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The potential for a plastic recycling facility to release microplastic pollution and possible filtration remediation effectiveness

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ABSTRACT

With current plastic production and the growing problem of global plastic pollution, an increase and improvement in plastic recycling is needed. There is limited knowledge or assessment of microplastic pollution from point sources such as plastic recycling facilities globally. This pilot study investigates microplastic pollution from a mixed plastics recycling facility in the UK to advance current quantitative understanding of microplastic (MP) pollution release from a plastic recycling facility to receiving waters. Raw recycling wash water were estimate to contain microplastic counts between 5.97 10^6 – 1.12 × 10^8 MP m^{-3} (following fluorescence microscopy analysis). The microplastic pollution mitigation (filtration installed) was found to remove the majority of microplastics >5μm, with high removal efficiencies for microplastics >40μm. Microplastics <5μm were generally not removed by the filtration and subsequently discharged, with 59-1184 tonnes potentially discharged annually. It is recommended that additional filtration to remove the smaller microplastics prior to wash discharge is incorporated in the wash water management. Evidence of microplastic wash water pollution suggest it may be important to integrate microplastics into water quality regulations. Further studies should be conducted to increase knowledge of microplastic pollution from plastic recycling processes.

1. Introduction

The existence of microplastics (MP) – plastic particles ranging 1μm to 5mm – is increasingly being seen throughout all ecosystems in the world. Research has shown that MPs travel in water systems from urban areas to freshwater courses and out to sea, as well as atmospheric systems transporting MPs from terrestrial systems to oceans and the ocean serving as a method of MP transportation around the globe (Su et al., 2022). MPs can comprise both primary and secondary particles; primary describing those manufactured intentionally, with secondary describing those broken down from larger MPs or macroplastics.

MPs can adsorb, transport and later release, environmentally and ecosystem detrimental contaminants such as organic pollutants and heavy metals. Alongside these adsorbed contaminants, MPs themselves have detrimental, and often fatal, effects on organisms of all sizes (Ruairuen et al., 2022; Joyce and Falkenberg, 2023; Klasios et al., 2021). These may range from the lethal impacts of the ingestion of MPs sized 1.25 μm by a keystone species of zooplankton (Lyu et al., 2021) to the bioaccumulation of MPs in larger mammals through biomagnification throughout food chains (Carlin et al., 2020; Rochman et al., 2019). Many types of MPs have also been detected in human blood, including polyethylene terephthalate (PET), polyethylene (PE), polystyrene (PS) and polypropylene (PP) (Leslie et al., 2022).

Plastic recycling facilities (PRFs) use processes whereby plastics are separated by type, broken down and granulated, and then pelletised for re-processing. The use of mechanical friction, abrasion, or equivalent methods to breakdown the plastics within these recycling processes may increase the MP concentration in the wash water volumes often used and subsequently discharged in these recycling processes (Altieri et al., 2021). The release of MP pollution in wash water discharge from plastic recycling facilities is significantly understudied and there is a research and knowledge gap in understanding how plastic recycling facilities may contribute to the environmental plastic pollution problem. Although recycling is low in priority to reaching a circular economy, there are some situations in which recycling is an essential method of waste reduction. For example, the recent global COVID pandemic has seen substantial increase in the volume of medical plastic waste produced, for which the standard waste treatment is either incineration or landfill. Global plastic production increased from 359 to 367Mt of global virgin plastic production between 2018 and 2020 (PlasticsEurope, 2021). Increased

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recycling of this plastic waste, and particularly single use plastics, is required to reduce plastic waste being sent to landfill and will be a part of the waste management solution for plastic pollution around the globe.

Although there is increasing research on the effluence of MPs from point sources such as wastewater treatment plants (WWTPs), little is known on the creation of MP pollution by plastic recycling processes. No legislation or standard exists within the UK - the country of study - to control the release of MPs into the environment from controlled activities. This study analyses the MP pollution generated by a case study PRF in the UK and determines the effectiveness of a filtration measure employed at the PRF, recommending technical and policy impact responses. The purpose of this study is threefold: 1) to identify if this PRF discharges MPs in wash water; 2) to consider the effectiveness of the filtration system in mitigating overall and small particle MP quantities in PRF wash water; and 3) to highlight PRF as a potential MP source of plastic pollution to the environment that may need further future investigation, monitoring and potential inclusion in future regulatory support.

2. Materials and methods

2.1. Study area and sample collection

Samples were collected from a state-of-the-art PRF in the UK accepting 22,680 tonnes of mixed plastic waste per year (Fig. 1, Supplementary Note 1). Samples were collected from the four wash water discharge flow paths within the PRF as they represented the MP discharge from this site. Wash water is assumed to be a mixed, homogeneous discharge but it is noted this is not monitored. A pre-filtration sample set was collected during the early activity of the PRF (during the first year of operation) prior to the installation of water filtration to the processes, with a post filtration sample set collected once the full filtration system was in operation. Wash water is used throughout the process, and it is discharged at each of the four sample sites (final outflow is a combined flow of all four discharge points). The filtration measures implemented are 50μm particle filters (mesh sieves forming part of a liquid/solid separator, supplementary note S2) placed at the 3 of the four outlet locations (sample point 1, 2 and 3) manual skimming from tank surfaces (supplementary note S1 provides PRF water flow). Comparison between these sample sets shows the effects of the filtration on MP quantity and particle size distribution.

At each process and associated discharge point, tanks collect excess water from the respective recycling process. From these tanks discharge points, three (triplicate) 2L water samples were taken (Fig. 1, points denoted in yellow). Collection and analysis of triplicate samples was required to ensure statistical and scientific validity of the results in line with standard scientific practice. However, it is acknowledged that if time and analytical resource were not a (global) significant microplastic analysis constraint then a great number of replicate samples could have been taken to further minimise result uncertainties. It is acknowledged that ideally the tanks would have been totally homogenised prior to sampling but due to tank size and health and safety constraints this was not possible. During the collection of post-filtration samples, the water samples were taken from the volume of water after the filtration measures, the filtered flow. All samples were collected in multiple 1L clear glass bottles which had been rinsed, heated in a furnace at 450°C for 6 hours, then again rinsed three times internally and once outside with Milli-Q (Guo et al., 2022; Hidayaturrahman and Lee, 2019; Lares et al., 2018; Leslie et al., 2017; Murphy et al., 2016). These bottles were wrapped in aluminium foil, with a foil lining inside the plastic lid to prevent plastic contamination internally (Bayo, López-Castellanos and Olmos, 2020; Funck et al., 2021; Lares et al., 2018). During collection of the water samples, the containers were filled but not overflowing, minimising any potential spillage from the glass containers during travel and associated contamination and ensuring no loss of floating MP particles from overflowing. Samples were stored in a dark fridge (4°C) until analysis (following sample storage protocols used in Bayo, Olmos and López-Castellanos, 2022; Lares et al., 2018; Allen et al., 2019). Field blanks, processed as full process analytical blanks, were collected during sampling campaigns. Blanks were collected using the same sterilised glass jars, left open for the full sampling duration, with 200ml of Milli Q water to retain any atmospherically deposited particles.

2.2. Microplastic Extraction from Water Samples

Each sample was filtered onto a 47mm glass fibre filters with a pore size of 1.6μm (Whatman GF/A). The filtered volume was dependent upon the MP concentration in each sample and thus varied between sample points to ensure the deposited particles on the filter were not too dense to inhibit analysis by Nile Red under fluorescent microscope (Supplementary Information, Table S1). The volume required was determined by shaking the bottle for 20 seconds to give an even distribution of particles throughout the sample, followed by the measurement of 200ml of sample into a large glass beaker to ensure an adequate representation of the sample (Table S1). For certain samples with large MP concentrations and thus small volumes of sample used, a volume of Milli-Q was also used to dilute the sample for easier distribution of particles onto the filter (Fig. S5).

The sample was poured in a swirling motion to promote an even distribution of particles on the filter and vacuum filtered through the filter, using a system comprising a stainless-steel filter holder attached by a silicone bung to a conical flask connected by a hose to a vacuum pump. The measuring beaker and inside walls of the filter holder were triple flushed with approximately 250ml Milli-Q to ensure all particles were rinsed onto the filter, which was also vacuum filtered through.

Following this, 40ml of 30% w/w hydrogen peroxide (H202) was applied to the filter and left to digest the organic content on the filter for at minimum 48 hours at room temperature, with the filter holder lid remaining intact to minimise the risk of contamination from deposition particles (Peeken et al., 2018; Leslie et al., 2022; Leslie et al., 2017; Okoffo et al., 2019; Gies et al., 2018). All handling of H2O2 was performed under a fume hood (Ziajahromi et al., 2017). Upon completion of this digestion, any remaining H202 was vacuum filtered through followed by 200ml of Milli-Q to remove any residual H202 from the filtered particles. All blanks followed the same procedure.

The filter was carefully removed from the holder using sterilised stainless-steel tweezers and placed in a sterilised 50mm diameter aluminium tin. Nile Red dye was then (glass) pipetted onto the filter, ensuring the dye was absorbed over the entire filter surface. This tin was heated at 30°C for approximately 2 hours, until the filter was dry, with the tin lid left loosely on to allow the escape of vapour but also to prevent any deposition and subsequent contamination onto the filter. Once dry, the lid was screwed on tightly and placed in the fridge for at least an hour, allowing the MPs and Nile Red to cool sufficiently before microscopy.

The Nile Red fluorescence microscopy method was used to analyse the size and shape of MP particles in the water samples following the methodology published and used in Shim et al., 2016; Erni-Cassola et al., 2017; Maes et al., 2017, Nel et al. 2021 and Kukkola et al. 2023. This method has been found to be appropriate for simple quantification of microplastics (but does not define polymer type) (Erni-Cassola et al., 2017; Shim et al., 2016). The Nile Red dye was prepared by the dilution of Nile Red into 99% methanol to a concentration of 1μg/ml and filtered (Whatman QMA, 2.2μm pore) to remove crystals or contamination within the solution which may be confused with sample MP particles during the Nile Red analysis (Erni-Cassola et al., 2017; Allen et al., 2019).

2.3. Microplastic quantification

A Nikon LV100ND fluorescent microscope with a green filter (emission/excitation 460/525nm) was used to image a specified, representative area of each filter to a magnification of either x10 or x5, and where

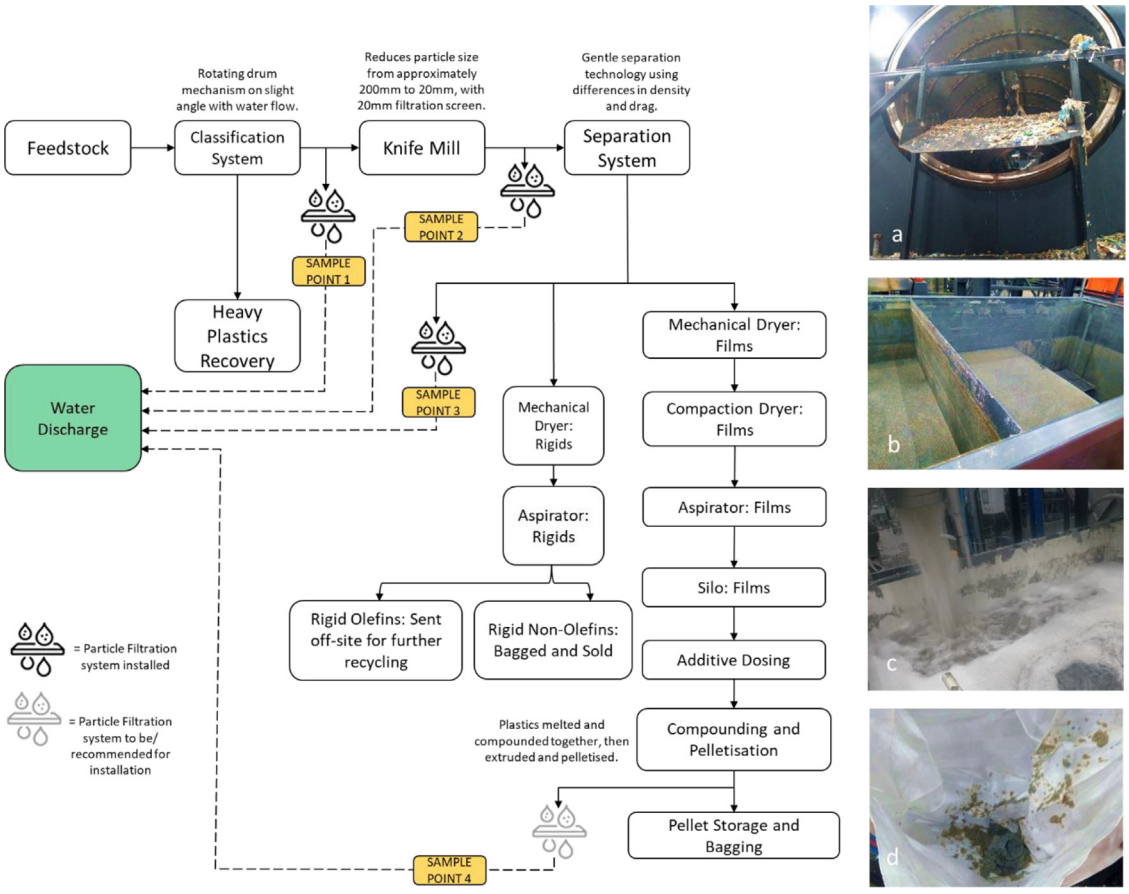


Fig. 1. Block flow diagram of PRF showing plastic flow through processes and cumulative water discharge from each point, with images respectively showing rotating drum with discarded plastics towards heavy plastics recovery, pre-filtration separation system water with MPs sitting as a sludge on top, the filtration system in operation, and manually skimmed sludge of MPs from post-filtration sample. a. rotating drum showing large plastics discarded towards heavy plastics recovery (pre-filtration). b. separation system tank showing pre-filtration levels of MPs sitting as a sludge on the water surface upstream from the sample 3 discharge point. c. water tank with filtration installed showing flow of filtered water into tank. d. sludge containing MPs manually skimmed from tank surface (post-filtration)

necessary x20,depending on average particle size within each sample. Organic material, after H2O2 digestion, is known to present limited fluorescence within the emission/excitation 460/525nm above an elevated threshold (Erni-Cassola et al., 2017) and therefore the differentiation between plastic and organic particles is clear using this method and appropriate fluorescent thresholds (Erni-Cassola et al., 2017). Where particles were large and of a variety of sizes (and therefore z-foci), multiple images were taken at different focal points. Microscopic imaging was performed under bright field (BF) light and Nile Red fluorescence to assist particle identification accuracy, maximise particles detected and quantified (BF images provided visual check to size and shape and highlighted any clumping of particles on the filters). Fluorescent images were collected using fluorescence exposure of 30ms at 10-15% fluorescence light strength (power). Fluorescence microscope imaging was undertaken in a clean and controlled laboratory setting and dark environment to minimise light influence and contamination of samples.

ImageJ (version 1.53, (Schneider, Rasband and Eliceiri, 2012)) analysis was performed across the total line section of images of each filter. Microplastic particles were identified following the identification key process defined in Kukkola et al., 2023, Figure 4, after image processing following Erni-Cassola et al., 2017 ImageJ methods and fluorescence thresholds. Following the processing of each image to allow software analysis of the particles, the area, perimeter, circularity and Feret diameter of each particle was found (Dražić, Sladoje and Lindblad, 2016). A minimum Feret diameter of 1μm was set to avoid overcounting of pixels. Particles with a 1:3 width to length ratio were categorised as fibres, with the remaining particles categorised as fragments. The area and Feret diameter were then used to categorise the particles into 30 categories ranging from <1.6-5μm to 4000-5000μm.

2.4. Data analysis

Data from particle analysis in ImageJ was exported to excel for all particles and samples. The sample area analysed (A_S) was identified through the image area assessed and verified against the sample filter diameter and area (A_F). All sample counts were blank corrected (MP_B – MP count for total blank filters). The total particle count for each analysed filter area (MP_S, MP/mm^2) was used to estimate the MP count for the total filter (MP_F) through proportional extrapolation ($MP_F = ((MP_S/A_S) - MP_B) x A_F$).

The MP particles were binned into size groups of <5μm, 5-10μm, 10-20μm, 20-30μm, 30-40μm, 40-50μm, 50μm steps up to 1000μm then 1000μm steps up to 5000μm (see supplementary data) for each filter and all blanks. Using these binned values for each sample, the percentage of total MP particles within each range was calculated. The mean and standard deviation for each sample location, pre and post filtration, was then calculated from the triplicate samples.

The MP per volume of filtered wash water (MP_V, MP/L) was then calculated using the MP per filter (MP_F) divided by the filtered wash water volume (V_F, MP/L) ($MP_V = MP_F / V_F$). There is an assumption due to PRF design (and through observation) that the wash water at each of the discharge sampling points (1-4) are homogeneously mixed. However, to address the potential loss if ineffective mixing occurs, three scenarios from most to least conservative were made for the calculation

of total MP discharge. These were: 1) that the MP floated in only the top 1cm of the wash water (0.2% of total tank volume), with the remainder of the tank volume being 'plastic free' in each of the sludge or collection tanks upstream from the discharge point; 2) that the MP floated the top 20cm of the upstream tanks (16.7% of total tank volume) with the remaining tank volume being 'plastic free'; and 3) that the sample was representative of a homogeneously mixed upstream tank and discharge. Acknowledging the tank volumes (tanks relative to sample point 1, 2 and 4 = 3760L, sample 3 = 9350L), PRF water inflow if 26.7L/s and the daily PRF leakage/loss of 80L/hour (supplementary note 2), the relative MP count per tank discharged per day was estimated (supplementary note 6, supplementary dataset). To enable tentative comparative analysis, MP/L and MP/tank was calculation for each sample point for both pre- and post-filtration system implementation.

All possible MP discharge concentrations were reported (Table S3, i.e. if post filtration discharge from sample point 1 dominated the overall PRF discharge then the discharge MP concentration could be close to 7.47×10^{10} MP/m^3). An assumption of equal discharge from each location could be used to estimate the PRF wash water discharge microplastic concentration ('Equal proportional discharge estimate' table in the Sample Summary tab of the supplementary data).

However, the proportional discharge rates from each tank and sample location are not recorded and therefore a conservative (worst case) approach to estimating potential daily microplastic discharge was used. To provide a conservative estimate, maximum concentrations (from the most concentrated MP discharge point) were presumed to dominate the PRF discharge (provide the largest proportional outflow). Therefore, the MP concentrations presented in the abstract, figures and supplementary information illustrate the estimated maximum MP discharge concentration from this PRF wash water for the different MP particle size ranges (MP>1.6 μm, MP>10 μm, MP>20 μm and MP>50 μm, enabling comparison with previously published studies) and for the three wash tank mixing scenarios (MP only floating in the top 1cm of tank water through to fully homogeneous mixing).

However, to ensure the 'best case scenario' is also estimated, the minimum potential MP wash water discharge concentration was also used in the total discharge estimates ('Minimalist MP discharge estimate table in the Sample Summary tab of the supplementary data). This estimated to concentration (and therefore enabled potential total MP mass results) for pre and post filtration findings based on the presumption that the lowest MP concentration discharge was the predominant (or only) discharge point from this PRF.

2.5. Quality assurance and control

During collection, the sampler wore cotton clothing to ensure minimal plastic contamination of the water samples (Murphy et al., 2016; Allen et al., 2019). Furthermore, the sampler stood downwind of the sample point to minimise contamination of plastics and organics into the samples (Allen et al., 2019). Between each use, all glass and stainless-steel laboratory equipment used in these processes were washed, then rinsed three times internally and once externally with Milli-Q to prevent MP contamination (Bayo, López-Castellanos and Olmos, 2020; Lares et al., 2018). Sample processing was conducted in a controlled dedicated MP laboratory in which measures were taken to minimise plastic contamination, such as the prohibition of any plastic clothing, use of a HEPA filter and 100% cotton lab coats, and the use of nitrile gloves when handling chemicals (Sun et al., 2019; Wang et al., 2020; Zhang et al., 2021). All glassware and equipment when not in use was wrapped in aluminium foil to protect the samples and other liquids used against contamination by airborne MP particles (Ziajahromi et al., 2017; Sun et al., 2019; Bayo, López-Castellanos and Olmos, 2020).

During preparation of the containers, a 'full process field blank' sample was also prepared, again in triplicate. These bottles were prepared in line with the sample methodology, with the addition of 200ml of Milli-Q into the bottle (to help retain any deposited particles). During sampling, the blank sample lid was taken off and the container left to sit in the location of sample collection and for the same length of time as each sample collection. This blank thus gives an indication of the field contamination in the sampling area and from the field sampling actions (Allen et al., 2019; Sun et al., 2019).

The laboratory positive control analysis was also completed. Positive controls were created using ground 2-50μm polystyrene fragments or known particle size distribution and mass, suspended in Milli-Q water at a set concentration (Supplementary Note 4). 2ml was extracted from a solution of spiked Milli-Q water (whilst magnetically spinning the sample to ensure even distribution of the particles throughout). This volume of spike was diluted into 100ml Milli-Q already resting on the filter. Following the vacuum filtration of this liquid, a further 100ml of Milli-Q was filtered through, including the flushing of the sides of the filter holder with Milli-Q. In line with the methodology used throughout this research, three repetitions of this positive control were performed. These filters were analysed in accordance with the analysis of the field samples. The positive control results (95% recovery, standard deviation of 29%, supplementary note 4) showed satisfactory MP recovery efficiency and validated the Nile Red fluorescence analysis results.

All samples were blank corrected using full process field blanks. Blank correction was conservatively completed relative to particle size and particle count. Full process blank samples presented 61 ± 4.5 MPs per sample (<10% of the smallest MP sample count, Fig. 2 and Fig. S2).

3. Results and discussion

Whilst multiple studies have been conducted focussing on MP pollution from WWTPs, very limited research exists on the study of MP pollution from PRFs. Within this emerging research field, this research highlights PRFs as a potential key point source of MP pollution for receiving waters and, in comparison with the few other studies in this field, shows the release of very high concentrations of small MPs, particularly environmentally relevant sized MPs of <10μm.

3.1. MP quantification in discharge

Microplastic particles were found in all samples. Pre-filtration samples of MP >10μm were compared to post-filtration sample results to quantify the filtration efficiency in the removal of MP from discharge wash water. The comparative analysis of pre and post filtration implementation was undertaken for MP >10μm. Pre-filtration MP >10μm counts (representing discharge without any filtration mitigation) ranged from $4.93 \times 10^8 - 9.55 \times 10^9$ MP m^{-3}. Post-filtration MP >10μm counts ranged from $2.17 \times 10^5 - 1.01 \times 10^9$ MP m^{-3} (Fig. 3a, supplementary data). There is a general decrease in MP >10μm particles of 4.82×10^8 to 9.39×10^9 MP m^{-3} (mean $3.41 \times 10^9 \pm 3.37 \times 10^9$ MP m^{-3}) as a result of filtration implemented in the PRF.

The filter counts were multiplied to the full volume of the respective standard tanks (3760L, Table S3). It is assumed that the MP count are generally not evenly distributed throughout the entire tank volume, as MPs could clearly be seen floating on the water surface (Fig. 1). As the sampling process did not allow for sampling from varying levels within the tank volume, an assumption was made that the volume of the top 1-20cm of the tank could provide likely estimations of the MP count. This yielded three assumptions of MP concentration within a 1cm depth, a 20cm depth, and a fully homogenised tank (Fig. 3b). The majority of MPs in the post filtration samples were found to be >10μm (93-100%) (Table S5). The sample MP counts were then extrapolated up to create an equivalent MP count per tank (presented as MP m^{-3} relative to the tank total volume) (Fig. 3b). If it is assumed all MPs are floating and contained within the upper 1cm of water in each tank, then the discharge from each of the four release points in the PRF ranges from $4.1 \times 10^5 - 6.2 \times 10^8$ MP m^{-3} (all particles >1.6μm, Table S3), and $4.1 \times 10^5 - 6.2 \times 10^8$ MP m^{-3} for MP >10μm (Fig. 3b). This increases by an order of magnitude if the upper 20cm of the tanks are considered to contain

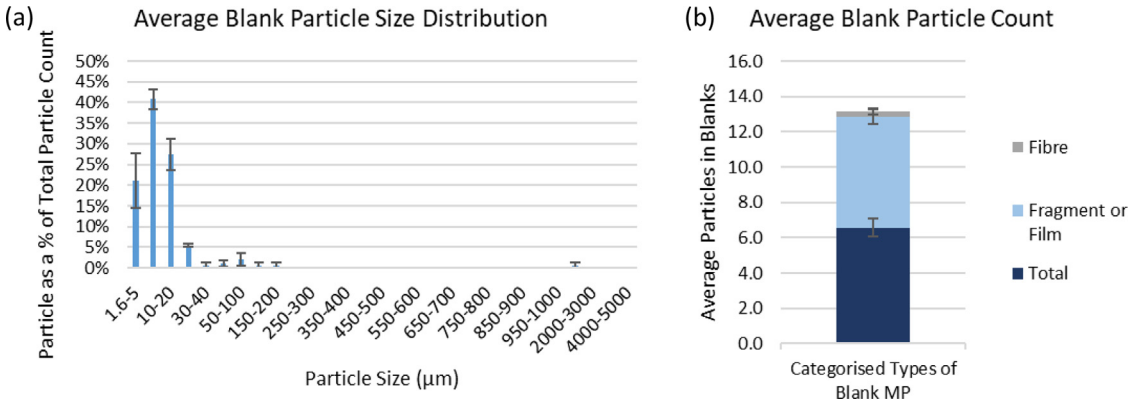
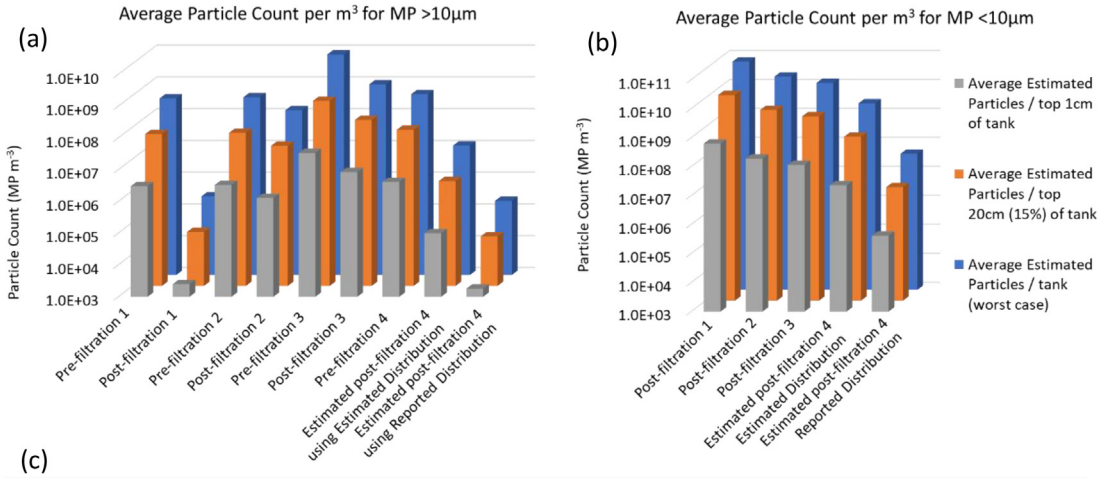


Fig. 2. Average particle size distribution (a) and count for field blanks (b).



(c)

Possible Plastic Recycling Facility Microplastic Discharge, MP count m^{-3}	Average Estimated Particles / tank (worst case)	Average Estimated Particles / top 20cm (15%) of tank	Average Estimated Particles / top 1cm of tank
MP > 1.6µm	7.47E+10	1.25E+10	6.23E+08
MP > 10µm	1.01E+09	1.68E+08	8.38E+06
MP > 50µm	4.25E+08	7.09E+07	3.54E+06

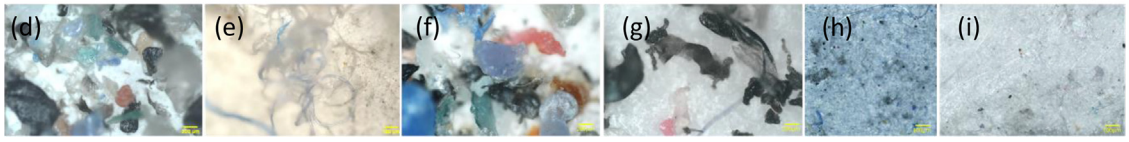


Fig. 3. Summary of average particle counts per m^3 per sample, showing higher particle counts in all pre- compared to post-filtration samples (a). Post-filtration MP <10µm (the majority of the post-filtration MP particles) representing the MP being discharged from the Plastic Recycling Facility (PRF) (b). The estimated total PRF discharge, comprised of all 4 sampled discharge points after filtration (c) and representative bright field images from pre-filtered samples (d-g) and post-filtration (h-i).

the MPs (as represented by the samples; $8.2 \times 10^6 - 1.2 \times 10^{10}$ MP m^{-3} for MP >10µm, $8.3 \times 10^6 - 1.3 \times 10^{10}$ for all particles >1.6µm), and two orders of magnitude if it is assumed (worst case scenario) that the sample is representative of a homogenised tank ($4.9 \times 10^7 - 7.5 \times 10^{10}$ MP m^{-3} for MP >10µm, $5.0 \times 10^7 - 7.5 \times 10^{10}$ for all particles >1.6µm). When all four discharge points are combined to represent the potential total MP discharge through wash water from the PRF, MP discharge was estimated to range from 6.2×10^8 to 7.5×10^{10} MP m^{-3} (MP >1.6µm, Fig. 3c).

Pre-filtration sample 2 (mean 9.94×10^9 MP m^{-3}) was found to have almost double the MP count of sample 1 (mean 5.42×10^9 MP m^{-3}), suggesting that the knife mill process itself generated a high number of MPs compared to the rotating drum classification process. This could be due to the breakdown process in the knife mill being much more vigorous than the separation process in the rotating drum, therefore breaking MPs down into a higher number of smaller MPs. Pre-filtration sample 4 (mean 7.16×10^8 MP m^{-3}) had a much lower MP count than other sample locations. This could potentially be caused by number of processes and discharge points the plastics encountered throughout the PRF process (Fig. 1, Fig. S1). Alternatively, the nature of the melting, compounding and extrusion processes might result in the 'soaking up' of many MPs into the final product, giving a lower MP count at this wash point.

All samples have low fibre MP counts, varying 0-1% in post-filtration samples and 2-10% in pre-filtration, suggesting that small numbers of fibres are generated by the PRF processes (Table S2). The higher occurrence of fibres in pre-filtration samples could potentially be due to an undercount of smaller particles (focus on MP >10µm due to lack of

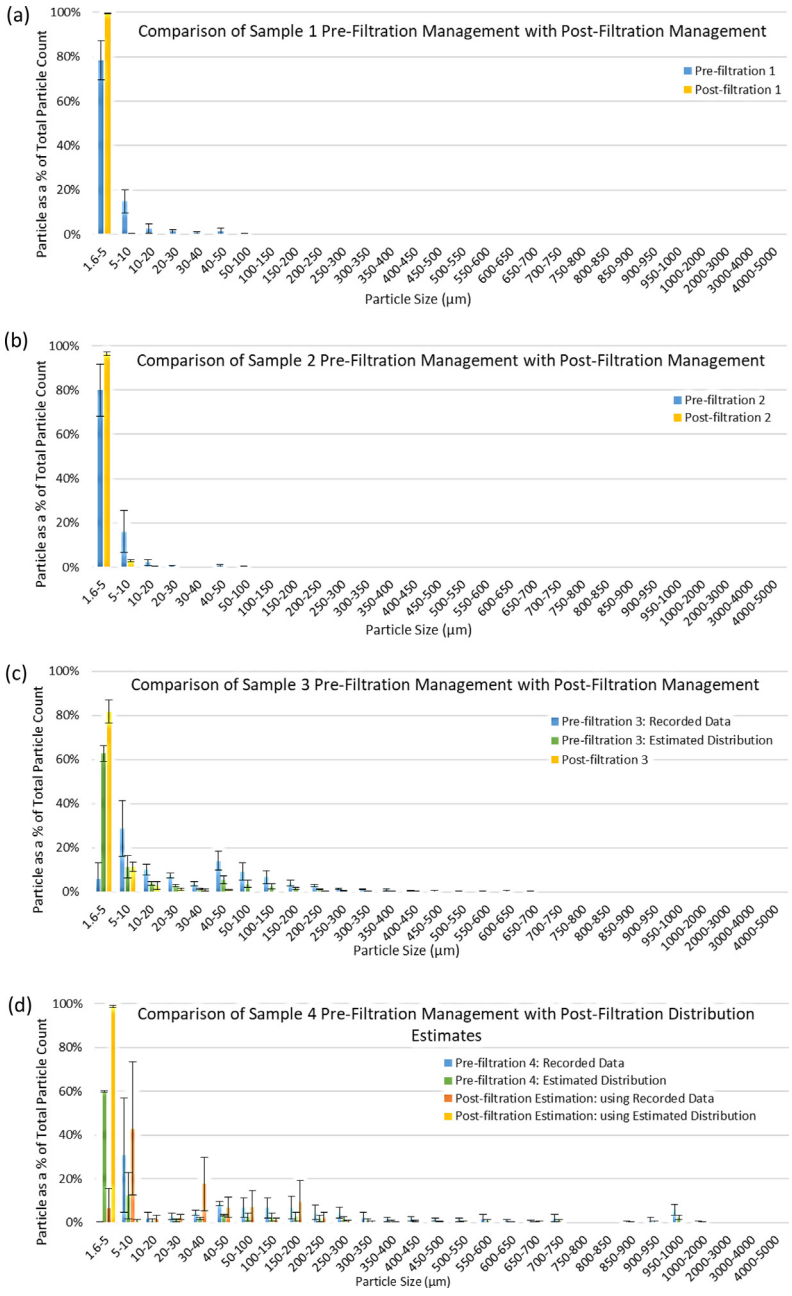


Fig. 4. Particle size distribution for pre and post filtration samples (standard deviation shown as error bars) (a-d).

size fractionation in this study) and thus over-representation of larger particles and fibres (specifically pre-filtration sample 3 and 4, Table S5), a smaller total count of particles giving any fibre present an over-representation (sample 4), or a higher occurrence of fibres due to extrusion and pelletisation processes (sample 4, Table S2).

3.2. MP particle size distribution

Pre-filtration samples 1 and 2 both consist almost entirely of MP <10µm (96% and 93% respectively), with 78% and 80% (respectively) <5µm, indicating that both the rotating drum and the knife mill generate a similar size distribution of MPs. Post-filtration samples 1 and 2 exhibit a larger peak in MP <5µm (100% and 96% respectively), with 100 and 99% of MP <10µm (respectively). This suggests that this method of filtration used at the rotating drum and the knife mill is not effective in removing most of the particles, which lie <10µm (Fig. 4a, b).

Pre-filtration samples 3 and 4 do not exhibit a similar peak in particle size distribution (Fig. 4c, d). 35% of sample 3 MPs lie <10µm, with more MPs at 5-10µm than <5µm, coupled with another small peak of 14% at MPs 40-50µm. Only 31% of sample 4 MPs lie <10µm, of which 0.2% MPs lie <5µm. The high quantity and size of larger MPs in pre-filtration samples 3 and 4 suggests that MPs <5µm and <10µm were underestimated due to the masking of smaller MPs by larger MPs. With the removal of larger MPs through size fractionation (sieving or multiple stage filtration), the smaller MPs could be analysed and quantified in the post-filtration samples. This is the assumed reason behind the increase in total MP count from pre-filtration to post-filtration sample 3 when the total size range (1.6µm – 5mm, sample 3 pre and post filtration: 6.17×10^9 MP m^{-3} and 1.47×10^{10} MP m^{-3}, Table S3) is considered rather than MP >10µm (Fig. 3). Estimates of a more likely pre-filtration distribution for samples 3 and 4 (supplementary note 5) show a trend closer to the expected negative log trend found in studies on MP con-

tent within water environments (Kooi and Koelmans, 2019; Erni-Cassola et al., 2017; Enders et al., 2015; Bergmann et al., 2017; Eo et al., 2019). These estimations are extrapolated using post-filtration sample 3 distribution (82% of MP <5μm) or post-filtration average distribution of samples 1-3 (93% of MP <5μm) in which the larger MPs were filtered out, hence enabling an estimation of smaller MPs.

No filtration was installed at sample 4 location in the PRF, and therefore estimations were calculated for post-filtration 4 using averages for samples 1, 2 and 3 and using both pre-filtration 4 recorded data and the more likely estimated distribution (Supplementary note 5). If a filtration measure were placed on the discharge point of sample 4 the particle size distribution may mimic that seen in samples 1-3 post filtration, skewed to the <5μm particle size. Filtration at this location could potentially remove up to 7.85×10^8 MP m^{-3} (potentially >90% of the MP >10μm).

3.3. MP mass discharge from the PRF and comparison to WWTP and river MP concentrations

A tentative, site specific early estimation of the mass of MP discharged from the PRF via wash water provides an indication of the annual microplastic point source wash water pollution discharge from this plastic recycling facility. Pre-filtration PRF discharge is calculated to range from 1.4×10^5 to 4.3×10^6mg m^{-3} (see Estimated Particles per Top 1cm and per Top 20cm (MP >1.6μm), supplementary note 6), and 6.3×10^3 to 2.0×10^6mg m^{-3} post-filtration (see Estimated Particles per Top 1cm and per Top 20cm (MP >1.6μm)). This equates to 96 – 2933 tonnes per annum of MP discharged in pre-filtration water and 4 – 1366 tonnes per annum of MP in post-filtration water. Given 22680 tonnes of plastic waste are brought the PRF for processing per year, the MP released relative to the tonnage imported to the plant is up to 0.06 tonne/tonne for post-filtration discharge (estimated discharge using the Top 20cm (MP > 1.6 μm) results, supplementary data). This equates to approximately 6% of the mass of plastic waste brought to the PRF for recycling (0.004-0.13 tonne/tonne), but rises to up to 13% if no filtration is provided to the wash water (pre-filtration Estimated Particles per Top 20cm (MP >1.6μm), supplementary note 6). If the current state of filtration is considered (filtration at sample points 1-3 but no filtration at sample point 4) then the annual MP discharge currently occurring from this PRF is estimated to be 59 – 1184 tonnes (as shown in the graphical abstract*, 'estimated mass calc' tab of the supplementary data), which is up to 5% of the imported plastic waste to this facility. While this is only a relatively small proportion of the total imported plastic mass, the mass released even after full filtration (4 – 1366 tonnes per annum) is not insignificant to a receiving waterway of sewer network. Given that the discharged MP particles are predominantly <10μm and therefore pose a risk to ecosystem health, these results highlight the need for PRF discharge monitoring, implementation of mitigation filtration beyond the implemented 50μm sieving and regulation to combat this PRF point source MP pollution.

To provide a comparison with surrounding water quality, the PRF results are considered relative to reported wastewater treatment plant MP concentrations and surface water MP studies. When compared against Scottish WWTP influent (found by Murphy et al. 2016; LoQ of 11μm), the WWTP influent, at 1.57×10^4 MP/m^3 ±5230, is several orders of magnitude lower than the estimated total of post-filtration concentration from this study for MP >10μm (MP range from 1.8×10^3 to 1.0×10^9 MP m^{-3} for estimations based on the assumption of MPs present at the depths of 1cm, 20cm, and a fully homogenised tank). The difference could potentially be due to a more varied WWTP feedstock (not solely plastic focussed) or the presence of multiple WWTP water quality management systems (primary, secondary and potentially tertiary). This would allow for some pre-treatment processes to have reduced MP content before sampling. Similar comparison can be made with a Finnish WWTP influent of 5.68×10^8 MP m^{-3} (Talvitie et al., 2017) (LoQ of 20μm). This MP concentration is much more comparable to this study's post-filtration total estimates for MP >20μm (1.7×10^3 to 5.8×10^8 MP m^{-3}).

In comparison to other published studies, previous research at a separate PRF using visual methods (without fluorescence) counted MP >0.001mm^2 (1000μm^2) in the discharge water from the operational PRF. This is equivalent to an approximate diameter and length of 35μm and 50μm for fragments and fibres respectively, finding 5.0×10^7 MP m^{-3} (Larsen, 2021). Estimates for post-filtration concentrations of MP >50μm range 6.3×10^2 to 1.2×10^8 (1cm depth to full homogenous tank).

There are currently only two published analyses of MP discharge in PRF wastewater (Guo et al., 2022; Suzuki et al., 2022), however these only identify MPs to LoQ of 100μm and 315μm respectively (Fig. 5).

Pre-filtration wastewater samples of three Chinese PRFs were found by Guo et al. (2022) to contain a range of MP concentrations from 23.43 – 1836.37mg L^{-1}, with post-filtration effluent containing 8.13 – 83.83mg L^{-1}. Effluent of three Vietnamese PRFs was found by Suzuki et al. (2022) to range between 26 – 3400mg L^{-1}. In comparison with these studies, the results of this study are converted to mg L^{-1}. Mass estimates are calculated assuming a commonly utilised general MP mass of 1g cm^{-3} and using the known particle counts and particle size distributions. It is acknowledged that this may be a simplistic and tentative estimation method but does provide some early insight into the possible relative and comparative mass MP discharge estimates. Both the pre-filtration and post-filtration values found in all assumed depths (1cm, 20cm and full tank) sit in a similar range with values found by Suzuki et al. (2022) and Guo et al. (2022) (Fig. 54a, b). These total post-filtration results, along with post-filtration results found by Guo et al. (2022), sit much higher than MP concentrations found in studies on surface waterbodies globally spanning 2014-2019 (Moss et al., 2021) (Fig. 5d). This shows the vast impact that the discharge of water from these PRFs would have on rivers globally if released directly into rivers, noting that the waterbodies listed in the figure are the top 23% regarding MP content in the study by Moss et al. (2021).

Kallenbach et al. (2022) reveal MP pollution in downstream river sediment as a result of PRF activity but focuses on quantification of MP concentration in macroinvertebrates and solid sediment. As a result, Kallenback et al. (2022) published findings cannot be directly compared with this study. Although Kallenbach et al. (2022) identified MPs to a LoQ of 75μm, no particles <110μm were found in the sediment. Kallenbach et al. (2022) gives one possible reasoning for this to be that the PRFs studied release only large particles into the wastewater, which contrasts the results found in this study. However, without detail into the PRF processes used for the PRF studied in the previously published research, it is difficult to understand the reason for this difference.

3.4. A note on atmospheric MP resulting from PRF activities

The field blanks were processed to give the total MP count in the PRF atmosphere (deposition during sampling) and a particle size distribution of these MPs (Fig. 2a, b). The field blanks helped ensure this study provide first MP counts for wash water specifically, but it is acknowledged that a total potential release (wash water plus atmospheric discharge) is important and should be the focus of future research. The results show high levels of MP content in the PRF atmosphere, with 61% of these MPs concentrated <10μm. PM10 (particle matter (PM) such as MPs <10μm) have been linked to illness in humans (Prata, 2018; Consonni et al., 2018), with human respiratory systems able to intake particles ranging 1nm-20μm (Leslie et al., 2022). Studies show diagnoses of interstitial lung disease in plastic processing factory workers (Warheit et al., 2001; Eschenbacher et al., 1999; Boag et al., 1999) due to inhalation exposure to the plastic particles and their associated chemicals. MPs have more recently been found to accumulate in human lungs ranging 1.6-5.6μm (fragments) and 8.3-16.8μm (fibres) (Amato-Lourenço et al., 2021) and to penetrate deep into the respiratory system (Jenner et al., 2022). While quantification of the atmospheric MP (beyond blank representation sup-

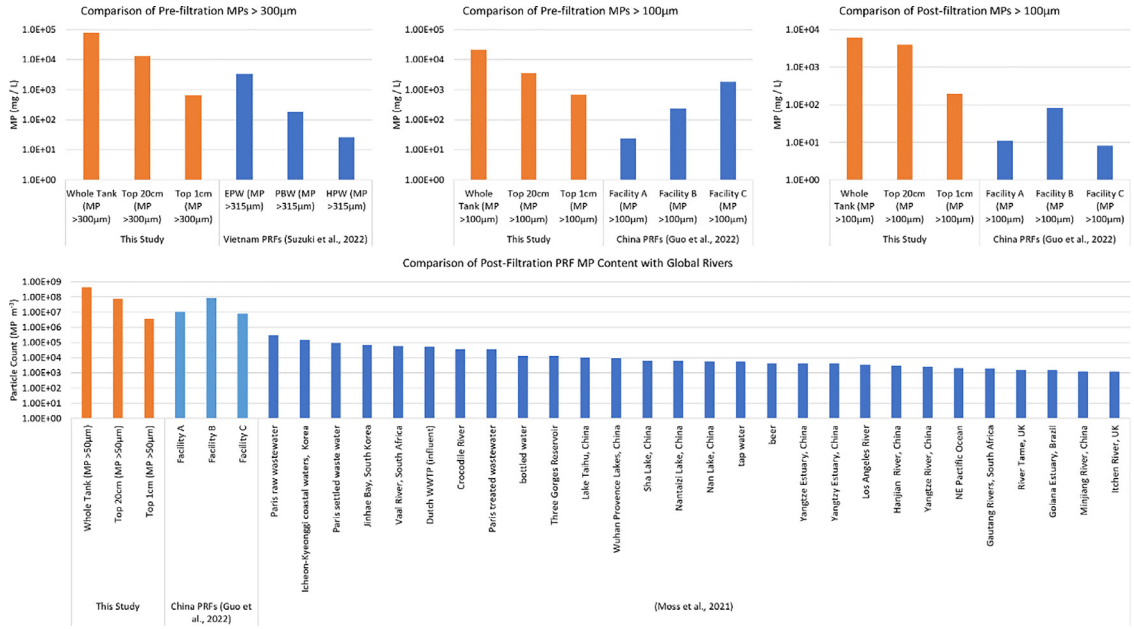


Fig. 5. Comparisons of pre-filtration and post-filtration estimated MP concentrations (for three assumed depths of MPs) with other studies conducted on MP pollution from PRFs (Suzuki et al., 2022, Fig. 5a; Guo et al., 2022 pre and post filtration, Fig. 5b, c), showing comparable results with both studies, and with surface water MP concentrations summarised by Moss et al. (2021) (Fig. 5d) from studies between 2014-2019, showing this study's MP concentrations to be much higher than all surface water concentrations.

porting the sampling activities) is not possible within this study, the blank results suggest that workers in PRF may be exposed to notable atmospheric and inhalable fraction MP. Further research is needed to quantify PRF worker exposure, and to quantify the atmospheric MP emission to the greater environment, and it is recommended that air quality monitoring regulations include observation of atmospheric MP in PRFs in the future (Enyoh et al., 2022; Kabir et al., 2021; Lithner et al., 2021). Following the precautionary principle, it is also recommended that any workers present in these PRF conditions wear masks to protect against the high likelihood of inhalation of such MPs. N95 respirators could provide protection against MPs down to sizes of approximately 0.1-0.3µm (Qian et al., 1998).

4. Conclusions

This pilot study provides an insight into the potential for plastic recycling facilities to function and microplastic pollution sources and considers filtration mitigations measures to combat the MP release to receiving waters. While it is acknowledged the results present insight into one recycling facility in one location, it highlights the need for further future research into considering the potential of PRF to be a point source of MP pollution, the impact of plastic recycling facilities beyond their recycled product and facilities design to include mitigation measures for wash water to prevent receiving water pollution.

The results show the generation of vast numbers of MPs in PRF water for the PRF in this study and demonstrates the potential for release of MPs into water through plastic recycling processes such as used by the PRF in this study. PRFs could therefore be considered a source of MP to receiving waterbodies (rivers or the sewer network) of a wide range of particles (if no filtration is used) and specifically the environmentally relevant <10µm and <5µm MPs (given current filtration implementation). The current process of recycling is a potential source of plastic pollution to the environment that it is designed to help prevent. Future studies of PRFs are highly recommended to provide a comprehensive understanding of MP creation and release due to PRF actions (both atmospherically and via water discharge) across multiple facilities globally. To advance this assessment it is recommended size fractionation of samples prior to analysis is undertaken, to ensure smaller particles accurately represented in analysis and to prevent potential 'hiding' and sample overloading.

The installed filtration efficacy comparing pre-filtration and post-filtration samples identified filtration to be effective for larger MP particles but allowed smaller MP (<10µm) to be discharged into the receiving waterway/network. The results show that the rotating drum (sample location 1) and knife mill (sample location 2) wash tanks in the PRF require the installation of filtration better suited to the particle profile found (designed to filter smaller particles) and that the separation system (sample location 3) and the compounding and pelletisation (sample location 4) wash tanks require additional filtration to remove smaller particles in addition to the current filtration. Further research on this potential source of MP pollution is needed to support progress within the plastic recycling industry without these facilities potentially contributing to an increase of global MP pollution.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

CRediT authorship contribution statement

Erina Brown: Conceptualization, Resources, Methodology, Data curation, Investigation, Writing – original draft, Writing – review & editing. **Anna MacDonald:** Methodology, Investigation, Writing – original draft, Writing – review & editing. **Steve Allen:** Methodology, Writing – original draft, Writing – review & editing. **Deonie Allen:** Resources, Methodology, Data curation, Investigation, Writing – original draft, Writing – review & editing.

Data availability

Data used in this paper are provided in the Supplementary Data.

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Supplementary materials

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