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Research Paper

Losses and emissions in polypropylene recycling from household packaging waste

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ABSTRACT

In this study we replicated a typical high-quality post-consumer polypropylene (PP) recycling process to investigate its losses and emissions and study potential improvements. To our knowledge this is the first time that quantitative measurements on all process steps have been performed instead of an accumulated yield and emissions in water. In the process an overall PP yield of 85 wt% based on pure PP input is achieved. The loss of target material is largest at the two mechanical dryer steps (6.6 wt%) and in the wet grinder combined with friction washers (4.0 wt%). In the process we observed approx. 3.9 wt% of the PP input as microplastics in the wastewater before the dissolved air flotation unit which is capable of 97–99 % mass-based removal of microplastics (MPs). Around 330 μg of PP was emitted to air at the mechanical drying step for each kg of input material. This is a very low mass fraction, but considering the particle size distribution the number of particles is vast. This emission can be reduced by using air filters at locations where MPs are generated. To reduce losses and emissions we investigated a few potential process changes. Compared to current practice, positive results were achieved by ensuring that the knives of the wet grinder remain sharp. The mechanical drying process can be improved by lowering the centrifugal speed which reduces the generated microplastics here from 4 wt% to 1 wt% without significantly affecting the moisture content.

1. Introduction

Demand for plastic is high and is likely to increase in the coming years (Plastics Europe 2023). Only a circular plastic chain can meet this demand without increasing fossil fuel dependency and the subsequent acceleration of planetary crises (Lange et al. 2024). A crucial step in a circular plastic chain is the recycling of plastic products after use, producing high-quality recyclate. Despite the urgency, use of recycled plastics in new products is still low: in 2022, 400 million tons of plastic was produced globally, of which only 9 wt% was sourced from recycled plastic (Plastics Europe 2023). (Inter)national policies such as the upcoming EU Packaging and Packaging Waste Regulation (PPWR) (European Commission 2022) or the National Circular Economy Programme (Government of The Netherlands 2023) specify targets for waste recycling, *e.g.* in the PPWR by 2034 at least 55 wt% of all plastic packaging should be recycled. In 2020, the overall European recycling rate for post-consumer plastics packaging reached 46 % (under the former Packaging and Packaging Waste Directive calculation methodology which is highly overestimating recycling rates) (Plastics Europe 2022). Reaching these targets will require increased recycling infrastructure and organisation, alongside technological innovation.

In the case of plastic packaging, mechanical recycling is still the major recycling technique. During any mechanical recycling process, losses and emissions occur, reducing material efficiency and creating potential environmental impact (Lase et al. 2022). Losses are defined as the mass of target plastic that exits the recycling process anywhere else than in the final product stream, *e.g.* due to incorrect sorting or being washed away. Emissions consist of any material, including the target plastic, that are discharged throughout the recycling process. These should be minimised and managed.

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One form of losses and emission are microplastics – plastic particles under 5 mm. These particles are ubiquitous, having been found in bottled water (Gambino et al. 2022) and food products (Kwon et al. 2020), remote ecosystems (Horton and Barnes 2020), and throughout the human body (Ragusa et al. 2021; Ragusa et al., 2022; Jenner et al. 2022; Leslie et al. 2022). Although the presence of microplastics in our surroundings is undeniable, a complete risk assessment for microplastics is still lacking. Given the lack of evidence that human exposure to microplastics is safe, the precautionary principle (Rio Declaration 1992) determines that (micro)plastics emission should be reduced where possible (United Nations 1992). To minimise microplastic emission from mechanical recycling, it is essential to improve our understanding of plastic losses and emissions throughout the process and to develop effective mitigation strategies. Reducing microplastic formation during mechanical recycling would both increase material efficiency and reduce environmental emissions. Where reduction is no longer possible, mitigation routes for the capture of microplastics should be investigated. A study of wastewater discharge from a plastic recycling facility in the UK found microplastic counts between 5.97 x 10^{6} –1.12 x 10^{8} MP m^{-3} (Brown et al. 2023). In the effluent of a Turkish recycling facility similar numbers were found (Çolakoğlu and Uyanık 2024). Analysis of surface water around a plastic recycling facility without water treatment in

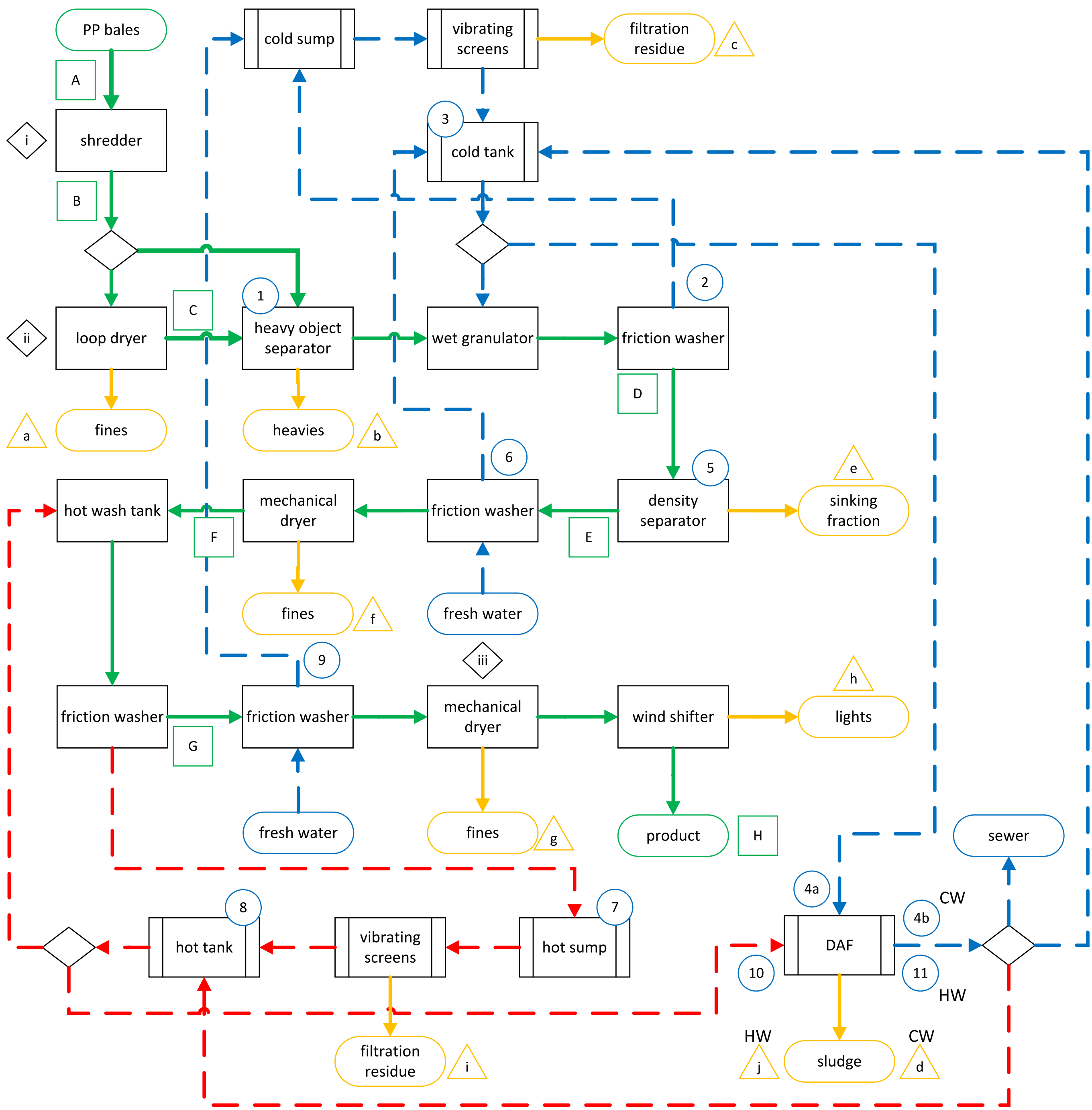

Fig. 1. Schematic representation of the used PP recycling process. Unit operations for the target material are depicted as rectangular boxes, whereas the unit operations for water streams are depicted as banded rectangular boxes. Solid arrows indicate product (green) and residue streams (amber), whereas dashed lines are the cold (blue) and hot (red) water flows. In the flow diagram diamonds indicate options in flow direction. In the diagram the different sampling positions are indicated for product (squares A-H), residue (triangles a-j), water (circles 1–11) and air (diamonds i-iii).

Vietnam also indicated increased microplastic concentrations (Suzuki et al. 2022).

Other emissions in a mechanical recycling process are related to residual packed product, adhesives, inks, labels, *etc*. and are also considered in this work. In this context, a significant lack of understanding exists for the polluting potential of recycling facilities. To gain a detailed insight in the process and enable reduction of its losses and emissions, we measure the full mass balance throughout the mechanical recycling of rigid polypropylene (PP) packaging waste, sorted from source separated kerbside collection in the Netherlands. We track and characterise PP lost throughout the process and other emissions to water and air. To the best of our knowledge, no detailed mass balance for a mechanical plastic recycling process is publicly available. To mitigate the losses and emissions, we explore different process routes that can achieve this.

2. Materials and methods

2.1. Polypropylene recycling process

In this study, we used the industrial-scale modular and flexible sorting and recycling facilities at the applied research organisation NTCP (Heerenveen, the Netherlands) to mimic industrial practices in a controlled environment. Typical throughputs are 2 and 0.5 ton/h for sorting and recycling, respectively. Before mechanical recycling, rigid PP must be extracted from kerbside collected packaging waste. This is typically done in a cascade of mechanical object sorting steps, including near-infrared (NIR)-based optical sorters, to achieve a stream in which at least 94 wt% of the objects is primarily PP (typical specification). In this study, material from a Dutch material recovery facility (MRF) was purified towards white and transparent objects by optical sorting. This stream will still contain some non-target materials such as non-PP objects, labels, adhesives, moisture and surface contamination. The used PP recycling process is described below, and any references refer to the schematic representation of the process in Fig. 1. Unless stated otherwise, all process equipment is supplied by Linder Washtech, Austria. Detailed information is available in the Supplementary Information (S.I.).

2.1.1. Shredding and dry wash

The sorted PP was shredded in an Antares 1900 shredder (80 mm circular mesh screen). Although uncommon in PP recycling, for exploration purposes, we dry washed a small fraction of the shredded material in a MTH 100/150 loop dryer (3 mm circular mesh screen) with the aim of removing moisture and surface contamination, further discussed in the mitigation section. In this experiment, air emissions were measured above the shredder (i) and above the cyclone (ii) after the loop dryer.

2.1.2. Cold wash

The shredded material is first treated in the cold wash section using a heavy object separator, wet grinder (GSH 500/600), friction washer FW 600 35°, density separator (Terraplast), friction washer (FW 600 35°) and mechanical dryer (MTH 100/150). The heavy object separator prewets the material and removes objects with a density substantially higher than water (b); this protects the downstream equipment. The wet grinder (15 mm circular mesh) creates plastic flakes large enough to enable further transport and prevent losses in screens and small enough to allow for high friction in the washing process. Subsequently the material is washed in a friction washer. Water and fine particles are separated from the flakes through a screen (2.5 mm circular mesh) (2). Water is circulated in a closed loop over the cold-water tank (3), the wet grinder and friction washer. The contaminated water is collected in the cold sump and returned to the cold-water tank over a double vibrating screen (mesh sizes 2 mm and 500 μm) where particles are removed (c).

2.1.3. Density separation

Next, the material (D) passes through a density separator where the sinking fraction containing higher density plastics is collected as a reject stream (e) and the floating fraction (E) passes through another friction washer to rinse the material using fresh water. Subsequently, the material is dried in a mechanical dryer where fines and water are separated from flakes (F) through a screen (f) with a circular mesh size of 3 mm.

2.1.4. Hot wash

In high-quality mechanical recycling of PP hot wash is applied to remove more contaminants such as organic product residue, inks and adhesives. We used a batch hot wash process where 75 kg flakes were washed in 500 L water. The water was preheated in the hot tank (8) where a typical industrial detergent formulation for plastic recycling (MacDermid Enthone RP 14-LF including potassium hydroxide) was added at a concentration of 21.5 g/L. This results in a solution with 0.038 M KOH (pH > 12). The washing water with fines was separated from the product through a screen (circular mesh size 2.5 mm) at the bottom of the friction washer and collected in the hot sump (7) and pumped back to the hot tank with filtration over a double vibrating screen with subsequent mesh sizes of 1 mm and 200 μm. The material from the friction washer (G) was processed further in a second friction washer (FW 600 35°) with the main purpose to reduce the moisture content. The material was dried further in a mechanical dryer (MTH 100/150) removing more fines as a reject stream (g). Air emissions are measured above the cyclone after the mechanical dryer (iii). In the last step, material passed through a zigzag wind shifter (ZZS 180x400-4 – Kat. 1) where film material was removed in the light fraction (h) and the product was collected in big bags (H). For each of the described washing sections, a mass balance was determined. All input, output and reject streams are collected during a measured timeframe and the compositions of these streams are determined afterwards. For solid samples (flakes or fines), the amount of moisture and surface contamination and the composition is determined. Water samples are collected at the relevant locations in the process and the composition of the solids in these samples is determined.

2.1.5. Water treatment

In the cold and hot wash steps above, the wastewater contains a mixture of dissolved and suspended solids. Use of water treatment can remove some of these solids and generally consists of two steps in a polypropylene recycling facility. In the first step, vibrating screens remove the larger suspended solids (see above). In the second step, a dissolved air flotation (DAF) unit (Nijhuis Industries, the Netherlands) removes more of the remaining suspended contamination. Water samples of the influent and effluent of the DAF are collected during the cold wash (sample 4a and 4b in Fig. 1) and during the hot wash (sample 10 and 11 in Fig. 1). In the DAF, a coagulant and flocculant were added to form larger particles (flocs). With the help of dissolved air, these flocs were made to float and subsequently removed by a scraper creating a sludge residue stream. In this trial, 0.5 L/m^3 Biofloc (coagulant) and 10 ppm Superfloc (flocculant) were used as recommended by Nijhuis. The hot wash results in high pH wastewater (pH > 12). Treatment of hot wash wastewater by dissolved air floatation (DAF) is optimal around pH 6.5, where no degradation of the flocculant takes place, and its effectiveness is highest. The pH was reduced by the addition of sulfuric acid (37 wt%) before the DAF unit.

When the water treatment with dissolved air floatation results in clean water, it is suitable for reuse thereby reducing the use of fresh water. The reuse is limited by the presence of dissolved contaminants which are not removed by dissolved air floatation. Here, during the cold wash two different types of water loops were applied. In the first part of the test the DAF is not used, and water is only looped over the vibrating screen, after reaching a high level of contamination the complete water system is treated in the DAF. In the second part a small fraction of the wastewater from the cold tank is continuously pumped towards the DAF and subsequently sent to the sewer, the water level in the cold wash loop is balanced by the addition of fresh water.

2.1.6. Residue streams

The process description shows that many residue streams (a – k) exit a polypropylene recycling facility. The exact composition of the residue streams varies, but generally contain a large quantity of plastic. Most of these residue streams are currently incinerated or end up in landfill but the high plastic content allows for a more valuable use and is discussed below.

2.2. Analytical methods

The online and offline analysis methods of samples throughout the process are described in detail in section 2 of the S.I. and presented here in short.

2.2.1. Air measurements

Air measurements were performed using both real-time online particle size measurements (particulate matter < 1 μm (PM1), PM10 and total suspended particles (TSP)) and sampling onto filters for offline analysis with thermogravimetric analysis followed by thermal extraction desorption-gas chromatography/mass spectrometry (TGA TED GCMS) and scanning electron microscopy (SEM) visualisation.

2.2.2. Water sample analysis

Grab-samples were collected at several points in the process. Total solids (TS) and total suspended solids (TSS) analyses were performed following the Standard Methods for the Examination of Water and Wastewater guidelines (Lipps and Baxter, 2023). For TS and TSS samples, TGA was performed to study solid composition in the water phase. The relative proportions of organic & volatiles, plastic and inorganics in the samples were determined. Differential scanning calorimetry (DSC) was used to analyse the presence of different polymers and TGA TED GCMS was used to determine PP mass concentration. For water samples conductivity was determined using an HI9930 EC/TDS-meter (Hanna instruments) and turbidity is determined using a Eutech TN-100 Turbidimeter. We also used standardised methods to determine the chemical oxygen demand (COD) and dissolved organic carbon (DOC). Total nitrogen (total-N) is measured using spectrophotometrically using Hach standard cuvette tests (Hach, Tiel, the Netherlands). DAF influent samples were diluted and analysed using LCK 338, while effluent samples were analysed using LCK 138. Elemental analysis of the sludge and water samples was carried out using microwave-assisted acid digestion and inductively coupled plasma optical emission spectroscopy (ICP-OES). Separate water samples for organic micropollutant analysis (OMP) analysis were taken from the cold water and hot water sump, at end of the cold and hot wash, respectively. Samples were collected following a similar approach described in for TS and TSS. Water samples were analysed for thirteen per- and polyfluoroalkyl substances (PFAS) and thirty-six other OMPs most commonly used as artificial sweeteners, pharmaceuticals and pesticides. For the PP mass balance, the amount of polypropylene in water with a size above 100 μm was quantified using TGA TED GCMS. Nile Red (NR) staining was used to estimate the microplastic concentration in particles per L in the DAF water samples (influent/effluent), to assess the effectiveness of the DAF to remove small MPs.

2.2.3. Analysis of solid fractions

For flake samples, the moisture content and surface contamination are determined. The flake size distribution is determined using a Retsch AS 200 Basic vibratory sieve shaker equipped with sieves according to the ISO 3310–1 and ASTM E11 standards. The type of plastic is determined on flake samples using a Thermo Scientific Antaris II FT-NIR (Fourier transform near-infrared) with an integrating sphere. A representative sample of 100 flakes is selected from a product or residue stream and the type of material is determined flake by flake. The composition of the sample is calculated based on the mass ratio. The mass ratio between flexible and rigid flakes is determined manually in a 10–20 g flake sample. The mass ratio between floating and sinking material is determined by adding water to a ∼ 50 g sample. The floating fraction is removed from the top and the heavy fraction is removed subsequently by filtration. Both fractions are dried in an oven at 95 °C for 4 h and the dry mass is determined afterwards.

3. Results & Discussion

3.1. Input material

Polypropylene bales of source-separated consumer packaging waste according to the DKR-324 specifications, implying a maximum of 6 wt% objects of which the main material is not PP. Manual characterization was used to determine the actual composition and is corrected for moisture content. The target objects (rigid PP) account for a total of 90.7 wt% with the remaining composition being made up from 0.7 wt% PP film, 4.1 wt% non-PP objects (*e.g.*, other plastics, paper, glass) and 4.5 wt% surface contamination. Although 90.7 wt% of the bale is composed of the target objects, the actual amount of recyclable PP is only 85.8 wt% because of labels, sleeves and caps (4.9 wt%). The sorted material was shredded, and the shredded material is used as the input for the mass balance of the washing process.

3.2. Mass balance for PP

A mass balance for polypropylene was made throughtout the washing process and calculated based on the amount of polypropylene at each step with corrections being made for moisture content, surface contamination and composition. The mass balance was calculated based on the polypropylene content of three types of samples: (intermediate) product, residue streams and water output. The (intermediate) product was collected before the cold wash, after the cold wash, after density separation and after hot wash. The collected residue streams are from the sinking fraction of the heavy object separator, the sinking fraction of the density separator, fines from the mechanical dryer (after cold wash and after hot wash) and the light fraction of the wind shifter. Water samples were collected after the wet grinder/friction washer combination and all other friction washers (three in total). The total quantified losses add up to 13.2 wt% with a product output of 85 wt% leaving 1.8 wt% unidentified (Fig. 2).

3.2.1. Cold wash

In the cold wash, polypropylene was lost at two locations: 1.8 wt% in the heavy fraction of the heavy object separator and 2.2 wt% in water from the wet grinder/friction washer. The presence of polypropylene in the sinking fraction could be due to the presence of additives (*e.g.*, $CaCO_3$ or TiO_2) increasing the density > 1 g/cm^3 or because of turbulence. Water sampled at the friction washer (sampling point 2) contained an average concentration of 0.67 ± 0.1 g/L PP particles > 100 μm (quantified using TGA TED GCMS). The 2.2 wt% PP particles that are lost at the friction washer are likely generated throughout the previous process steps, and released here, as this is the first step in the process where fines are removed through a 1.5 mm screen. Fines generation mainly occurs in the shredder and wet grinder due to the size reduction in these process steps. Size reduction steps are a compromise, bigger flake sizes lead to insufficient washing while smaller flake sizes increase the amount of fines generated and thereby losses.

3.2.2. Density separation

In the density separation step, 0.6 wt% of the polypropylene input was removed with the sinking fraction. This is again likely due to additives and turbulence. In the subsequent drying steps, 1.0 wt% polypropylene losses are found in the water from the friction washer. These

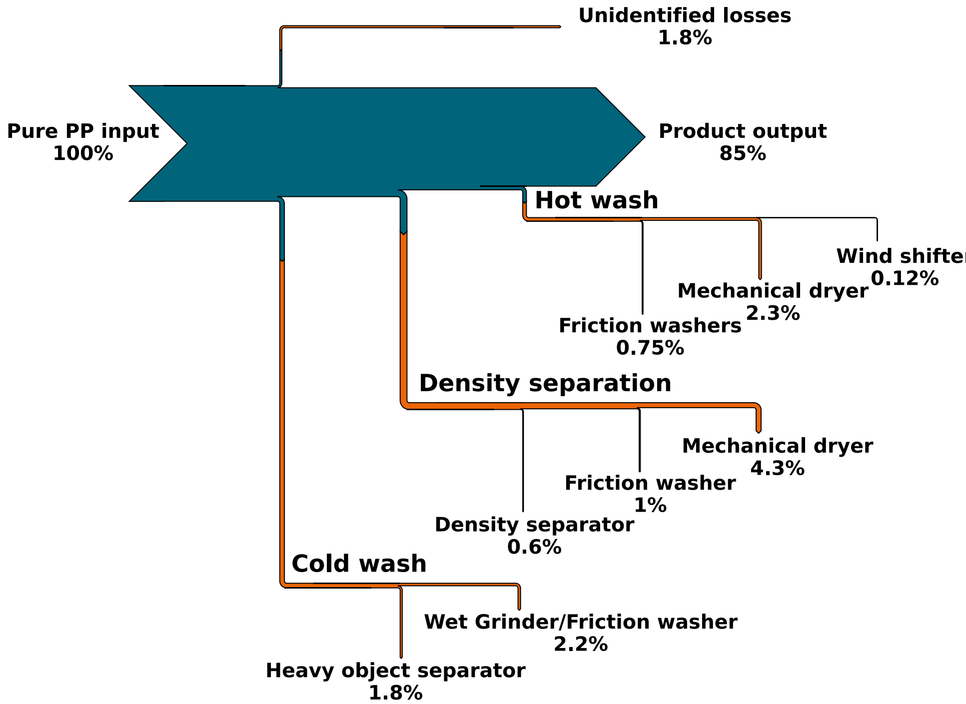

Fig. 2. Mass balance (in wt%) over the complete PP recycling process following the amount of polypropylene in each of the streams, hence the input is 100 wt% PP. Green represents the product stream and orange the residue streams.

losses in the water are calculated using the average concentration quantified using TGA TED GCMS (3.04 ± 2.1 g/L of PP particles above 100 µm, sampling point 6). The large variation between samples can be explained due to the pulsing nature of the friction washer combined with batch sampling. 4.3 wt% of input material is lost as fines at the mechanical dryer. This is the largest loss of a single process step and accounts for 29 wt% of the total losses.

3.2.3. Hot wash

More fines are generated due to friction in the hot wash. The complete mixture is released from the hot wash onto a friction washer where fines are removed with the water. The polypropylene content of the effluent from this friction washer is estimated at 0.61 wt%, a sample from the subsequent friction washer contains 0.14 wt% of polypropylene adding up to a total polypropylene loss of 0.75 wt%. Additionally, 2.3 wt % of losses are found in fines from the mechanical dryer and 0.12 wt% of rigid polypropylene in the light fraction of the wind shifter. In the fines from both mechanical drying steps, a total of 6.6 wt% of polypropylene losses are found, accounting for 44 wt% of the total losses. This shows the influence of mechanical drying steps on the polypropylene losses and the potential for mitigation at this process step will be discussed later.

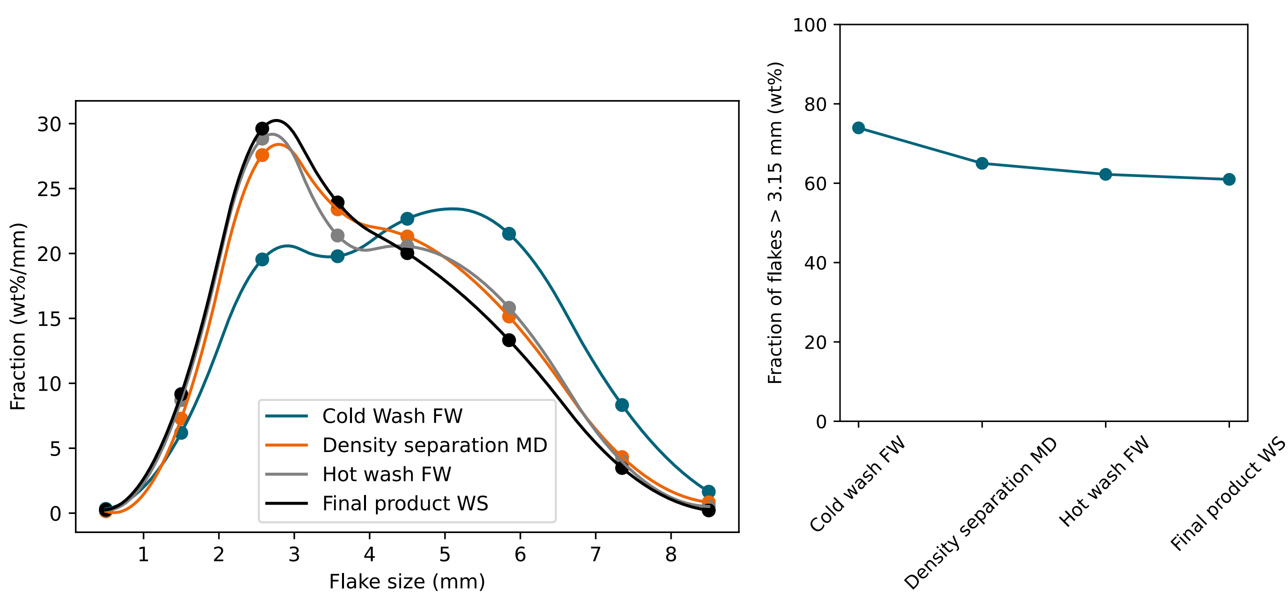

Fig. 3. Left: Flake size distribution based on mass fractions determined with a vibratory sieve shaker on product samples at 4 locations during the process. Right: Mass fraction of flakes > 3.15 mm on the same samples.

3.3. Flake size distribution

Most of the losses described above are in the form of small plastic particles that can pass through the screens of a friction washer or mechanical dryer. Fines generation largely depends on the applied friction in the process and can be visualised with the flake size distribution (Fig. 3, left). During the process, the mass fraction of bigger flakes decreases, while that of smaller flakes increases. Between the sample points, fines are removed in friction washers and mechanical drying steps which is not considered in this figure. This means that the actual mass fraction of fines (flakes < 3.15 mm) generated, is larger than the figure suggests. With screen sizes of 2.5 and 3 mm in the friction washer and mechanical dryer, respectively, the fraction of flakes > 3.15 mm gives an indication of the quantity of 'valuable' flakes (Fig. 3, right). The largest decrease of flakes > 3.15 mm (74 to 65 wt%) is observed between the sample after cold wash and the sample after density separation. This part of the process is composed of a density separator, friction washer and mechanical dryer and the biggest loss is found here at the mechanical dryer (4.3 wt%). This indicates a correlation between the losses at the mechanical dryer and the reduction in flake size and is studied further in the mitigation section. The sizes of fines that are removed at both mechanical drying steps are also determined.

3.4. Emissions

3.4.1. Emissions to air

3.4.1.1. Shredding and dry wash. The aerodynamic particle sizer (APS) was placed directly above the shredder to perform online measurements of the emitted particles (Fig. 4). Shredding caused a significant increase in the concentration of particles to air in all three particle size bins (see Fig. 4A).

Quartz filters were sampled from the same position to measure the concentration of PP using TGA TED GCMS. The PP concentration was higher during shredding (7.6 μg/m^3 for TSP) than during overnight measurements (0.5 μg/m^3 for TSP, see Fig. 4B). During shredding activity, the concentration of PP in the air corresponded to 13.9 wt% (PM1), 0.3 wt% (PM10) and 2.8 wt% (TSP) of the total particle count measured by APS. The low concentration of PP in airborne particulate matter is in line with this being the first processing step where most airborne particles originate from surface contamination. To investigate

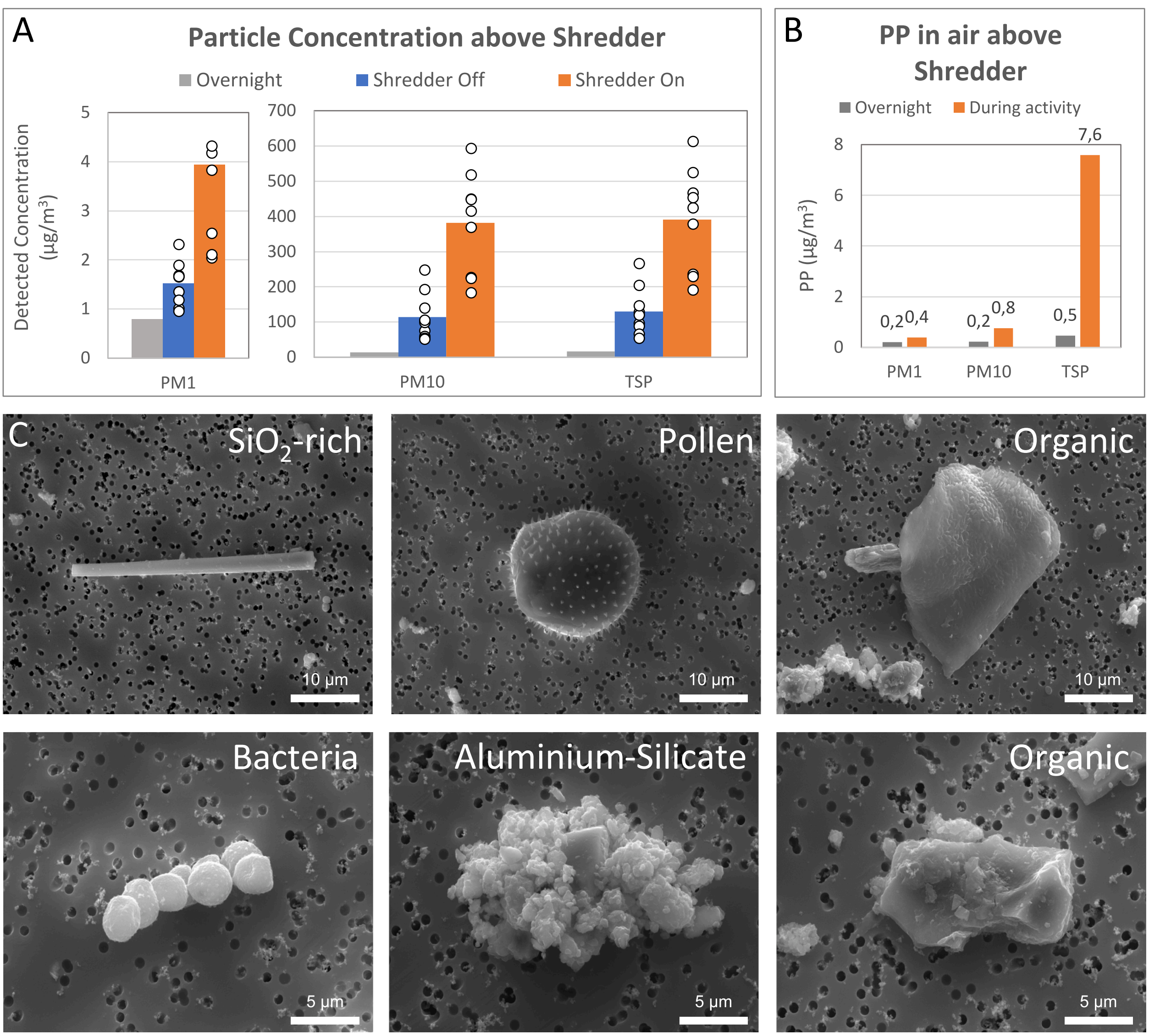

Fig. 4. Particle detection in air sampled above the shredder. A: Average detected concentration for particle sizes PM1, PM10 and TSP, measured overnight, during daytime activity and when shredder was actively being used. B: PP concentration (μg/m^3) in air above shredder determined by TGA TED GCMS C: illustrative SEM images of particles emitted during shred activities.

other types of emitted particles, SEM-EDX (energy dispersive X-ray) analysis was performed (Fig. 4C). Different types of particles were present, ranging from fibre-like particles with an elemental composition mainly consisting of silicon and oxygen (back scattered electrons (BSE) images and EDX spectra in S.I.) to irregularly shaped particles and bioaerosols. A range of different particles is to be expected in any recycling facility due to the heterogeneous material streams.

3.4.1.2. Cold wash. Air was sampled next to the cyclone outlet directly following the mechanical dryer, where the emission of PP was expected to be highest. Overnight at this location, very little PP was found (Fig. 5A). During the cold wash, the amount of PP in the air increased to 0.49 μg of PP/m^3/kg input material (Fig. 5B). Measurements were also taken at a background measurement location at ground level in the hall, at a distance of around 15 m from the cyclone. Assuming that, outside the direct influence of the cyclone, small PP particles spread homogenously throughout the hall, an estimation can be made as to the total PP emitted to air during the cold wash. When the mechanical dryer was on during the cold wash, 338 μg of PP (TSP) was emitted per kg processed material. During the cold wash, ongoing activity in the hall caused

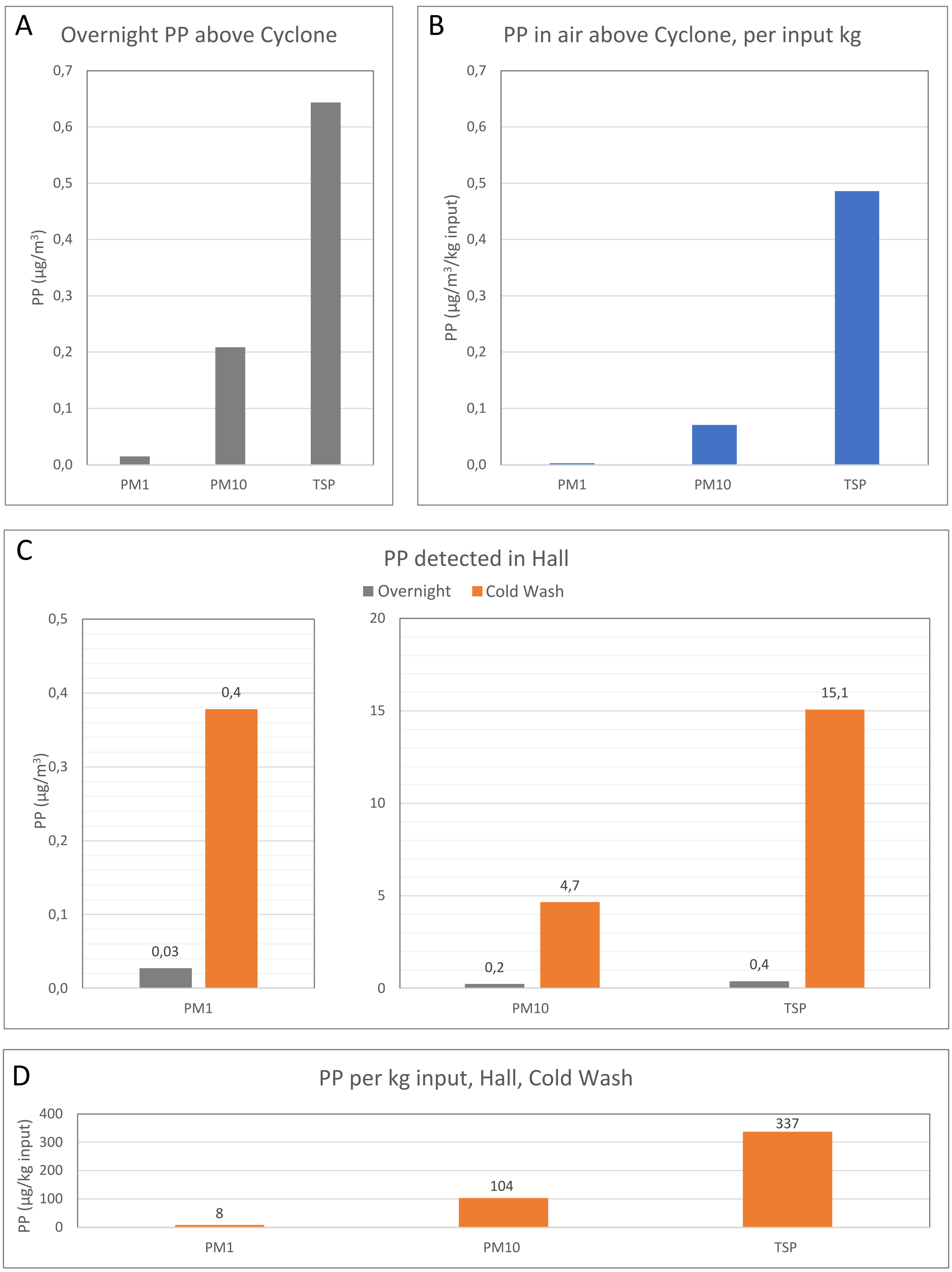

Fig. 5. A. Overnight background concentration of PP (μg/m^3) detected above cyclone. B. PP (μg/m^3) measured above cyclone during cold wash, per kg of input plastic. C. Overnight and cold wash concentration of PP (μg/m^3) measured at ground level. D. Estimated total PP (μg) emitted during cold wash, per kg of input plastic.

unpredictable ventilation. This value was therefore not corrected for hall refresh rate, meaning that the actual PP emission is likely higher. The 338 µg of PP emitted per kg of input material reads like a small mass fraction, but considering the particle size distribution the number of particles is vast. With the current estimated recycling amount of rigid PP in the Netherlands of 20 kton (PP input) this would result in an annual release of 6.6 kg of airborne microplastics. Assuming particles of 20 µm, this would amount to $1.7x10^{12}$ particles per year.

3.4.2. Emissions to water

3.4.2.1. Cold wash. During the cold wash, the majority of the surface contamination was removed from the plastic flakes, decreasing from 7.1 to 1.1 wt%. This surface contamination was transferred to the water phase as a mixture of organic and inorganic contaminants. The surface contamination, together with plastic fines, accumulated in the cold-water loop leading to an average TS concentration of 3.5 g/L (Sample 2).

At the heavy object separator (Sample 1), the material first came into contact with water, and soluble contaminants began to dissolve here. Using conductivity values as a proxy for the total dissolved solids (TDS), the conductivity increased from 0.5 mS/cm (tap water) to 2.5 mS/cm during the processing of 1500 kg of material. On the other hand, the conductivity in the cold-water loop that ran through the wet grinder and friction washer (Sample 2) increased from 0.5 mS/cm to 0.9 mS/cm only, during the processing of 800 kg (after 1000 kg part of the circulated water was pumped towards the DAF). Considering both the amount of material processed, and the water volume, this gave an increase of 0.44 µS/cm and 0.11 µS/cm for every kg of material processed in 1 m^3 of water for the heavy object separator and cold-water loop, respectively.

Non-soluble substances were also removed and transferred into the water as shown in the turbidity values. Hence, at the heavy object separator, there was a steady increase in turbidity, as no fresh water was added here during washing. For the cold sump, a fraction of the circulated water was pumped to the DAF after 1000 kg of material was processed (dotted line), with the volume stabilized by addition of fresh water to the cold tank. This change in process conditions resulted in a reduction in contamination level reflected in the decreased turbidity of the water in the cold sump. After the wet grinder and friction washer, the polypropylene stream was purified in a density separator (Sample 5). The level of contamination of the water inside the density separator was monitored by turbidity/conductivity and showed no significant change.

Samples from the first (Sample 2) and second (Sample 6) friction washers after the wet grinder and float-sink, respectively, were taken in triplicate over a period of 1 h. Whilst the first friction washer had TS values in a relatively narrow range, between 3.1 and 3.7 g/L (n = 5), the second friction washer caused water and fines to be forced out in a pulsing effect, resulting in TS values ranging between 1.4 and 4.7 g/L (n = 5). TGA/DSC analysis on the TS obtained from these samples showed that the sample after the first friction washer had a much higher (non-plastic) organic composition than that of the second friction washer. This is to be expected as in the first friction washer most surface contamination is removed including food residue. Using the TGA we estimated the average plastic proportion across all aliquots of Sample 2 to be 49 wt% (n = 9) and 81 wt% (n = 15) for Sample 6. This is compared to the DSC which estimated Sample 2 to be 56 wt% (n = 8) and Sample 6 to be 78 wt% (n = 9).

In the water loop, water was continuously passed through a vibrating screen, which acts as the first level of treatment before the DAF (see Fig. 1 and S.I.). The cold wash influent DAF sample, taken during the cold washing process, is most representative in demonstrating the role of the vibrating screen on the overall water quality. From the first friction washer (Sample 2), which used water recirculated through the cold loop, to this DAF influent sample, the TS concentration within the water decreased from approx. 3.5 g/L to 1.4 g/L. The TGA/DSC data for these samples show little variation in the ash content and a slight decrease in the (non-plastic) organics; however, the plastic content decreases from approx. 1.9 g/L to 0.05 g/L, representing a reduction of 97 wt%. The vibrating screen removes particles > 500 µm therefore, most dissolved organic and inorganic pollutants can pass through this. The solid plastic particles, however, are readily removed in this step, meaning that only the smallest MPs will be sent to the DAF, resulting in a significant reduction in plastic mass in these DAF influent samples (thermograms in S.I).

After the cold wash, samples were taken and analysed for PFAS and micropollutants. None of the targeted PFAS were detected and only small amounts of micropollutants. Acesulfame, an artificial sweetener, is one of the micropollutants detected at a concentration of 12 µg/L. This artificial sweetener is expected due to its ubiquitous use in beverages. Details can be found in the S.I.

3.4.2.2. Dissolved air floatation. DAF water treatment was used for wastewater from both the cold and hot wash process. Characteristics of the influent and effluent of the DAF are summarized in Table 1.

Influent (sample 4a) and effluent (sample 4b) of the DAF were collected (Fig. 1) and analysed for TS and TSS. Suspended solids can be removed in the DAF unit and is a relevant parameter to assess the effectivity of the process.

Higher TSS concentrations were observed when the DAF was operated without water refreshment compared to partial water refreshment (Table 1). The DAF removed ~ 96 wt% in both samples. Due to the addition of tap water during the cold wash process to replace the lost effluent water, a lower effluent concentration was achieved.

The removal of microplastics by the DAF was also quantified. Acknowledging the low small sample volume analysed, when extrapolated we estimate that the influent of the DAF contained approximately 1.9 and 1.6 million MP particles/L without and with continuous refreshing, respectively. The DAF was able to remove ~ 62 wt% and ~ 57 wt% of MP particles by number, respectively, resulting in effluent counts of 0.73 (without refresh) and 0.70 (with refresh) million MP particles/L. TGA TED GCMS showed that 97 to 99 % of the total mass of PP in the cold wash wastewater was removed, resulting in effluent concentrations of 0.4 mg/L (without refresh) and 0.05 mg/L (with refresh). We estimate that with water refresh, approximately 0.8 to 1.0 mg/kg (MP/processed PP) was emitted in the effluent. Correspondingly, we estimate that this would be around 13 million MP particles.

A study by Brown et al. (2023) analysed wastewater from a plastic recycling facility before and after 50 µm particle filters were placed on

Table 1
Analytical data of the influent and effluent of the dissolved air flotation unit during the PP recycling process. A: Batch treatment of cold wash water without refreshment of the washing water during processing. B: Treatment of cold wash water with continuous water refreshment. C: Batch treatment of water from a hot wash cycle.

	A: Cold wash		B: Cold wash with refresh		C: Hot wash	
Quantity	*Influent*	*Effluent*	*Influent*	*Effluent*	*Influent*	*Effluent*
Total solids [mg/l]	2610	1367	1422	1024	2578	1875
Total suspended solids [mg/L]	1243	50	499	19	385	111
MP conc. Nile Red [millions/L]	1.93	0.73	1.62	0.70	0.73	0.42
MP conc. TED GC MS [mg/L]	15.18	0.40	14.24	0.05	5.00	0.13
total-P [mg/L]	19.49	14.62	14.45	11.48	27.33	23.35
total-N [mg/L]	68.70	33.00	39.40	26.10	9.23	10.65
DOC [mg/L]	59.3	50.3	49.0	25.5	231.8	152.0
COD [mg/L]			3366	329	3334	1558
Turbidity [NTU]	934	194	592	111	379	130

outlet locations. As expected, the filtration step was highly effective at removing MPs greater than the mesh size. The highest amount measured 1.01 million MP particles/L after the 50 μm additional filter. This indicates that the DAF is at least as efficient as a 50 μm filter but with much less risk of clogging.

The disparity in removal efficiencies determined by mass and particle number can be explained by the more effective removal of larger microplastics by the DAF as confirmed by the particle size distribution of the DAF influent and effluent (see S.I. for Mastersizer and Nile Red based size distribution). The results indicate that approximately half of the particles below 100 μm are removed during the DAF treatment. This is demonstrated with only a slight change in the average Feret diameter of the particles observed, with the influent average decreasing from ~ 21.1 μm to ~ 18.7 μm. The overall plastic mass discharged is relatively small but represents a substantial absolute count of MPs which require adequate post treatment.

Water in the hot wash process was continuously looped with filtration over a vibrating screen, without any refresh of clean water. The turbidity and COD both show that the level of contamination is increasing with more material being processed. The presence of KOH and surfactants increases the COD level to 7.6 g/L before any polypropylene washing takes place and is as reference value subtracted from all data points. The conductivity and pH are not suitable to assess the level of contamination in the wastewater as these levels increase by the presence of ~ 0.16 wt% KOH. Under normal operation conditions, this water is continuously refreshed, but with processing only 1200 kg this was not necessary.

During the hot wash, the presence of KOH and surfactants resulted in substantially more dissolved solids when compared to the cold wash process. Before DAF treatment, the water was neutralized with H_2SO_4. As the amount of TSS is much lower in hot wash wastewater than in cold wash wastewater, we have focussed on the DAF efficiency for the treatment of cold wash wastewater. Details for the hot wash can be found in the S.I.

3.4.3. Residue streams

Besides air and water emissions, the residue streams from a recycling process need to be considered as emissions. Generally, these residue streams are incinerated or landfilled, however the composition of some residue streams allows for more valuable uses. More information on the potential use of these streams can be found in the S.I.

4. Mitigation of losses and emissions

In several parts of the recycling process significant losses and emissions are identified. Possible reduction of these losses and emissions are investigated with 3 small-scale trials in specific sections of the process.

4.1. Dry washing

The first mitigation is to pass the shredder output through a loop dryer. This reduces the amount of surface contamination and thereby emissions to the cold wash wastewater. Rotation speeds of 700 and 1100 rpm (tip speed 3.4 and 5.4 m/s, respectively) were used and surface contamination and moisture in both the material and residue were assessed.

The shredder output is composed of 7.1 wt% surface contamination and 3.1 wt% moisture leaving 89.8 wt% potential product (Fig. 6). After the 700 rpm loop dryer, surface contamination is reduced to 5.1 wt% and moisture to 2.3 wt%. However, this also leads to a loss of product output as the residue from the loop dryer contain 47.5 wt% product. At 1100 rpm, the moisture (0.7 wt%) and surface contamination (2.0 wt%) is reduced even further, however, the increased rotation speed causes even more product loss as here 57.3 wt% product material is found in the residue from the loop dryer.

At 700 rpm, 94 wt% of the target ended up in the product stream, whereas at 1100 rpm this was only 87 wt%. The remaining material is lost as fines together with surface contamination and moisture. We conclude that the removal of surface contamination using a loop dryer before washing is not a suitable method for the relatively brittle PP as it would lead to increased losses. Filtration and DAF treatment of the cold wash wastewater seem a better option.

4.2. Wet grinder and friction washer

The initial flake size distribution is governed by the wet grinder. The impact of wet grinder screen size (15 vs 25 mm) and knife age (old *vs* new), and friction washer use on the flake size distribution was studied. As expected, the larger wet grinder screen results in a significant shift in the distribution to larger flakes (Fig. 7). This minimises flakes < 3.15 mm and will likely reduce losses in the later stages of the recycling process. Conversely, a relatively large fraction of the flakes is > 8 mm and a reduced efficiency of washing is possible. An increased washing efficiency with smaller flake sizes is not tested here but is expected and

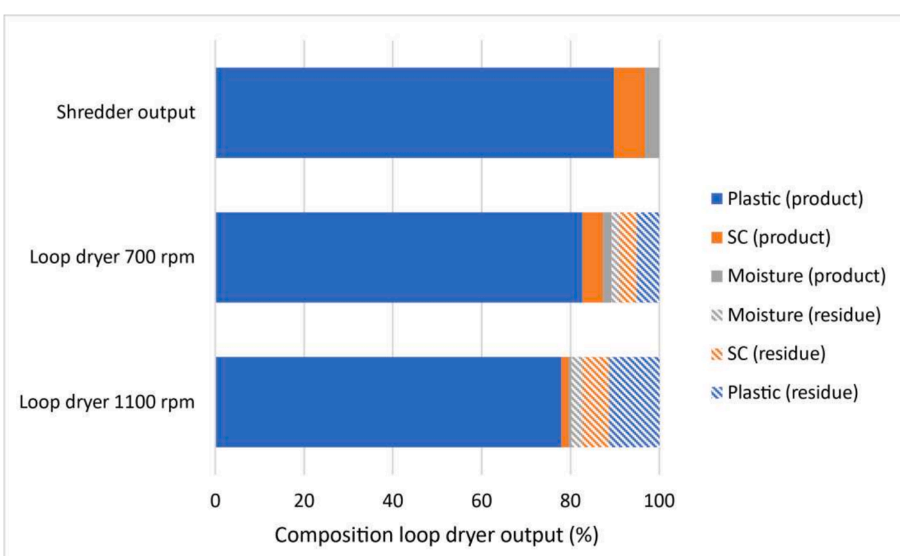

Fig. 6. Composition of product and residue output before and after loop dryer at 2 speeds (700 and 1100 rpm).

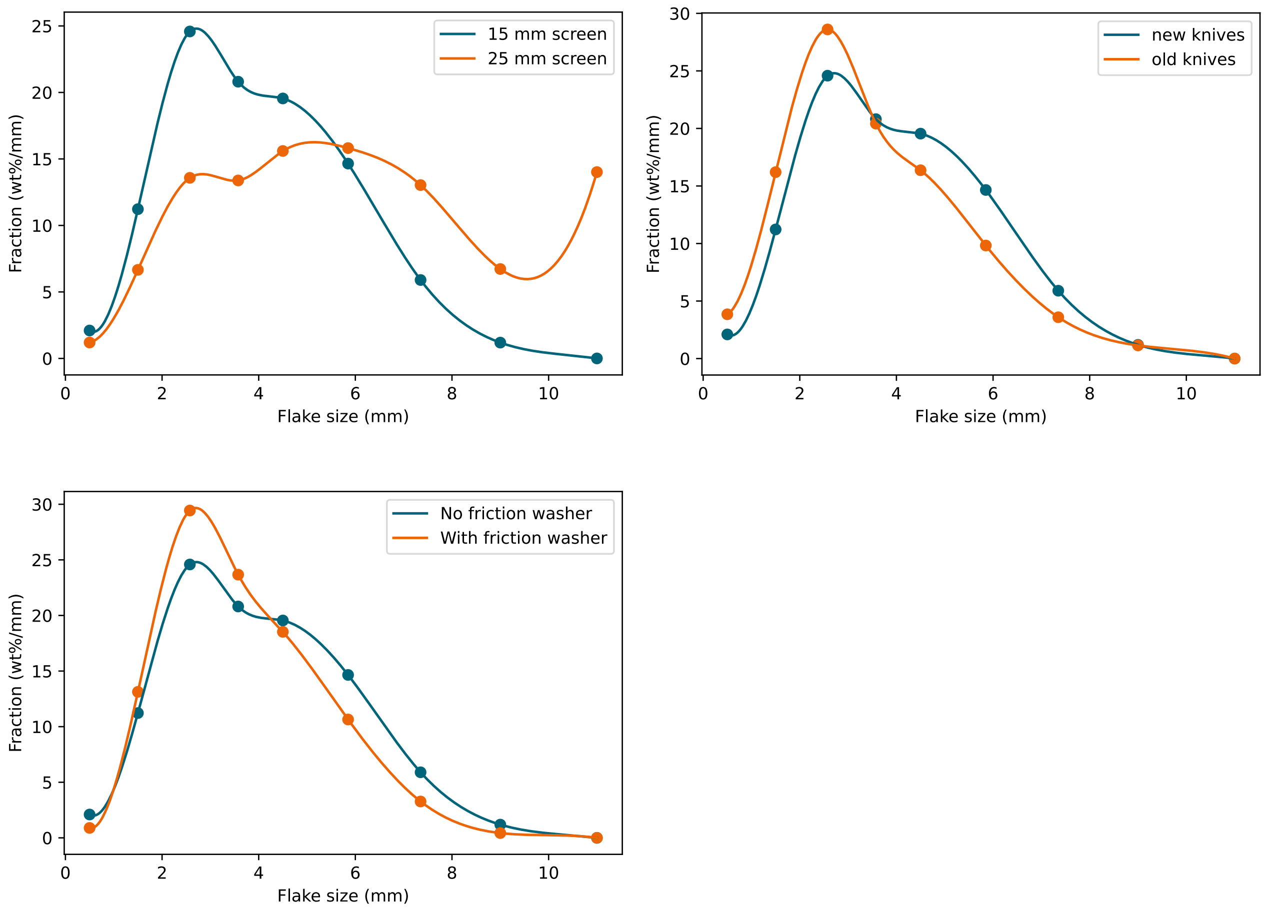

Fig. 7. Flake size distributions under several process settings. Reference settings (blue): 15 mm screen, new knives, no friction washer. The lines serve as a guide to the eye.

mentioned by friction washer producers. The flake size distribution is only slightly affected by knife age. The fraction of flakes < 3.15 mm is larger with worn knives showing that timely replacement of knives can contribute to the minimization of losses in polypropylene recycling. In the friction washer, part of the polypropylene material is separated through the screen and ends up in the wastewater. However, the fraction of flakes < 3.15 mm increases after the friction washer at the expense of the fraction > 4 mm. The forces inside the friction washer lead to the formation of fines as well. It is not realistic to skip the friction washer as it plays a key role in the cleaning process.

4.3. Mechanical dryer

The majority of polypropylene losses (6.6 wt%) occur as fines in the mechanical drying step. These are partially present in the product stream going into the mechanical dryer and are partially generated in this step. The impact of mechanical drying on polypropylene was studied by processing clean polypropylene material at four different rotation speeds: 885, 1121, 1298 and 1475 rpm (tip speed 39, 49, 57 and 65 m/s, and relative G 408, 654, 877 and 1133, respectively). The material follows the same process as the first part of the hot wash process (hot washer, 2x friction washer, mechanical dryer) only not as a batch process, but with continuous flow of 400 kg/h plastic and 25 m^3/h water at 20 °C. At the 4 rotation speeds, the drying efficiency is very similar and all < 0.2 wt% which is sufficient for further processing. A linear correlation is found between the losses and the relative centrifugal forces on the material and shows a 4x increase from the slowest to the highest speed (from 1.0 to 4.0 wt%).

The correlation between the losses from the mechanical dryer and the impact on the flake size distribution discussed in the mass balance is further investigated here. The flake size distribution changes significantly between the lowest speed (885 rpm) and the highest speed (1475 rpm) (Fig. 8). The mass fraction of flakes > 3.15 mm is 86 wt% for the input and reduces to between 80, and 54 wt% for the lowest and highest speeds, respectively. This mitigation experiment shows that a substantial minimization of losses and emissions can be made in the drying step. Optimization of the mechanical dryer speed can reduce losses, but also different ways of drying (*e.g.* thermal drying) need further investigation.

Airborne PP concentration was measured above the output cyclone of the mechanical dryer during these tests. The mechanical dryer outputs higher volumes of air at higher speeds, which was accounted for, leading to a final value for PP concentration measured in mg of PP release per 200 kg of input plastic.

The measurements show an increased emission of total PP (TSP) from 0.55 to 4.20 mg/kg plastic as the dryer speed increases. The PM1 and PM10 fractions do not show consistent changes with mechanical dryer speed, suggesting that adjusting the mechanical dryer speed only has an influence on the particle generation of particles above 10 μm.

4.4. Other potential mitigations

As mentioned in section 3.4.1 MPs and other small particles can become airborne in some processes in PP recycling. The majority of these can be captured by using dust filtration units on cyclones in the process and a general air filtration system for the building.

Additional treatment after the DAF can be considered to limit the emissions of MPs further via the PP recycling effluent. Low-pressure membrane filtration processes, such as micro- (MF) and ultrafiltration

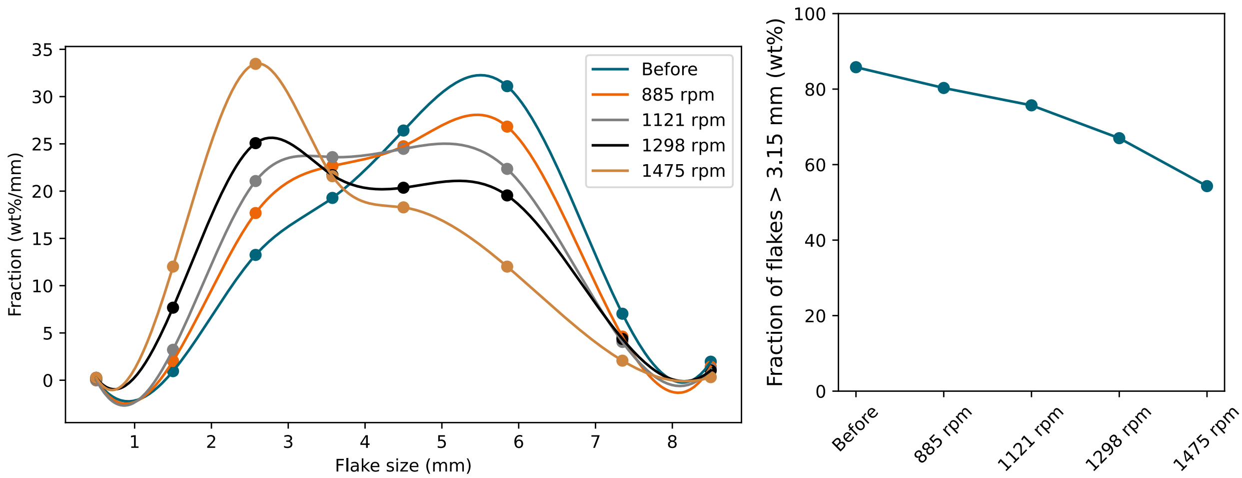

Fig. 8. Flake size distributions after mechanical dryer at 4 different speeds (left) and mass fraction of flakes > 3.15 mm (right). The lines serve as a guide to the eye.

(UF), can remove MPs with particle sizes up to ~ 20 nm. If membrane filtration is considered, implementing membrane bioreactors (MBRs) may be a more advantageous option for plastic recycling wastewater (Poerio, Piacentini, and Mazzei 2019; Xiao et al. 2019). MBRs are hybrid systems that combine membrane filtration with biological wastewater treatment. While the membrane filtration step allows for the removal of suspended particles, the aerated biological process can remove nutrients and organics, including certain micropollutants such as surfactants and pharmaceuticals (Goswami et al. 2018). Previous research concluded that PP microplastics have a minor influence on the biological performance of MBRs (Wang et al. 2022). Additionally, high removal of MPs by MBR systems has been reported (Talvitie et al. 2017; Bayo, López-Castellanos, and Olmos 2020), making them potentially interesting as an additional treatment step. Further research into using MBRs as additional treatment following the DAF is still required to understand its full applicability in PP recycling wastewater treatment. If higher water quality standards are needed, implementing high-pressure filtration membrane filtration processes, such as nanofiltration (NF) and reverse osmosis (RO), could be considered. While this would lead to high-quality effluent, such systems will come with substantial capital and operational expenses. Therefore, water recycling before treatment should be utilised as much as possible in the recycling process itself.

5. Conclusions

We have shown that in a typical high-quality post-consumer polypropylene recycling process an overall PP yield of 85 wt% is achieved. PP losses are largest at the two mechanical dryer steps (4.3 wt% after cold wash and 2.3 wt% after hot wash) while additional substantial losses occur in the friction washers (a total of 4.0 wt%). The flake size distribution of the product shifts to lower flake sizes throughout the process and the smaller flakes are removed from the process via residue streams or water. The dry mass of all non-aqueous residue streams is ~ 191 kg for every 1000 kg of input material whilst ~ 3.9 wt% of the PP input is lost as microplastics in the wastewater. Emissions to air were 330 $\mu g_{PP}/kg_{input}$ at the mechanical dryer.

We have also investigated ways to mitigate losses and emissions. Ensuring that wet grinder knives are regularly sharpened or replaced leads to less flakes < 3.15 mm. The mechanical drying process can be improved by lowering the centrifugal speed which the generated microplastics in this step from 4 wt% to 1 wt% without minimal effect on the moisture content. The DAF is capable of 97–99 % mass-based removal of microplastics. Those that are not removed are typically small in size thus a large number of small microplastics are still emitted. When also considering the vibrating screens the mass-based removal of microplastics in wastewater (screen + DAF) achieves a reduction from 1900 mg/L (DSC/TGA) to 0.05 mg/L (TGA TED GCMS) – a removal of > 99.99 wt%. It is not effective for PP recycling to include a dry washing step at the beginning of the process to reduce the surface contamination as it increases the creation of microplastics significantly. There are also a number of options to avoid or reduce losses and emissions that haven't been experimentally investigated. The dry residues are rich in plastic and may be suitable for cascade recycling through *e.g.* pyrolysis or even mechanical recycling. The residues containing large amounts of organic materials are potentially suitable for digestion. The emission to air can be reduced by using air filtration units at locations where MPs are generated.

Apart from making sure that plastic packaging waste is being collected and sorted for recycling, in order to actually have it recycled, we recommend using more quality control along the whole process and adjust the process accordingly. As a starting point, the presented TGA, TGA TED GCMS, and DSC can be used. In addition, one should investigate the options for cascade recycling for plastic packaging waste.

CRediT authorship contribution statement

Johann B. Kasper: Writing – review & editing, Writing – original draft, Visualization, Validation, Investigation, Formal analysis. **Luke A. Parker:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis. **Sander Postema:** . **Elena M. Höppener:** Writing – original draft, Visualization, Investigation, Formal analysis. **Alexandra H. Leighton:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Formal analysis. **Alexander M.D. Finnegan:** Writing – review & editing, Writing – original draft, Validation, Investigation, Formal analysis. **Sam B. Rutten:** Writing – original draft, Visualization. **José Nijman:** . **Amanda Larasati:** Investigation, Formal analysis. **André C.C. Soares:** Investigation, Formal analysis. **Marcel C.P. van Eijk:** Writing – review & editing, Supervision, Project administration, Investigation, Funding acquisition, Conceptualization.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2024.11.029.

Data availability

The used data is shared in the Supplementary Information.

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