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Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada

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ABSTRACT

Municipal wastewater treatment plants (WWTPs) are conduits through which microplastics (MPs) are released into aquatic environments. However, the technical challenges in working with wastewater sample matrices have precluded reliable particle count budget calculations. We applied newly-adapted methods for MP collection and analysis to a study of a major WWTP serving a population of 1.3 million people near Vancouver, Canada. Suspected MP particles, including fibres, were counted and categorized using light microscopy in influent, primary effluent, secondary effluent, primary sludge and secondary sludge. Fourier Transform Infrared Spectroscopy (FT-IR) confirmed that just 32.4% of the suspected MPs were plastic polymers. Using FT-IR corrected data, we estimate that 1.76 ± 0.31 trillion MPs enter the WWTP annually, with 1.28 ± 0.54 trillion MPs settling into primary sludge, 0.36 ± 0.22 into secondary sludge, and 0.03 ± 0.01 trillion MPs released into the receiving environment. This corresponds to a retention of microplastics of up to 99% in the WWTP.

1. Introduction

There is increasing concern about the scale and nature of microplastic (MP) pollution in the world's aquatic environments (Gall and Thompson, 2015), leading to fundamental questions about their source. Municipal wastewater treatment plants (WWTPs) are an important source of MPs to aquatic environments as even low concentrations of MPs (e.g. < 0.1 MP/L) in effluent may contribute significantly to MP pollution due to the large volumes that are continuously discharged by WWTPs (Browne et al., 2011; Kershaw and Leslie, 2015; Mason et al., 2016; Mathalon and Hill, 2014; Talvitie et al., 2015, 2017; Ziajahromi et al., 2017).

Microplastic concentrations in effluent reflect diurnal and seasonal variations and further depend on operational factors at the WWTP such as population served and treatment processes (Lares et al., 2018; Mason et al., 2016; Michielssen et al., 2016; Ziajahromi et al., 2017). A recent study from 17 WWTPs in the USA estimated that between 50,000 and 15 million MPs per day were discharged (Mason et al., 2016), the majority being fibres and fragments (Mason et al., 2016; Michielssen et al., 2016). While MP fibres in wastewater can originate in part from the washing of synthetic clothing in household laundry (Browne et al., 2011), other MP particles include the abrasives from some cosmetic products and toothpastes, as well as non-point source contamination in urban stormwater (Talvitie et al., 2017; Ziajahromi et al., 2017).

A few studies have indicated that primary and secondary wastewater treatment remove the majority of MPs (Carr et al., 2016; Lares et al., 2018; Murphy et al., 2016; Talvitie et al., 2015, 2017). However, due to the limited number of studies that have quantified MPs in both wastewater and sludge in the same WWTP, it is not clear whether this loss can be attributed to the breakdown of particles into particle sizes that evade detection by current methods or to separation into the solids stream (Michielssen et al., 2016).

The diversity in size, structure, colour and polymeric composition of MPs in samples renders it challenging to isolate, enumerate and characterize MPs in organic matter-rich wastewater matrices. Method development remains one of the key challenges for researchers who are seeking to characterize MP transport and fate within WWTPs. Current methods often fail to process untreated influent wastewater because the solids content rapidly clogs sampling devices. While most past studies have used various screens and sieves ranging from 20 µm to 6000 µm (Table 1; Lares et al., 2018; Murphy et al., 2016; Simon et al., 2018) to pass influent samples (volumes ranging from 0.8 to 30 L) before processing, there is, to our knowledge, only one study that has developed a method to process bulk influent wastewater samples, wherein solid and

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Table 1
Sampling and analysis methodologies used for the study of microplastics (MP) and other Small Anthropogenic Litter (SAL) in municipal wastewater vary widely. The majority of studies use variations of stacked sieve devices for the extraction of MP. Sieving may impact the ability to retain MP fibres, such that we adapted our recently published oil extraction protocol (OEP) to omit sieving in influent samples (Crichton et al., 2016). To be able to process samples with the OEP, sample volumes need to be 1 L or less. However, MP counts in primary and secondary effluent are low, and sample volumes needed to be higher to stay within the limit of detection. Sieving was therefore a requisite to condense sample volumes.

Reference	Sample quantity (L)				Sieved	Mesh size (μm)	Sample processing	Analysis
	Influent	Primary	Secondary	Tertiary				
Magnusson and Norén, 2014	2	ND	ND	ND	Yes	300	Water filtered onto plankton net	Microscopy and melt test
Tagg et al., 2015	0.25	ND	ND	ND	No	0.2	Centrifugation followed by H_2O_2 digest and filtration	FT-IR
Talvitie et al., 2015	0.3	1–20	1–20	2–285	Yes	20–200	Pumped through custom stacked sieve device	Microscopy
Talvite et al., 2017	0.1	1–14.5	1–14.5	2–14.5	Yes	20–300	Pumped through custom stacked sieve device	Microscopy and ft-ir
Carr et al., 2016	0.1 L	Unknown	$1.89 * 10^5$–$2.23 * 10^5$		Yes	45–180	Manual inspection for influent, stacked sieve and surface sieve for secondary and tertiary effluent	Magnifying glass and microscopy
Michielssen et al., 2016	1–2 L	10–20	10–20	34–38	Yes	20	Poured through stacked sieves followed by manual inspection	Microscopy
Murphy et al., 2016	30	30	50	NA	Yes	65	Sieving followed by vacuum filtration onto membrane filter	Microscopy and FT-IR
Sutton et al., 2016	ND	ND	$1.9 * 10^4$–$25.8 * 10^4$		Yes	125–355	Sieving and wet peroxide oxidation	Microscopy
Mason et al., 2016	ND	ND	$5.00 * 10^2$–$2.10 * 10^4$		Yes	125–355	Sieving and wet peroxide oxidation	Microscopy
Mintenig et al., 2017	ND	ND	390–1000 L		No	20	Filtration onto cartridge filters followed by enzymatic and H_2O_2 digestion, and density separation using $ZnCl_2$	Microscopy and FT-IR
Ziajahromi et al., 2017	ND	3 L–100 L	27 L–150 L	200 L	Yes	25–500	Custom stacked sieve device	Microscopy and FT-IR
Lares et al., 2018	0.8 - 3 L	4–30 L		NA	Yes	250–500	Stacked sieve device, followed by wet peroxide oxidation and cellulase treatment	Microscopy, FT-IR and Raman
Simon et al., 2018	1 L	ND	ND	ND	Yes	2000–500	Sieving followed by cellulase treatment and wet peroxide oxidation	FT-IR
Our study	1 L	30 L	30 L	ND	Influent: No; Primary & secondary: Yes	1–65	Oil extraction followed by H_2O_2 digest and vacuum filtration	Microscopy and FT-IR

liquid fractions were separated using centrifugation (Tagg et al., 2015).

Likewise, only a small number of studies have successfully extracted MPs from wastewater sludge samples. In one study, researchers employed a combination of sieving, elutriation and density separation on 30 g sludge samples with a minimum size limit of 250 μm (Mahon et al., 2017). To estimate size fractions down to 45 μm, the authors collected subsamples from the fraction that washed through the sieves (Mahon et al., 2017). In another study, 25 g sludge samples were passed through a 300 μm plankton net followed by microscopic evaluation and FT-IR (Magnusson and Norén, 2014). Another study processed 5 g sludge samples by digestion (Yang et al., 2011). While this harsh treatment removed most of the sludge matrix, it also led to the potential loss of several polymers including polyethylene (PE) and polypropylene (PP) due to melting (Carr et al., 2016).

Similar problems were reported by Mintenig et al. (2017) who treated drained sewage sludge with sodium hydroxide (NaOH) solution (Mintenig et al., 2017), a treatment that has been reported to affect integrity of polyester and PE (Cole et al., 2011). Another study dried 2.5 g combined sludge (primary and secondary) at 50 °C and dissected the dried fraction manually using a dissecting microscope (Murphy et al., 2016). While this method avoids any harsh treatment for the removal of organics, it is very subjective due to the manual handling and identification of potential MPs. The same method was applied by Lares et al. (2018) on samples collected from activated sludge, membrane bioreactor sludge and digested sludge. Finally, Talvitie et al. (2017) analyzed 1 g wet sludge and 0.2 g dry sludge by first diluting the sample with 1 L tap water and subsequent filtration over a custom made stacked sieve device with size fractions between 300 and 20 μm (Talvitie et al., 2017). This method also avoids treatment with harsh chemicals but the sample size is small compared to others.

The objective of this study was to carry out a pilot study of MPs in a major regional WWTP serving Vancouver, Canada, through the development of new approaches to MP isolation and quantification. In order to achieve this, we 1) obtained samples at major points within the WWTP, including primary influent, primary effluent, secondary effluent, primary sludge and secondary sludge; 2) developed or adapted methods to separate, clean, count and characterize MPs in WWTP samples; 3) performed FT-IR analysis on a subset of MPs to confirm their polymeric identities; and 4) estimated an annual particle count budget for MPs in this WWTP.

2. Methods

2.1. Sample collection

All samples for this pilot study were collected in the processing stream of a major secondary WWTP near Vancouver, British Columbia, Canada. This WWTP serves a population of 1.3 million residents and treats approximately 180,044 ML/year of municipal wastewater and stormwater from combined sewers, annually. The treatment process begins by passing the water through vertical screening bars (13 mm) for the removal of large debris. This is followed by solids removal via primary clarification and biological degradation via trickling filters and solids contact tanks. Secondary clarifiers provide additional solids removal prior to chlorination (seasonal only) and discharge to the Fraser River upstream from the marine waters of the Strait of Georgia.

Wastewater samples (influent, primary effluent, and secondary effluent) were collected on September 16, September 29, and October 28, 2016 and sludge samples (primary sludge and secondary sludge) were collected on September 14, September 27, and October 11, 2016.

All samples were collected and processed in duplicates alongside procedural and background blanks. Samples were collected into clean glass jars that had previously been rinsed three times with distilled water that was pre-filtered over a 1 μm borosilicate membrane filter (binder-free; Sartorius, Bohemia, USA). Water was collected using a Teledyne ISCO Glacier Portable Water Sampler with PTFE lined tubing

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