. Flocculates and settling tanks encourage the separation of sewage sludge from the post-processing effluent (hereafter simply 'effluent') prior to any disinfection, polishing or advanced (tertiary) treatment, before being discharged into a nearby waterbody.

A small number of studies now document the passage of plastic particles through individual wastewater treatment plants (Baltic Marine, 2014; Magnusson and Wahlberg, 2014; Carr et al., 2016; Murphy et al., 2016, Table 1). Carr et al. (2016) represents the most significant study to-date, having sampled 0.189 million liters of effluent at each of 8 different southern California facilities. All of these studies indicate that wastewater treatment facilities are quite efficient at removing microplastics from treated wastewater, with calculated removal efficiencies of 95–99%. Murphy et al. (2016) found that primary processing removed 78% of microplastic with subsequent secondary processing removing an additional 20%, while Carr et al. (2016) specifically noted that microplastic removal into wastewater sludge and other solids occurs largely as part of the skimming (which occurs only at some wastewater treatment processing facilities) and settling treatment processes.

Despite the efficacy of removal, all studies still noted microplastic counts within the effluent stream (Table 1). The counts vary considerably between the studies, supporting the need for multiple (preferably high-volume) samples across multiple dates. All of the studies to-date have been extremely limited in their scope, each focused on only a few facilities with limited geographic range, suggesting the need for broader testing to gauge broader regional discharge levels. Differences may also be attributed to variations in sample collection and processing, highlighting a need for a harmonization and standardization of these techniques.

Here we present a broad study of municipal wastewater treatment plant effluent from facilities across the United States, designed in response to some of the limitations described above. Only by conducting a wide-ranging survey of effluent contamination can we determine whether findings drawn from a handful of previously studied facilities can be extended to treated wastewater in general. Participating facilities were selected to cover a broad geographic range, as well as a variety of treatment technologies, in order better characterize the range and variability of microplastic contamination in effluent. This unique dataset also provides an improved means of estimating the discharge of one discrete source of microplastic, microbeads, to receiving waters in the US.

2. Methods

2.1. Study sites

A total of 90 effluent samples were analyzed, from 17 different wastewater treatment facilities of varying size, populations served, advanced filtration types, and at multiple locations across United States (Table 2). For confidentiality reasons, the facilities are identified only with regard to their general location and the major waterbody into which they discharge. Sutton et al. (2016) provide additional information about the San Francisco Bay facilities and samples. The study commenced with three facilities in Fall (September) 2013, with additional facilities added and sampled each academic semester through the Spring (May) 2015 (Table 2). The number of sampling events (i.e., the number of individual samples collected at each facility) varied by facility given our available resources and individual access. As each sample was collected on a different date, each is considered a separate and distinct sampling event (i.e., no replicates), allowing the variability of individual facilities with multiple sampling events to be discussed.

2.2. Sample collection

Effluent was sampled just prior to being discharged from either a sampling port or from an effluent flume using an extraction pump. In both cases the sampled effluent, representing a subset of the total effluent flow through the facility, was filtered through a set of Tyler sieves at a flow rate of 12–18 L per minute for a period of 2–24 h. Exact flow rates (liters per minute) were determined in

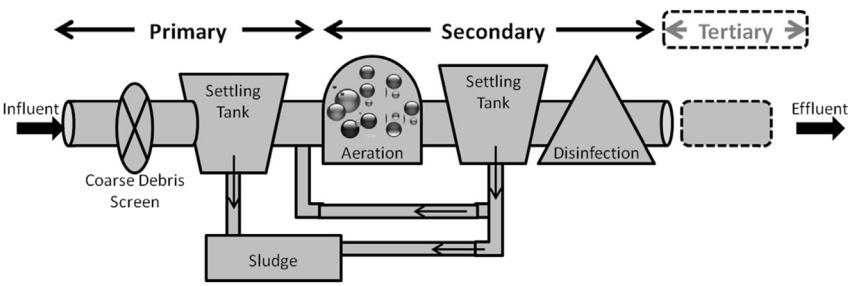

Fig. 1. Schematic of a typical municipal wastewater treatment facility indicating stages of processing. Tertiary treatment is optional and, if present, will vary between facilities. Coarse debris screening (identified here as part of the primary treatment) can also be considered to be preliminary treatment.

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Table 1
Overview of previous related studies for comparison to this work.

	Number of facilities	Volume of effluent sampled (l)	Number of replicates	Finest mesh size (μm)	Sample processing	Sample analysis	Per liter of final effluent		
							Microfibers	Microparticles[a]	Total
Baltic Marine (2014)	1	50	3	20	None	microscope	16.00	7.000	23.00
Magnusson and Wahlberg (2014)	1	1000	4	300	None	microscope	0.004	0.004	0.008
Martin and Eizhvertina (2014)	2	1	2	1.2	WPO[b]	microscope	32.91	0.250	33.16
Carr et al. (2016)	7	1.89×10^5–2.32×10^5	1-2	45	None	Raman	nr[c]	nr[c]	0.001
Murphy et al. (2016)	1	30–50	2-Jan	65	None	FTIR	nr[c]	nr[c]	0.250
This study	17	5.00×10^2–2.10×10^4	1	125	WPO[b]	microscope	0.030	0.017	0.050

[a] As fragments and pellets.
[b] Wet peroxide oxidation.
[c] Not reported.

Table 2
Overview of municipal wastewater treatment facilities included in the quantitative study in order of increasing average 24-h facility flow rate (in millions of liters per day, mlpd) on the date of sampling. Facilities are identified by the major body of water into which they discharge.

Facility ID	Location	Major water body discharge point	Sampling time period	Average 24-h facility flow rate (mlpd)[a]	Population served	Advanced filtration	Combined sewers
SFB1[b]	Northern California	San Francisco Bay	Fall 2014	2.35	56,000,000	n/a	No
LE1	Western New York	Lake Erie	Spring 2014	6.49	3,500	n/a	No
LE2	Western New York	Lake Erie	Fall 2013	13.0	12,000	granular (sand, anthracite coal)	No
LC	Eastern New York	Lake Champlain	Fall 2013 Spring 2014 Fall 2014	15.8	32,000	n/a	Yes
FL1	Central New York	Finger Lakes	Fall 2014	15.9	56,000	n/a	No
LE3	Western New York	Lake Erie	Fall 2013	21.5	15,000	n/a	Yes
SFB2	Northern California	San Francisco Bay	Fall 2014	32.1	140,000	granular (sand, anthracite coal)	No
SFB3	Northern California	San Francisco Bay	Fall 2014	44.7	138,500	granular (sand)	No
SFB4	Northern California	San Francisco Bay	Fall 2014	75.7	220,000	granular (dual media)	No
LE4	Northern Ohio	Lake Erie	Spring 2014	110	103,000	n/a	Yes
SFB5	Northern California	San Francisco Bay	Fall 2014	112	471,000	n/a	No
SFB6	Northern California	San Francisco Bay	Fall 2014	174	650,000	n/a	No
SFB7	Northern California	San Francisco Bay	Fall 2014	190	800,000	n/a	No
FL2	Central New York	Finger Lakes	Fall 2014	213	245,000	biological aerated filter	Yes
LM1	Eastern Wisconsin	Lake Michigan	Spring 2015	307	470,000	n/a	Yes
SFB8	Northern California	San Francisco Bay	Fall 2014	314	1,400,000	granular (gravel, sand, anthracite coal)	No
LM2	Eastern Wisconsin	Lake Michigan	Spring 2015	382	606,000	n/a	No

[a] mlpd = millions of liters per day.
[b] Airport.

triplicate both immediately before and after a sampling event to ensure consistency (with differences generally less than 5%) and then averaged over all measurements. Given the flow rate and the exact length of time over which a sample was collected, the volume of effluent sampled was determined (Table 3). Longer sampling periods afforded a greater, time-integrated, representation of particulate within the effluent flow. However, given the break-through of sludge from the secondary aeration tanks, longer sampling periods necessarily restricted the mesh-size of the filtration sieves utilized. A 0.355 mm-mesh sieve was stacked atop a 0.125 mm-mesh sieve for the shorter (2 h) sampling times, while the 0.355 mm-mesh sieve was used in isolation for the longer sampling periods (during which the finer mesh sieve would fill with sludge and other biological material preventing the passage of the effluent water through the filtration sieve set). For each sample, sieve contents were transferred into separate sample containers and immediately preserved in 70% isopropyl alcohol for later laboratory processing and analysis.

2.3. Sample processing

Consistent with other plastic pollution studies conducted by this laboratory (Free et al., 2014; McCormick et al., 2014; Sutton et al., 2016), labile organic matter within each sieve sample was digested using 30% hydrogen peroxide in the presence of an iron (II) catalyst, otherwise known as Fenton's reagent (Free et al., 2014; Masura et al., 2015). Plastic debris is considered to be resistant to this wet peroxide oxidation (WPO) processing (Masura et al., 2015). After processing, samples were filtered using the 0.125 mm sieve and all remaining particulates were transferred to glass Petri dishes using deionized water (DI) for visual analysis. Using a dissection microscope (Leica EZ4 HD, 40×), all microplastic particles were removed, enumerated and categorized as fragment, pellet, line/fiber, film, or foam (Free et al., 2014). Seven blank samples, in which DI water was stored within sample containers for periods of 1–14 days, were processed concurrently with effluent samples and none were found to have any microplastic particulate, indicating that the risk of sample contamination from the containers, laboratory, or processing was negligible.

As noted by Van Cauwenberghe et al. (2015), a variety of processing and detection methods for plastic pollution within environmental samples have been developed and employed. Here we rely on a processing method developed and supported by the National Oceanic and Atmospheric Administration (NOAA) Marine Debris program, which is stated to have negligible impact upon the most common plastic particulate within a sample (Masura et al., 2015). As some plastic materials may not be resistant to oxidation, it is possible that such materials were degraded through our chemical processing.

While instrumental analysis methods such as FTIR or Raman spectroscopy are necessary for polymeric identification (i.e., polyethylene, polypropylene, polystyrene, etc.), numerous studies have

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Table 3
Overview of microparticles found within effluent by number, type, and mesh-size. Facilities are organized in order of increasing daily discharges of microplastic particles.

Facility ID	No. samples	Volume sampled (l)		Average number of microparticles		Type (%)					Sizes (%)	
		Range	Average	Per liter	Per day	Fragment	Pellet	Fiber	Film	Foam	125–355 μm	>355 μm
LC	22	1600–41,000	13,000	0.004	52,773	65	6	13	13	3	n/a	n/a
LE1	4	3000–18,000	12,500	0.010	64,487	53	0	40	3	4	100	0
LE2[a]	13	980–3000	2100	0.009	101,365	21	5	68	2	5	47	53
FL1	11	1900–4500	4300	0.008	118,706	30	1	58	8	2	51	49
SFB1	1	–	500	0.195	456,691	9	0	90	0	1	53	47
LE3	8	500–3000	1600	0.047	1,237,402	21	5	68	2	5	35	65
SFB2[a]	1	–	1200	0.064	2,045,092	35	0	58	4	4	60	40
LM1	3	18,000–21,000	19,000	0.007	2,251,990	53	2	39	5	1	n/a	n/a
FL2[a]	7	1900–4500	4200	0.019	4,078,889	28	0	68	3	0	40	60
SFB7	1	–	1000	0.022	4,105,857	9	0	91	0	0	55	45
SFB3[a]	1	–	1000	0.092	4,134,574	4	0	94	2	0	45	55
LE4	10	700–1200	1000	0.042	4,769,334	70	4	8	15	4	100	0
LM2	4	12,000–21,000	18,000	0.017	6,055,005	77	2	15	6	0	n/a	n/a
SFB5	1	–	900	0.072	8,086,115	41	0	59	0	0	77	23
SFB4[a]	1	–	500	0.127	9,625,335	18	0	78	3	0	45	55
SFB6	1	–	400	0.071	12,433,886	29	0	57	14	0	46	54
SFB8[a]	1	–	1200	0.047	14,916,649	0	0	100	0	0	44	56
17	90	Averages:	4847	0.050	4,384,362	33	1	59	5	2	57	43

[a] Facilities include advanced/tertiary treatment.

utilized visual-only identification, such as that employed here, for microplastic identification (Bond et al., 2014; Lavers et al., 2014; Devriese et al., 2015; Rochman et al., 2015a; Romeo et al., 2015; Fossia et al., 2016; Hammer et al., 2016; Miranda and Carvalho-Souza, 2016; Nicolau et al., 2016; Peters and Bratton, 2016). Given the source (i.e., wastewater), fibers obtained in this processing would presumably be anthropogenic and derived from textiles, though a portion of fibers observed in wastewater may not be plastic, instead derived from other anthropogenic sources (Remy et al., 2015).

2.4. Statistical analysis

Two statistical analyses were performed to explore the effects of selected predictor variables on particulate output. Predictors included the facility-specific service population size (*Population*; log-transformed to reduce natural skew), presence of tertiary/advanced filtration (*Filter*), and whether the facility services combined sewers (*Sewers*) in the region served, as well as the tertiary filtration X combined sewer interaction. In Analysis 1, total particles in effluent (i.e., all particle types combined) were modeled as a function of the above-noted predictor variables (Table S1). Analysis 2, undertaken to distinguish the influence of fibers from that of fragments given the potential for non-plastic fibers to inflate values, included counts of only these two particle types in the samples as an outcome variable, with a dichotomous predictor variable (*Type*, fragments versus fibers) to distinguish between them (Table S2). This split outcome was modeled as the effects of the above predictors, plus interaction terms with *Type* to indicate the differential effects of the other predictors on fragments versus fibers in facility output.

In both analyses, generalized linear models (GLMs) were employed, with a Poisson link function, a natural choice for "count data" (Cameron and Trivedi, 2013). Output values were multiplied by 10,000 to create integer values necessary for Poisson analysis (thus converting outcome units within the analysis to particles per 10,000 L). This model choice produced roughly normal residuals in both analyses, suggesting that the distribution family was acceptable. Because individual measurements were nested within facilities, two-level multilevel models were specified, to separate the effects of interest, using a random intercept term for individual facilities. Modeling was performed with the lme4 package (Bates et al., 2015) in R version 3.3.0 (R Core Team, 2016). Variance-covariance matrices for both analyses are available in Supplementary material.

3. Results

For each sampling event, given the volume of water sampled, and the total particle counts, the abundance of particles released per liter of effluent was calculated (Table 3). Nine of the 17 facilities included in this study were evaluated on multiple sampling dates and reported values were averaged across those dates (Table 3). Our results indicate that municipal wastewater effluent contains, on average, less than 1 particle per liter of effluent, with values ranging from 0.004 to 0.195 and a 95% confidence interval of 0.050–0.024 (Table 3).

Values for the number of particles released per liter were combined with the 24-h facility flow rate on the date of sampling (Table 2) in order to extrapolate to the daily abundance of particles released by each facility (Table 3). For facilities with multiple sampling dates, values were obtained for each sampling event and reported values were averaged across those dates. Despite the low values of particulate found on a per liter basis, as these facilities process millions of liters of wastewater per day (Table 2), the estimated daily abundance of particles released within the effluent was found to be quite high, on the order of tens of thousands to millions of particles per day (i.e., individual facility values ranging from ~5×10^4 to nearly 1.5×10^7 particles per day). When averaged over the 17 facilities surveyed, we found that, on average, 4.4×10^6 particles were released per facility per day (95% confidence interval: $\pm 2.1 \times 10^6$) (Table 3).

Statistical models of the effects of selected predictor variables on particulate output (Table S3) fit the data well (Analysis 1: marginal $R^2 = 0.16$, conditional $R^2 = 0.62$; Analysis 2: marginal $R^2 = 0.18$, conditional $R^2 = 0.39$; Nakagawa et al., 2013). Statistically significant associations, or lack thereof, are explored in the Discussion.

4. Discussion

4.1. Microparticle characteristics across all facilities

Totaling across all facilities tested, fibers were found to be the

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most common type of particle (59%) followed by fragments (33%), with more minor contributions arising from films (5%), foams (2%), and pellets (1%) (Table 3). Smaller particles (0.125–0.355 mm) were found to be slightly more prevalent than larger particles (>0.355 mm) (57% vs. 43%; Table 3). Larger particles were dominated by fibers (80%), while smaller particles were more evenly distributed between fibers and fragments (Fig. 2). The increased prevalence of fragments within the smallest size classification is consistent with studies characterizing the sizes and topography of microplastics present within personal care products (Fendall and Sewell, 2009; Napper et al., 2015). In general, levels of fibers and fragments in final effluent were within the range reported in previous studies exploring a discrete number of treatment facilities (Baltic Marine, 2014; Magnusson and Wahlberg, 2014; Martin and Eizhvertina, 2014; Carr et al., 2016; Murphy et al., 2016) (Table 1).

There is some uncertainty as to whether all fibers detected in these effluent samples are derived from plastic, given the lack of spectroscopic verification. Browne et al. (2011) found high levels of plastic fibers, verified through spectroscopic analysis, emanating from synthetic materials in both wash water and within the sedimentary record near wastewater effluent outflows. Murphy et al. (2016) also identified significant levels of plastic fibers in effluent using FTIR spectroscopy. However, a recent study of aquatic invertebrates' digestive tracts indicates that some anthropogenic fibers may be cellulose-based rather than plastic (Remy et al., 2015). Carr et al. (2016) found fibrous material in effluent samples to be overwhelmingly derived from biological exudates. However, these samples were not subjected to oxidation, which is expected to remove the bulk of this signal from the samples in this study. Lenz et al. (2015) noted that 75% of fibers identified by visual inspection as plastic in marine samples were later verified as plastic via Raman spectroscopy. Thus while synthetic plastic is likely to be one source of fibers in the studied effluent, we cannot assume all fibers in these samples were plastic.

4.2. Inter- and intra-facility variation in effluent content

While fibers and fragments were consistently found to be the most prevalent particles within the effluent, which type dominated was facility-specific (Table 3). Fibers dominated at two (of the four) Lake Erie (LE2 and LE3) facilities, the Finger Lakes (FL) facilities and all of the San Francisco Bay (SFB) facilities ($n_{total} = 12$), with fragments dominating at the remaining two Lake Erie (LE1 and LE4) facilities, the Lake Champlain (LC) facility and the Lake Michigan (LM) facilities ($n_{total} = 5$). While it may be expected that fragments would be more prevalent in the effluent stream of facilities that also process stormwater (*i.e.*, combined sewer systems) due to run-off, two of the five facilities where fragments dominated do not have combined sewers and two (of the 12) facilities in which fibers dominated do have combined sewer systems. Notably, one thing that the five fragment-dominated facilities do have in common is that they all lack any type of advanced (tertiary) treatment. Nevertheless six, or half, of the fiber-dominated facilities also lack any type of advanced filtering treatment. Thus the composition of particles within an effluent stream would seem to depend upon more than just the presence of advanced (tertiary) treatment or combined sewers.

Nine of the 17 facilities included in this study were evaluated on multiple sampling dates. The variation in effluent particulate discharges across sampling dates for two representative facilities (LC and FL1) is shown in Fig. 3. These facilities were chosen for their differences, as well as their similarities. LC is a slightly smaller facility (than FL1) in terms of population served (32,000 vs. 56,000), but is a combined sewer system that processes stormwater, as well as municipal wastewater. Aside from during storm events (when the flow rate at FC will increase due to the influx of stormwater), both facilities have similar average flow rates (Table 2). Despite their similar service populations and flow rates, the LC facility discharges are dominated by fragments, while FL1 discharges are dominated by fibers (Table 3; Fig. 3).

The dominant particle type at a given facility is generally consistent across sampling dates, though exceptions exist and the overall composition of particles within the effluent streams is highly variable (Fig. 3). A few high flow days at the LC facility (due to a storm event) resulted in an increased abundance of foam particles within the effluent stream (likely due to run-off), but no consistent trend in particle type as a function of flow rate was found (Fig. 4). This highlights the need to sample a facility across multiple sampling dates, as well as sampling significant volumes of wastewater, in order to capture representative effluent discharges.

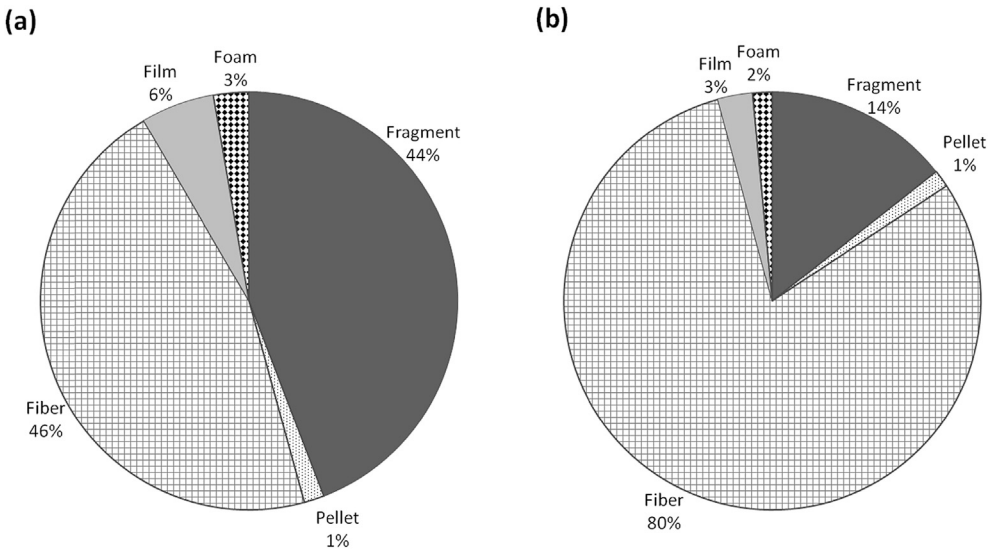

Fig. 2. Percentage of microparticle by type (*Fragments*, dark solid grey; *Fibers*, mesh; *Films*, light solid grey; *Foam*, black diamonds; *Pellets*, dotted) for each size classification, 0.125–0.355 mm (a) and >0.355 mm (b).

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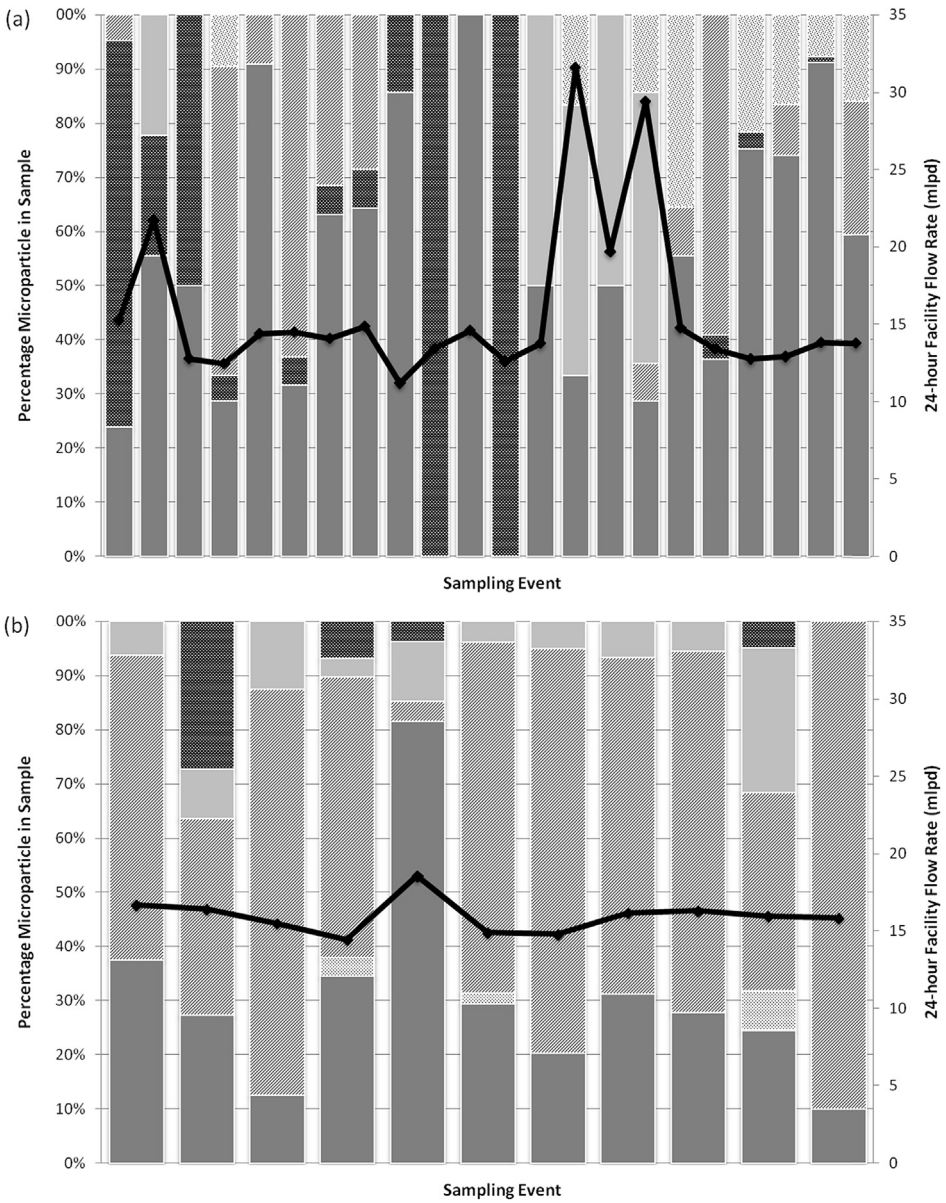

Fig. 3. Variations in microparticle compositions at two representative facilities, LC (a) and FL1 (b), across multiple sampling events. Percentage of microparticles by type (*Fragments*, dark solid grey; *Fibers*, diagonals; *Films*, light solid grey; *Foam*, confetti; *Pellets*, dotted) for each sample. Solid black line indicates facility flow rate on date of sampling.

4.3. Influence of facility characteristics on microparticle discharge

An analysis of effluent particle concentrations relative to variables such as population served, use of tertiary filtration, contributions of stormwater via combined sewer systems, and facility daily flow rate is warranted. Modeling results suggested that (log-normalized) *Population* had a statistically significant positive association with total particles ($B = 0.25$, $z = 2.08$, $p < 0.05$; Table S3). *Population* was also positively associated with number of fibers but unassociated with number of fragments, after controlling for other effects.

Our data do not show a clear correlation between advanced (tertiary) filtration and reduced microparticle discharges. Instead, the model suggested that the presence of filtration (*Filter*) had a marginally significant positive association with overall particle discharge, after controlling for other predictors ($p < 0.1$). *Filter* had no association with number of fragments, but was associated with higher numbers of fibers in output when other effects were controlled. The association between *Filter* and fibers seems to be responsible for the increased number of total particles in facilities with tertiary filtration. It is possible that the filtration media could promote microbial growth or accumulate microbial residues that could be mistaken for fibers, a potential explanation for this relationship. Sample processing using oxidation, as conducted in this study, is generally found to be effective in removing many such residues from marine samples (Masura et al., 2015), but has not been specifically evaluated for effluent samples. Combined sewers were associated with increased numbers of fragments in output, but not fibers.

Six of the 17 facilities included in this study have some type of advanced filtration (Table 2), but both on a per liter basis (range: 0.009–0.127; average = 0.060 ± 0.033) and in terms of total daily discharges (range: 1×10^5 to ~1.5×10^7; average = $5.8 \times 10^6 \pm 4.0 \times 10^6$), their releases were consistent

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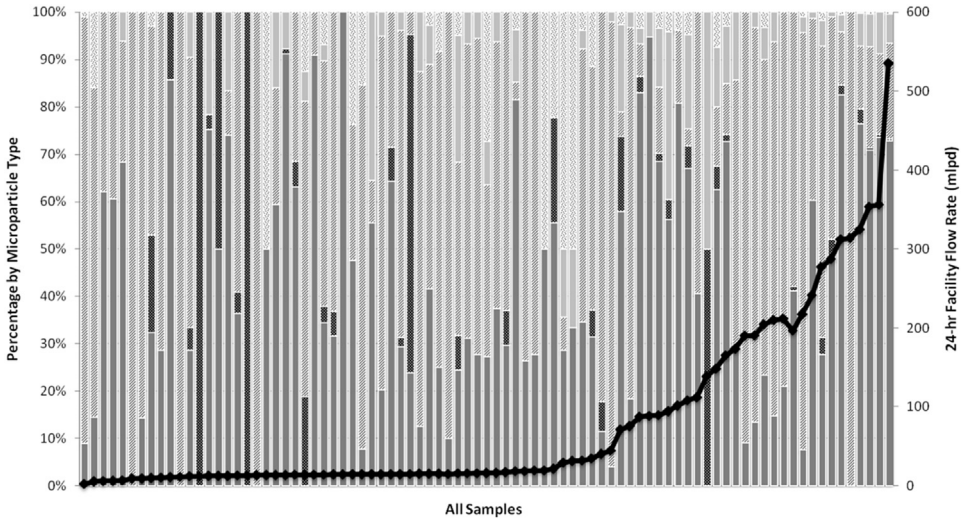

Fig. 4. Variations in microplastic compositions for all 90 sampling events across all 17 facilities. Percentage of microparticle by type (*Fragments*, dark solid grey; *Fibers*, diagonals; *Films*, light solid grey; *Foam*, confetti; *Pellets*, dotted) for each sample. Solid black line indicates facility flow rate (in millions of liters per day, mlpd) on date of sampling.

with those of the other facilities. At only one facility (LE2) were we able to sample both before and after the advanced filtration processing. These data (collected on 4 different sampling dates) did show an overall 15% reduction in particles within the effluent stream at this facility (data not shown), but given the limited nature of these data, and in light of the consistency of the findings of our six advanced treatment facilities relative to the other eleven facilities (that do not have advanced treatments), the potential of advanced filtration to increase removal of microplastic from the effluent stream is uncertain. A qualitative, presence/absence study of microbeads (excluding fibers) in effluent from 34 wastewater treatment facilities in New York found tertiary treatments such as membrane microfiltration, continuous backwash upflow dual sand (CBUDS) microfiltration, and rapid sand filters, did not guarantee microbeads would be absent from effluent (Discharging Microbeads, 2015). These findings are also consistent with a more recent study by Carr et al. (2016), which found that effluent filters (employed in tertiary processing) had a minimal impact upon microplastic particle removal. Thus existing studies indicate that advanced filtration may not be effective with regard to microplastic removal. However, it must be noted that a full examination of the impacts of both of these variables would necessarily involve comparisons of paired influent and effluent samples at facilities, rather than a reliance on effluent data alone.

Interestingly, facilities with higher 24-h flow rates (on the date of sampling) appeared to have a lower abundance of particles per liter (Fig. 5; filled circles). This could be attributed to dilution, as only a subset of the total effluent flow was extracted for filtering. Since we did not sample the entire effluent flow, nor were our filters immersed within the effluent flow, it is possible that the turbulence associated with a higher flow rate resulted in increased mixing, causing resident microparticles to be more evenly mixed throughout the effluent stream. This increased dilution of the microparticles throughout the effluent stream could have diminished the prevalence of particulate within those samples. Nevertheless, owing to higher quantities of water processed at these larger facilities, they were found to release higher total counts of particles on a daily basis (Fig. 5; 'x' markers).

Overall, our data suggest that the abundance and types of particles within municipal effluent streams may be dependent upon a complex variety of factors such as population served, adjacent surrounding land use (which would influence run-off that could be combined with wastewater), combined sewer systems, flow rate, advanced (tertiary) filtration, and sources (residential versus commercial versus industrial).

4.4. Estimated US discharge of microbeads via wastewater effluent

Rochman et al. (2015b) utilized studies available in 2015 (Magnusson and Wahlberg, 2014; Martin and Eizhvertina, 2014) to estimate the abundance of microbeads being released into US waterways via municipal wastewater effluent. This one particular source and pathway of microplastic pollution should be mitigated in time as the US 'Microbead-Free Waters Act' was signed into law in December 28, 2015, banning the manufacture of microbeads for personal care products by July 1, 2017 and the sale of such products by July 1, 2018. Nevertheless, this prior work provides a point of comparison for this present study, and both can provide valuable background data prior to the implementation of this regulation.

For these estimates, Rochman et al. (2015b) assumed all 'microparticles' (specifically excluding fibers) to be 'microbeads' derived from consumer products, such as personal care products. For comparison purposes here, we assume that microparticles we categorized as fragments and pellets are 'microbeads.' This assumption is consistent with the characterization (i.e., the topography and other physical traits) of these microparticles by other studies (Fendall and Sewell, 2009; Napper et al., 2015). Rochman et al. (2015b) estimated that 0.100 microbeads were released per liter of effluent, which is about an order of magnitude higher than our average value of 0.017 microbeads per liter of effluent (Table 2).

In order to extrapolate numbers (on a per liter basis) to total US microbead discharges per day, the quantity of wastewater being processed daily must be known. Rochman et al. (2015b) used a value of 80 billion liters of wastewater based upon US Census (2000) data. We found two alternative, more recent sources for estimates on the number of municipal wastewater treatment facilities and the average amount of wastewater being processed daily (Maupin et al., 2014; EPA DMR, 2014). Every five years, the US Geological Survey consults with local, state and federal agencies in order to compile and publish a report on US water use (Maupin et al., 2014). While they do not directly report on the quantity of wastewater processed, their most recent (2010) report does state that 42 billion gallons (or ~159 billion liters) of water are

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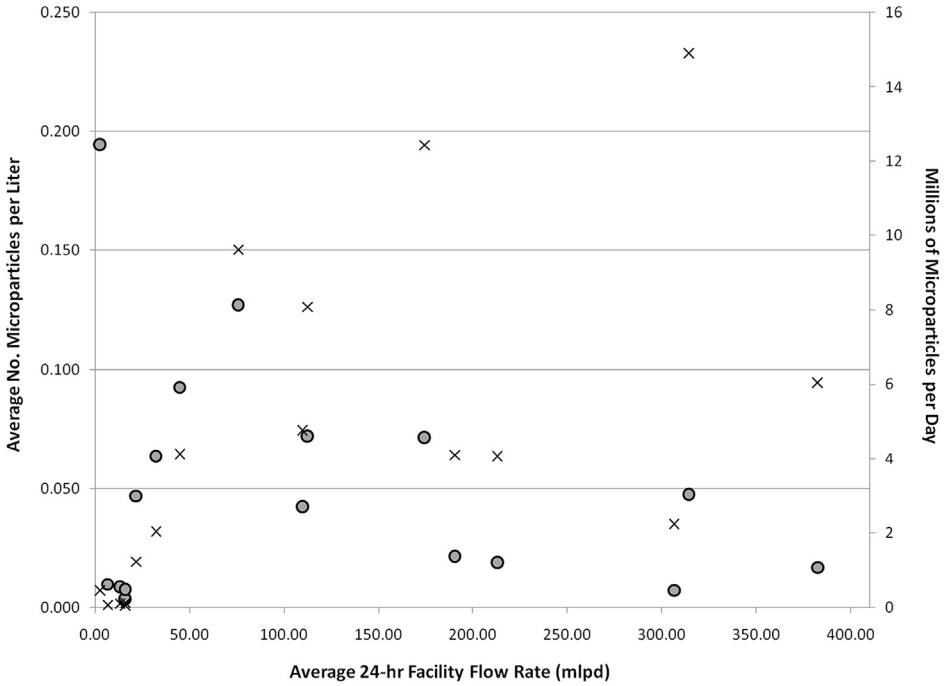

Fig. 5. Average number of microparticles per liter (filled circles) and per day (in millions; 'x' markers) as a function of the 24-h facility flow rate in millions of liters per day (mlpd).

Table 4
Estimates on the number of municipal wastewater treatment facilities, volume of wastewater treated per day, and billions of microbeads being discharged from these facilities based upon this study in comparison to a previous estimation (Rochman et al., 2015b).

This study:	0.017 "microbeads" (as fragments and pellets) per liter 1,490,683 "microbeads" (as fragments and pellets) per facility per day						
	Maupin et al., 2014	EPA DMR (2014)	Rochman et al. (2015b)	Estimations (billion microbeads per day)			
				Maupin et al., 2014	EPA DMR (2014)	Average	Rochman et al. (2015b)
No. of facilities	14,780	15,648		22	23	13	8
Total flow (mlpd)	159,022	206,834	80,000	3	4		

withdrawn daily for public use and that there are 14,780 municipal wastewater treatment facilities in the US (Maupin et al., 2014). Here we assume that the public supply water withdrawals are equivalent to the amount of wastewater processed (Table 4). As an additional source, we utilized the US EPA Discharge Monitoring Report (DMR) Pollutant Loading Tool (available on-line at: http://cfpub.epa.gov/dmr/) to obtain reported 2014 wastewater flows for all major and minor publically-owned treatment works. These data (EPA DMR, 2014) indicate that in 2014 there were over 15,000 municipal wastewater treatment facilities processing nearly 55 billion gallons (or ~207 billion liters) of wastewater (Table 4). Thus our analysis of available data indicates Rochman et al. (2015b) underestimated the quantities of wastewater being processed in the US and that a more accurate estimate is at least double their value (of 80 billion liters per day), or 159–207 billion liters per day (Table 4).

Using our measurements of microbead discharge and updated estimates for the amount of wastewater processed, we estimate that US municipal wastewater treatment facilities are releasing between 3 and 23 billion microbeads per day. The variability in these values arises from uncertainties in the estimates with regard to the number of municipal wastewater treatment facilities and the amount of wastewater they process on average each day (Table 4). Despite this variability, our average estimated discharge of 13 billion microbeads released into US waterways each day from municipal wastewater treatment facilities corresponds well with the 8 billion per day estimated by Rochman et al. (2015b).

5. Conclusion

In summary, our analysis of 90 samples taken from 17 different facilities across the US indicates that municipal wastewater treatment facilities were widely found to represent one pathway for microplastics to enter the aquatic environment. Given the prevalence of microbeads within personal care products (Fendall and Sewell, 2009; Napper et al., 2015), the prevalence of synthetic clothing and fabrics that release microfibers when washed (Browne et al., 2011), as well as the fact that wastewater treatment facilities were not designed to remove emerging contaminants like microplastics, this finding is consistent with expectations (Eriksen et al., 2013; McCormick et al., 2014; Rochman et al., 2015b) and the limited number of facilities for which data were previously available (Baltic Marine, 2014; Magnusson and Wahlberg, 2014; Martin and Eizhvertina, 2014; Carr et al., 2016; Murphy et al., 2016).

These findings must be evaluated in light of a number of considerations. First, the significance of the wastewater pathway for microplastic contamination relative to other pathways, like stormwater run-off, wind-blown debris, and *in situ* degradation of larger plastic items, is unknown and will require additional study. Microbeads and microfibers may prove to be a relatively small portion of the microplastic found in oceans, seas, and freshwater

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systems. Second, thorough study of the impacts of advanced (tertiary) treatment technologies and combined sewer systems is needed. Such studies require characterization of paired influent and effluent samples at minimum. A study in which influent at multiple facilities with various types of tertiary treatments is spiked with microplastic particles of varying sizes and shapes (i.e., beads versus fibers) could prove extremely valuable in examining the influence of such treatment on microplastic release. Third, the finding that facilities with higher flow rates tended to have lower particle concentrations suggests a potential for bias in the sample collection method. Additional testing using a different sample collection strategy, preferably testing across the entire effluent flow, should be conducted to address this possible bias. Finally, studies to specifically evaluate the range of compositions of the fibers commonly observed in effluent is essential to determining the relative contributions of plastic fibers versus anthropogenic, non-plastic or microbially-derived fibers in treated wastewater. Only with this information would it be possible to estimate the discharge of plastic fibers to US waterways.

Author contributions

The manuscript was written through contributions of co-authors SAM, DG, RS and DLR. All authors have given approval to the final version of the manuscript.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2016.08.056.

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