Preliminary Study on Low-Density Polystyrene Microplastics Bead Removal from Drinking Water by Coagulation-Flocculation and Sedimentation

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Key words: Microplastics, Drinking water, Coagulation-flocculation, Floc breakage

Abstract

Microplastics (MPs), sized ~150 µm, have been found in tap water at levels of ~ 5 particles/L, suggesting that water treatment plants are not effectively removing MPs. Therefore, there is an urgent need to evaluate their fate in drinking water treatment processes. Coagulation-flocculation and sedimentation are applied in water treatment to primarily decrease turbidity, and MPs contribute to water turbidity. This study focuses on the removal of polystyrene (PS) beads of 100 µm with density 1.04-1.06 g/cm^3. The low-density PS beads offer a removal challenge because they have similar density to the media. The effects of initial water pH and stirring speed on MPs removal by coagulation-flocculation and sedimentation were studied. The most effective conditions found for removing the PS beads from water, that led to removal rates up to 98.9 ± 0.94 %, were 3.4 mg Al/L of coagulant, pH 5, flocculation time of 7 min and sedimentation time of 30 min. For the first time, floc breakage and regrowth following the addition of Al, has shown to favour the removal of the PS beads. Based on this research, coagulation-flocculation can play a very important role in removing MPs during drinking water treatment.

Keywords: aluminium sulphate, polystyrene; microbead; water treatment; floc breakage

1. Introduction

Microplastics (MPs) have attracted great attention globally. At present, the investigation of microplastic pollution mainly focuses on the marine environment (Michida et al. 2019; Jones et al. 2019; Li et al. 2020; Kumar et al. 2021). As a relatively new type of pollutant, extensive attention has been paid to its occurrence, distribution, abundance, separation and identification methods, adsorption and desorption mechanisms, and ecotoxicological effects in current research, and MPs have been gradually detected in freshwater (Zhang et al. 2021; Zhao et al. 2021; Frank et al. 2021; Li et al. 2020; Christensen et al. 2020). Freshwater is abstracted and treated for producing drinking water. In this process, coagulation-flocculation-sedimentation is the main step for removing particulate matter in drinking water treatment plants (DWTP). However, the removal of MPs in this key step to produce drinking water has received little attention. In the UK, coagulation-flocculation stages are usually combined with pre-ozonation, sand filtration and granular activated carbon contactors. Also, sedimentation is a worldwide technique for water treatment and an important step to prevent the subsequent overload of filters.

The percentage of samples from DWTP containing MPs ranges from 24 % to 100 % and the MPs content from below the limit of detection to 1247 MPs/L across studies (Danopoulos et al. 2020). When finding MPs in the treated water, for accurate quantification, it is important to work with large sampling volumes specially when the concentration of MPs is low (Zihajahomi et al. 2017).

The variety of MPs in sources of drinking water is diverse. Among them, PS is one of the most abundant types of MPs in freshwater globally (13 %) (Li et al. 2020). It is used in rigid packaging and construction material (British Plastics Federation 2021a), among other uses. In the UK, the Water Industry Research (UKWIR) found that the most common MPs in DWTP are PS and Acrylonitrile Butadiene Styrene (ABS) (Ball et al. 2019). Specifically, in raw water where the content was ~ 113 MPs/L, after treatment, the water still contained 2-27 MPs/L (Ball et al. 2019). This shows that the current drinking water treatment processes need to improve.

In the production of drinking water from a river with initial concentration of 6614 ± 1132 MPs/L, the removal efficiency of conventional treatment processes (including coagulation/flocculation, sedimentation and sand filtration) was about 58.9-70.5 % (Wang et al. 2020). There, MPs > 10μm were removed with 50.7-60.6 % efficiencies which was greater than for the rest of MPs (Wang et al. 2020). Polyacrlyamide (PAM) was the coagulant used and it led to large amount of PAM in the sludge of the sedimentation tanks (Wang et al. 2020). Currently, there are no legal restrictions on the MPs content in drinking water, and there is no treatment technology that directly targets the removal of MPs.

Skaf et al. (2020) found high removal efficiency (99 %) of kaolin flocs using aluminium at pH 6.5 by coagulation-flocculation and sedimentation. Because zeta potential of polyethylene beads was similar to that of kaolin in water adjusted to pH 4-7, these authors assumed that MP beads could be removed under their study conditions. However, because there are a large variety of MP types, sizes and densities (around 1 g/cm^3), and Kaolin density is about 2.65 g/cm^3, their results cannot be generalized.

When a variety of coagulants (iron, aluminium and polyamine-based) was used to study coagulation-flocculation as a tertiary wastewater treatment process to treat secondary sewage containing microplastics (~ 10 μm) (Rajala et al. 2020), the optimal microplastic removal (i.e. 93 %) was achieved with polyaluminum chloride as coagulant. Both Shahi et al. (2020) and Lapointe et al. (2020) indicated that different plastic types, sizes, densities, solution environments and coagulants have an impact on the flocculation effect, and highlighted that further research is needed.

Among the studies on treatment of MPs through coagulation-flocculation, some focused on MPs of different polymers such as polyethylene, polypropylene, polyvinyl chloride, or a mixed solution of MPs (Wang et al. 2020; Skaf et al. 2020). However, the study focusing on the treatment of low-density PS MPs as a pollutant by coagulation-flocculation and sedimentation has not been reported and has special interest. PS is rigid and brittle (British Plastics Federation 2021b) which are properties that favour its degradation. PS' photo resistance outdoors is competitive; however, it can change depending on its additives (e.g. metal complexes, benzophenone or Ethylene Propylene Diene Monomer (EPDM)) (Zweifel et al. 2012). Photooxidation is a predominant weathering process that will favour the formation of plastic debris (Wypych 2018). These fragments can diffuse to freshwater used for the production of drinking water.

The density of PS (1.04-1.06 g/cm^3) (Cincinelli et al. 2020) is close to that of natural water and, hence, they may result in PS particles in suspension or floating in water. Therefore, they pose a greater potential risk than plastics that settle during drinking water treatment. In addition, it is recognised that flocs can be broken after flocculation in water treatment plants due to potential high shear zones, leading to low removal efficiency of the flocs. However, it is known that restoring the previous low shear conditions, flocs can grow back to the previous size (Yukselen and Gregory 2004). Considering the low density of the PS particles, we were interested on what the effect of breakage and regrowth of flocs on their removal as well. The aim of this paper was to preliminary investigate the potential impacts of coagulation-flocculation and sedimentation on low-density 100 μm PS microbeads, which were spiked in natural and tap waters.

2. Materials and Methods

2.1 Materials

All chemical reagents used were analytical grade and obtained from Sigma-Aldrich (UK), including $Al_2(SO_4)_3 \cdot 18H_2O$, Na_2CO_3, NaCl, 37 % HCl, NaOH and kaolin. PS beads (100 μm, 1.04-1.06 g/cm^3) were purchased from Dongguan Xingwang Plastics Co., Ltd. Water used in this research was tap water (pH 7.7±0.1; turbidity: 0.2±0.1 NTU; absorbance at 254 nm (UV-254) was 0.177±0.001 for the breakage and regrowth process and Regent's Park pond water (pH 8.4±0.1; turbidity: 0.8±0.3 NTU; UV-254, 0.64±0.59) for other tests. All MPs stock solutions were prepared at 5 g/L and were stored in the dark at 4 °C.

2.2 Coagulation-flocculation and sedimentation tests

A PB-900 programmable Jar tester (Phips & Bird, USA) was used with a total of six beakers (1 L) with one flat-bladed mixer with diameter (d) = 0.0504 m. PS beads (100 μm) stock solutions (Dongguan Xingwang Plastics Co., Ltd., China) were added to Regent's Park pond water at 10 mg/L. For imaging and MPs counting purposes only, MPs were dyed with red acrylic paint prior to the coagulation-flocculation experiment; the optimization of the treatment steps was carried out with undyed beads.

The coagulant used was $Al_2(SO_4)_3 \cdot 18H_2O$ at 3.4 mg Al/ L based on previous work (Yu et al. 2010). During coagulation, the solution pH was adjusted with 0.1 M $NaHCO_3$, and the pH of the untreated water (before adding the coagulant) was adjusted to 1, 3, 5, 7, 12 and 13 by adding 0.1 M HCl or 0.1M NaOH (Fisher Scientific).

To investigate the effect of flocculation mixing speed and sedimentation time, coagulation speed was maintained at 300 rpm (G = 345 s^{-1}) for 1 min, and then the mixing intensity was decrease to seven individual test speeds (50, 100, 150, 200, 250 rpm; G = 23, 66, 122, 188, 263 s^{-1}) for 7 min of flocculation (Zhou et al. 2021). The mixing intensities were converted into velocity gradient using Equation (1) (Rushton et al. 1950) and Equation (2) (Camp 1954):

$$P = N_p \rho N^3 d^5 \tag{1}$$

$$G = \sqrt{\frac{P}{\mu V}} \tag{2}$$

Where P is the power requirement (W), N is the rotational speed of the impeller (rpm), N_p is the power number (dimensionless), d is the impeller diameter (m), V is the tank volume (m^3), and ρ and μ are the density and absolute viscosity of the water (kg/m.s) at temperature 'T'. The following parameters were used: $N_p = 7$ (Cornwell and Bishop 1983); $V = 8x10^{-4}$ m^3; water temperature 25 ºC; $\rho = 1x10^3$ kg/m^3; $\mu = 0.0091$ kg/m.s; d = 0.0504 m. Finally, the sedimentation step spanned for 30 min (Ma et al. 2019a). All experiments were carried out in triplicate. The effect of the duration of the different flocculation speed was investigated from 100 s to 800 s (Ma et al. 2019a) with increments of 100 s. In all tests, coagulation speed was set at 300 rpm (G = 345 s^{-1}) for 1 min. Sedimentation time was screened and the optimum time, based on maximum number of MPs separated from solution and counted, was selected.

2.3 Floc breakage and re-growth experiment

In a dynamic test, the PDA 3000, Photometric Dispersion Analyzer (Rank Brothers Ltd., Cambridge) (Figure S1 in Supplementary Information) was sampled every two seconds. Kaolin (50 mg/L) and PS MPs (10 mg/L) were prepared in 800 mL of tap water (central London). Coagulant (3.4 mg Al/L) was added to the raw water as specified in Section 2.2. The pH of the suspension was adjusted to 5 with 0.1M HCl and stirred at 300 rpm (G = 345 s^{-1}) for 1 min. Then, the stirring speed was reduced to 50 rpm (G = 23 s^{-1}) for 10 min. Next, it was increased to 300 rpm (G = 345 s^{-1}) for 1 min to break the flocs and then back to 50 rpm (G = 23 s^{-1}) for 10 min for flocs re-growth. In the case of the addition of coagulant for a second time, the additional dosage of alum (0.8 mg/L) was added into the stirred suspension during the floc breakage phase (Yu et al. 2010). All experiments were carried out in triplicate.

2.4 Quantification of MPs

For the quantification of MPs, an optical microscope (model Euromex Oxion Material Science, Netherlands) and Countess^{TM} cell counting chamber slides (C10228, Thermo Fisher Scientific, UK) were used for the visual inspection of MPs with microscopy. A glass graduated pipette (5 mL) was used to draw the diluent (0.85 % NaCl aqueous solution) into a test tube. An aliquot (1 mL) of water sample with suspended MPs was taken (using polypropylene micropipette tips) and it was added to a glass test tube. The suspension was shaken to resuspend the MPs adhered inside the test tube. Then, the test tube was manually shaken several times. An aliquot (1 mL) of the tube was placed in between the flat counting chamber and the cover glass, allowing the suspension to flow naturally into the counting chamber for up to 2 min. The concentration of MPs in the suspension was determined by visually counting the MPs with the optical microscope and the volume of sample was taken into account. The MPs percentage removal was obtained from the difference between the concentration of MPs before and after the treatment and was normalised by the starting concentration of MPs.

3. Results and Discussion

In this work, PS beads of 100 µm were selected because this size belongs to a relatively abundant size fraction (9.7 %) in the final clarifier effluent (Wolff et al. 2021). This size range has shown to be toxic in fish (Ding et al. 2020) and PS particles (0.2 µm), although smaller than the ones studied here, were observed to cross the membrane in red blood cells with microscopy (Rothen-Rutishauser et al. 2006).

This study used spiked MPs at 10 mg/L which is greater contamination than in the freshwater. The study concentration stems from the need to carry out accurate mass measurements and compare initial and final concentrations after the effect of coagulation, flocculation and sedimentation, while using an analytical balance for the preparation of solutions with MPs and working with 1 L jars. Given that, unlike molecules and ions, microplastics only become suspended in water (and not dissolved in water), preparing a concentrated solution for further dilution would entail uncertainty on the concentration of MPs in the working solutions. Therefore, to maintain low uncertainty in the MP levels, the authors opted by spiking MPs at levels greater than those in freshwater. The disadvantage of this is that there may be agglomeration of PS MPs in solution, which will be minimised by the stirring in the jars. The agglomeration and location of the beads during the clarification process, including in the floc are illustrated in Figure S2. The MPs in Figure S2 were dyed to illustrate their distribution in the study treatment. Figure 1 shows flocs sampled directly from the sludge after sedimentation without changing properties of the flocs.

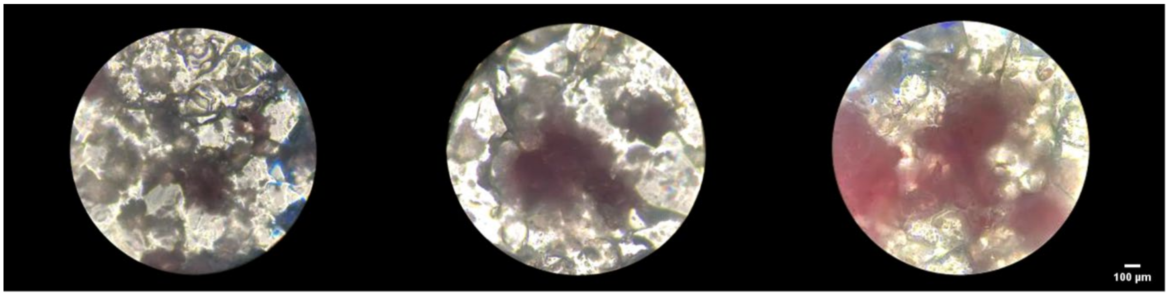

Fig. 1 Flocs including PS microplastics (dyed in pink) that have undergone a coagulation-flocculation and sedimentation treatment observed with the microscope (400X) (Water used: Regent's Park pond water (pH 8.4±0.1; turbidity: 0.8±0.3 NTU; absorbance at 254 nm, UV-254, 0.64±0.59), Coagulation-flocculation condition: 3.4 mg Al/L from $Al_2(SO_4)_3{\cdot}18H_2O$, PS MP 10 mg/L, initial pH 5. The coagulation time was 60 s with 300 rpm (G = 345 s^{-1}), flocculation time was 400 s with 50 rpm (G = 23 s^{-1}), and sedimentation time was 30 min).

3.1 Effect of flocculation stirring intensity on MPs' removal

Stirring speed has a crucial influence on flocculation. Faster the stirring speeds will cause greater breakage of the flocs and may lead to a reduction of the effect of the treatment. Previous studies selected stirring speed of 100 rpm (G = 66 s^{-1}) when using Al as coagulant (Zheng et al. 2011; Ma et al. 2019b). The range of stirring speeds investigated in this research were ≤250 rpm (G = 263 s^{-1}) (see reaction condition in Section 2.2) and while this favours the dispersion of the PS beads and the reproducibility of the system, it can affect the size of the flocs. Figure 2 shows the efficiency of the removal of MPs with the mixing conditions. The MPs removal initially increased to up to 95 % and then decreased rapidly from stirring intensity above 67 rpm (G = 36 s^{-1}). This may be explained by the fact that increasing mixing intensity, decreased the size of the flocs, making the removal less effective (Moruzzi et al. 2019). Therefore, in practice, for PS MPs removal, controlling the stirring speed at 50 rpm (G = 23 s^{-1}) in the flocculation process led to working conditions close to the optimum ones with reproducible stirring. Figure 2 includes a regression polynomium adjusted to the critical range of stirring speeds. This facilitates calculating the removal of MPs within that range. Figures 3-5 also include regression curves adjusted to the experimental conditions around the optimal removal of MPs.

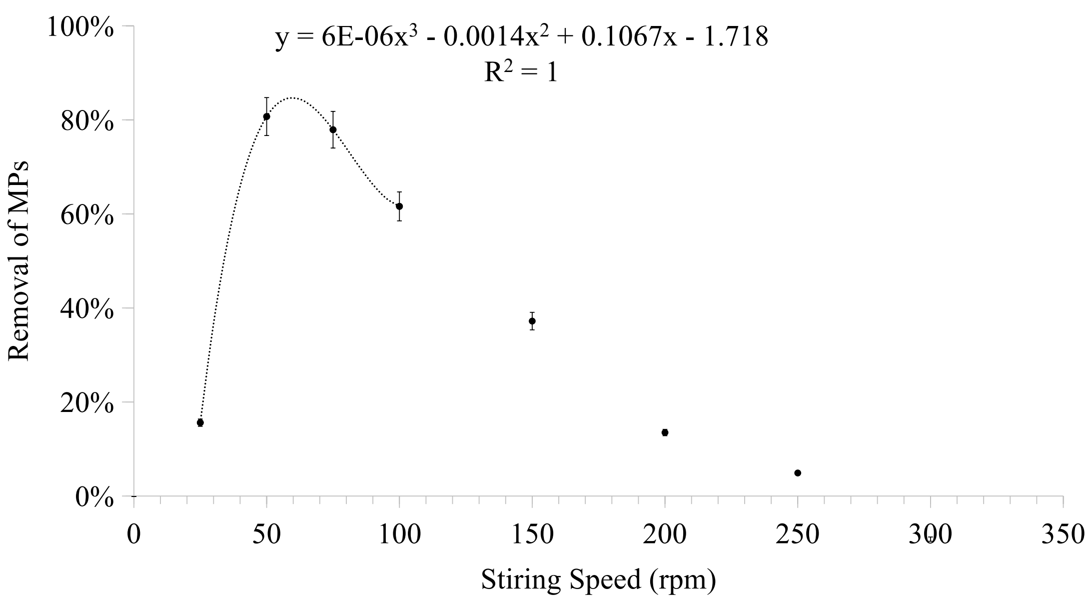

Fig. 2 Effect of flocculation stirring speed on the removal of 100 μm PS spiked in Regents Park pond water. The conditions used were: 3.4 mg Al/L from $Al_2(SO_4)_3 \cdot 18H_2O$, PS MPs 10mg/L, initial pH 5. The coagulation time was 60 s, flocculation time was 400 s, and sedimentation time was 30 min.

3.2 Effect of flocculation time on MPs' removal

The length of the flocculation time often determines the removal of suspended particles (Wu et al. 2012). Studies using Al salts as coagulant usually require about 15 min of flocculation time (Ahmad et al. 2006; Zhu et al. 2011; Wu et al. 2012). Shorter flocculation times than the optimum often lead to insufficient removal of particulates, while prolonged flocculation stages are unnecessary. From Figure 3, it can be observed that for stirring speed of 50 rpm ($G = 23\ s^{-1}$) when increasing the flocculation time to 400 s, or even longer, the removal of the flocs by sedimentation increased till 98.52 ± 1.04 % for the case of 100 μm PS beads. This behaviour can be explained by the flocculation kinetics as both stirrer speed and time dictates floc size and structure, and a dynamic equilibrium is expected (Oliveira et al. 2015; Moruzzi et al. 2013; Moruzzi et al. 2017), leading to the almost complete removal of MPs.

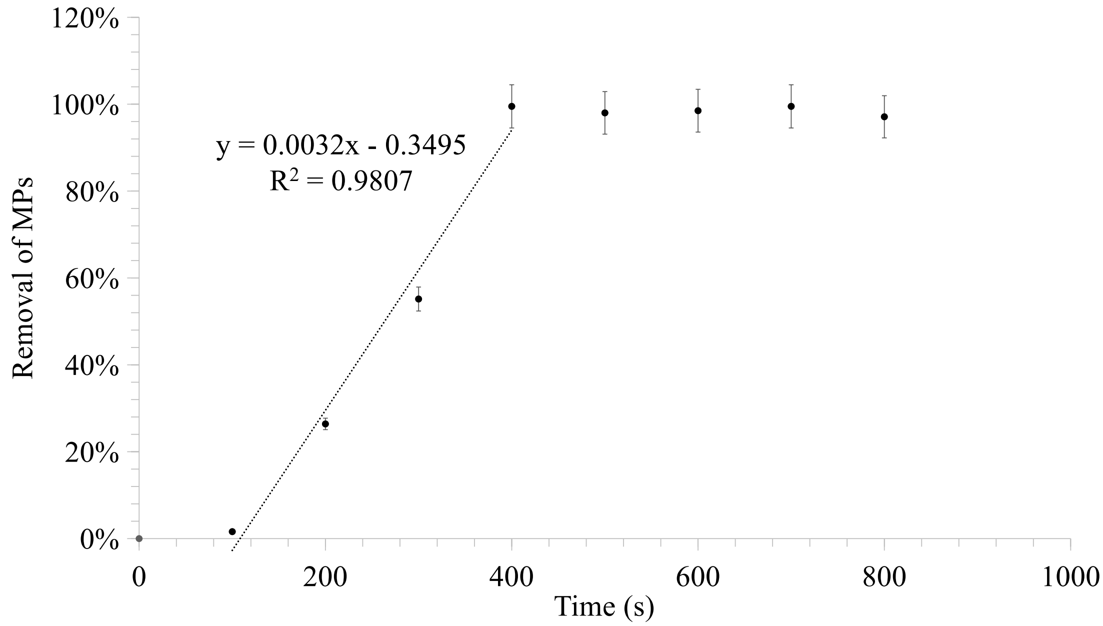

Fig. 3 Effect of flocculation time on 100 μm PS beads' removal from spiked Regents Park pond water. The conditions used were: 3.4 mg Al/L from $Al_2(SO_4)_3{\cdot}18H_2O$, PS 10 mg/L in water, initial pH 5, flocculation speed 50 rpm (G = 23 s^{-1}), coagulation time 60 s, sedimentation time 30 min.

3.3 Effect of initial water pH on the removal of 100 μm PS beads

Ionic strength has a crucial role in clarification (Yukselen et al. 2004) and the water pH generally has a great effect on the floc characteristics (Liu et al. 2013; Lee et al. 2012; Zhang et al. 2017; Zhao et al. 2014). Hence, to further investigate removal mechanisms of PS beads (as purchased and without the acrylic painting), the corresponding removal efficiencies were investigated at initial pH levels (before adding the coagulant) of 1, 3, 5, 7, 10, 12, 13, with the coagulation-flocculation conditions shown in Section 2.2. Among these pHs, the most relevant pH range of drinking and wastewater treatment (before adding the coagulant) is pH 5-7. After adding the coagulant, the pH of the suspensions was 3.27, 3.91, 4.88, 6.15, 8.41, 11.03, 11.75, respectively.

At acidic (pH 1 – 5), the MPs removal was ~ 54 % to 91 % (Figure 4) for flocculation speed 50 rpm ($G = 23\ s^{-1}$), coagulation time 60 s, flocculation time 400 s and sedimentation time 30 min. By adjusting the pH to > 6.8, the $Al_2(SO_4)_3$ flocculant hardly worked (the suspension remained turbid) and the MPs removal was low (~ 70 %). From Figure 4, adjusting pH to ~ 5 has favoured the removal of hydrophobic MPs because under these conditions aluminium sulphate has a large surface potential (Liu et al. 2013). Under these conditions, the removal of MPs achieved was 91 %. This may be explained by the fact the pH and the coagulant dosage determine which hydrolysis species is formed during coagulation. For example, in the case of aluminium coagulants, it is recognized that the optimal removal of particles from water is achieved under optimum pH conditions close to the point of minimum aluminium solubility i.e. 5.8 > pH > 6.5 where the sweep coagulation mechanisms occur (Gregory and Duan 2001).

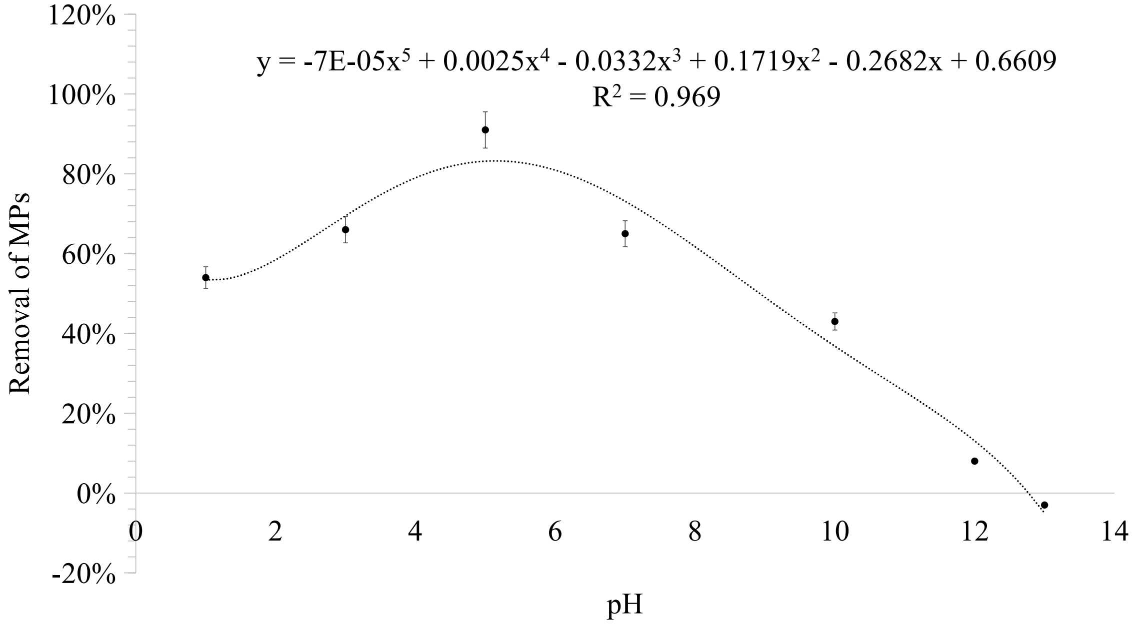

Fig. 4 Effect of coagulation pH on 100 μm PS beads' removal in spiked Regents Park pond water. The conditions used were: 3.4 mg/L as Al from $Al_2(SO_4)_3 \cdot 18H_2O$, PS MPs 10 mg/L, flocculation speed 50 rpm (G = 23 s^{-1}), coagulation time 60 s, flocculation time 400 s and sedimentation time 30 min.

3.4 Effect of sedimentation time on removal of PS MPs

After flocculation, sufficient sedimentation time will allow the suspended flocs to completely settle. This will minimise errors in the measurement of MPs because if there were smaller flocs floating in water, these could have been left in suspension and not sampled for MP counting with microscopy. Past studies trying to clarify kaolin (with density 2.6 g/cm^3 and particle size: 0.4 – 0.75 μm) in drinking water treatment found that $Al_2(SO_4)_3$ coagulation with sedimentation time of 30 min was effective to remove the flocs (Domopoulou et al. 2015), which is similar to the results found here for MPs with density lower than kaolin.

In the specific conditions of this study (removal of 100 μm PS beads (3.4 mg Al/L, PS MPs 10 mg/L, pH 5, stirring speed 50 rpm (G = 23 s^{-1}), coagulation time 60 s, flocculation time 400 s) sedimentation time was gradually increased until 40 min. The percentage of MPs removal reached 98 % at 30 min under these conditions (see Figure 5). After that, increasing sedimentation time did not lead to improvements in the removal of the study beads.

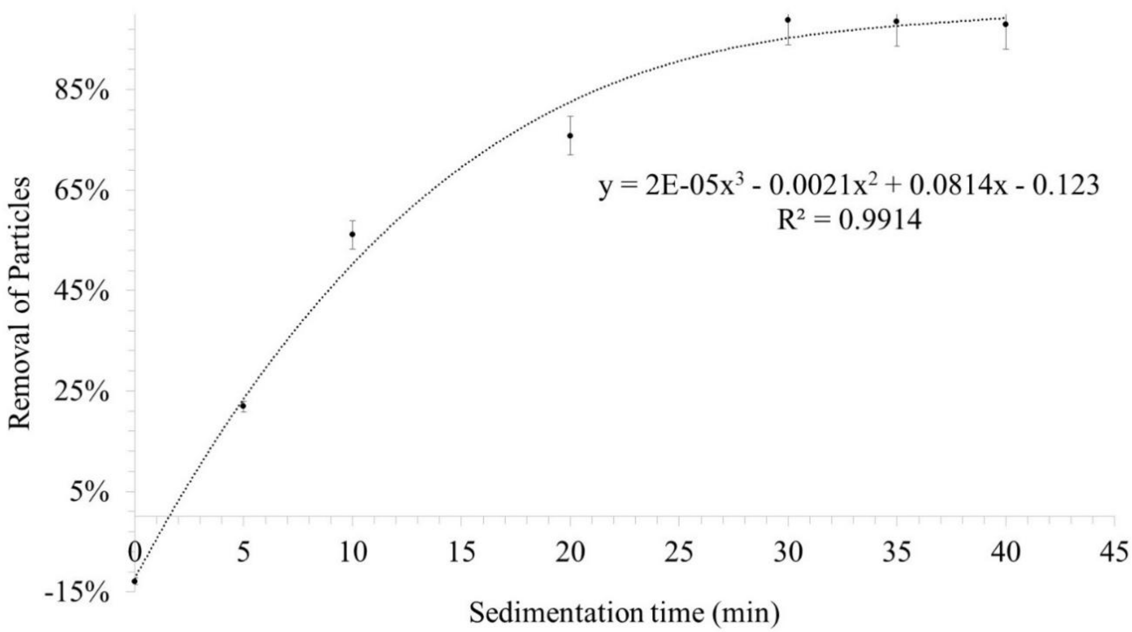

Fig. 5 Effect of sedimentation time on removal of 100 μm PS beads. The conditions used were: $Al_2(SO_4)_3 \cdot 18H_2O$ 3.4 mg/L as Al, PS MPs 10 mg/L, pH 5, flocculation speed 50 rpm (G = 23 s^{-1}), coagulation time 60 s, flocculation time 400 s.

3.5 Effect of floc-breakage and regrowth on MPs' removal

In this study, when flocs broke after increasing the stirring speed, additional dosage of coagulant (0.8 mg Al/L) led to the re-growth of flocs. These second flocs were larger than those before breakage (Figure 6). It is likely that, under the experiment conditions, additional MPs (which are hydrophobic and with non-formal negative charge) coated the surface of the broken flocs (positively charged) and as a result stronger and more interactions might have formed between the fragmented flocs. This interpretation agrees with a study that proposed that adsorption sites inside flocs can become exposed by the breakage and there is also a decrease of the zeta potential on the surface of the flocs (Yu et al. 2010).

The phenomenon of floc-breakage and regrowth with addition of coagulant improved the capacity for removing kaolin (Yu et al., 2010). In addition, floc removal after breakage/regrowth is dependent on the dosage of the additional coagulant. However, MPs beads have very different physical and chemical properties than kaolin clay in terms of density, surface area and surface chemistry. Therefore, the removal effect of reformed flocs and direct flocculation on PS MPs in the presence of kaolin needs to be investigated. To study floc breakage in detail, the average transmitted light intensity (Direct Current Value) and fluctuating root mean square (rms) components of the transmitted light intensity were monitored. This was done with the PDA instrument. The ratio (rms/DC), called as the Flocculation Index (FI) provides a measure of particle aggregation (Yu et al. 2010). The FI value is related to the size and concentration of the suspended particles and it significantly increases as aggregation occurs and decreases when aggregates break (Figure 6). From Figure 6, the FI value when adding coagulant increased even more than the original FI value after regrowth, therefore, this indicates that more particles were included in the flocs.

The removal of the PS 100 μm beads after floc breakage and regrowth reached 94 % at 1000 s, and this is about 16 % larger than traditional flocculation process (81 %) (Figure 6). Flocculation contact time throughout the floc breakage-regrowth process (i.e. 20 min) is therefore important in relation to the collisions between flocs including the PS beads but it also suggests that in case of floc breakage in a water treatment plant, flocs containing MPs may potentially be re-grown before greater removal of MPs by sedimentation. This potential advantageous step should be further investigated, particularly considering the different densities, types and sizes of MPs, water qualities and coagulant dosages as these may affect the results. These will be investigated in future experiments supported with zeta potential measurements.

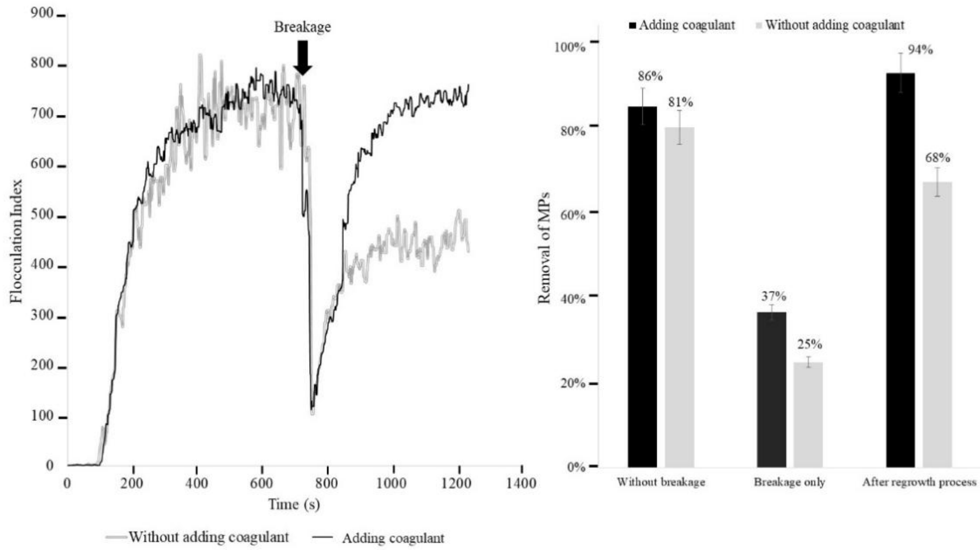

Fig. 6 Effect of floc-breakage on FI with and without additional coagulant

The current conditions for alum flocculation in drinking water treatment plants are 40 rpm flocculation for 10 min, sedimentation for 20 min, and pH ~ 6 before coagulation (Ma et al. 2019a; Combatt et al. 2020; Cardoso Valverde et al. 2018). According to the results of this study, if the stirring rate is increased to the equivalent gradient of velocity (G = 23 s^{-1}), the settling velocity is modified to the equivalent time of 30 min at Jartest, and the pH before coagulation is adjusted to ~ 5, the effect of flocculation on low-density PS microplastics will be their increased removal to 99 %. Adjusting the flocculation process however will impact other suspended solids and pollutants and needs further study.

A limitation of the present study is that it used commercially available pristine PS beads and research is starting to show that irregularly shape beads may have markedly different toxicity and may interact with flocs slightly differently than commercial bead. Hence it is recommended to harvest MPs in the environment or water treatment when possible (Yokota and Marissa 2020). However, we opted for using commercially available MPs in order to have sufficient availability of similar type of beads for the experiments planned in this work.

4. Conclusions

It is urgent to understand how to remove MPs in drinking and wastewater treatment given that these are an opportunity to reduce MPs' spread and protect the environment and humans. This study investigates the removal of low-density MPs during the flocculation process, which plays an important role in decreasing the turbidity of water and hence may be the key to remove MPs particles. This is a preliminary study that has screened the effect of the duration and stirring speeds in coagulation-flocculation and sedimentation when using a common coagulant for 100 μm low-density PS beads as a model. These MPs have been selected due to their toxicity and composition and size commonly found in effluents from clarifiers. The study on a single type of MPs has allowed to achieve greater detail in the removal conditions. The optimized coagulation-flocculation conditions found were 3.4 mg Al/L, pH 5, flocculation time 7 min, precipitation time 30 min. Under these conditions, and when natural water was used, percentage removals were 98.9 ± 0.94 %.

The breakage and regrowth process of flocs have shown to enhance the removal of 100 μm low-density PS beads by flocculation, when additional dosage is applied. Although this study used PS (1.04-1.06 g/cm^3) as model, these findings can potentially be applicable for other hydrophobic MPs and MPs of similar density (e.g. PP (0.9 g/cm^3); PS (1.06 g/cm^3), Polyethylene (PE, 0.92 g/cm^3) and nylon (1.14 g/cm^3)). Further research on different sizes of the MPs is needed as well.

Given that, the re-flocculation process has not been maturely applied in the water treatment industry as a MPs target technology. This paper points to considerations for the improvement of drinking water flocculation treatment process in the future. Future work should address how coagulation-flocculation-sedimentation conditions change over wider variety of MPs; and how these optimal conditions for MPs will be affected in the presence of organic pollutants and other suspended particles. It is necessary to investigate wider types of raw water and give further insights of removal mechanisms by monitoring the change of zeta potential of flocs under different conditions. Finally, this work confirms that coagulation-flocculation and sedimentation are important steps for the removal of MPs.

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Declaration of Competing Interest

The authors report no conflict of interest.

CRediT authorship contribution statement

Chaoran Li: Methodology, Investigation, Visualization, Writing - Original Draft, Resources; **Rosa Busquets:** Conceptualization, Supervision, Writing - Review & Editing; **Rodrigo B. Moruzzi:** Writing - Review & Editing; **Luiza C. Campos:** Conceptualization, Supervision, Writing - Review & Editing

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Supporting Information

Preliminary Study on Low-Density Polystyrene Microplastics Bead Removal from Drinking Water by Coagulation-Flocculation and Sedimentation

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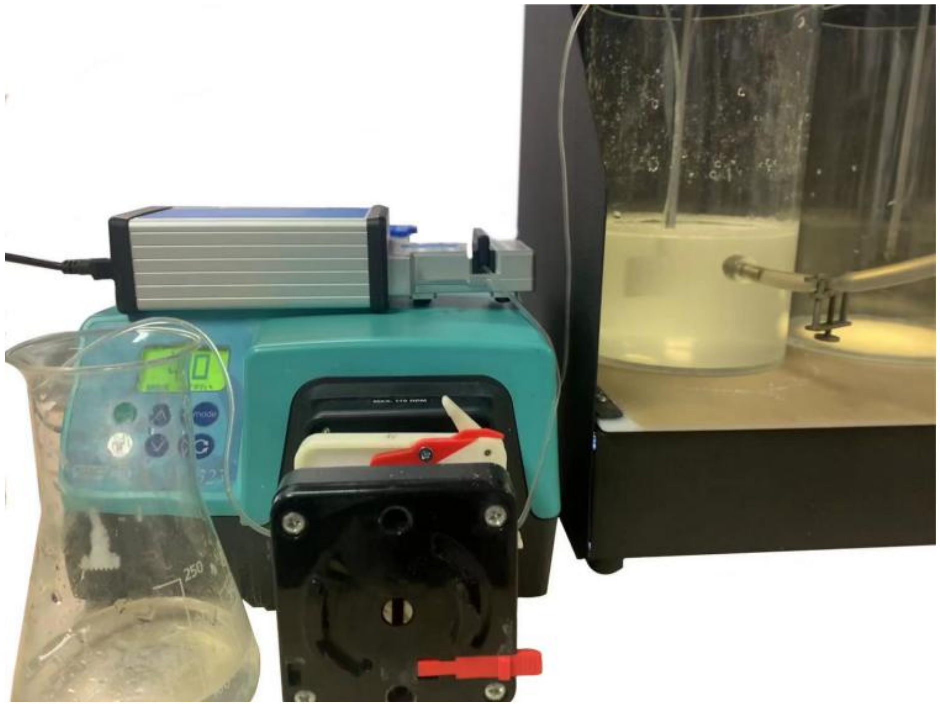

Fig. S1 - PDA device and jar tester flocculator assembly used in this study.

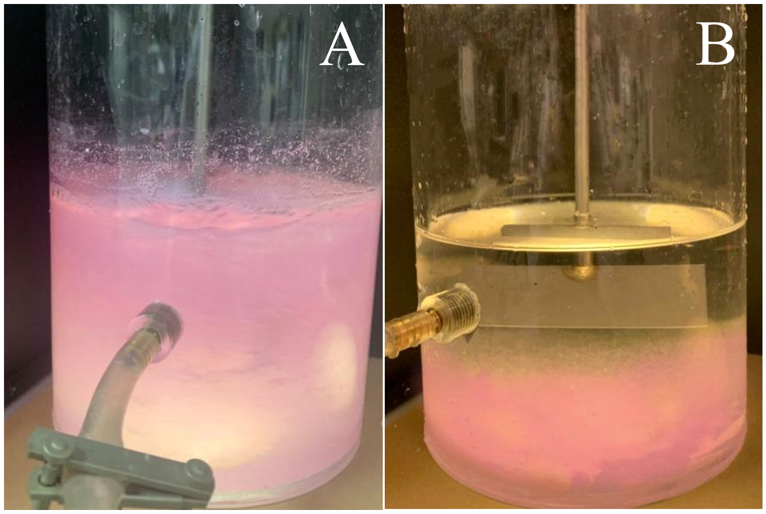

Fig. S2 – Water solution during the coagulation-flocculation process (A) and after sedimentation (B). The water used was from the pond in Regent's Park (pH 8.4 ± 0.1; turbidity: 0.8 ± 0.3 NTU; absorbance at 254 nm, UV-254, 0.64 ± 0.59), Coagulation-Flocculation condition: 3.4 mg Al/L, PS MP 10 mg/L, initial pH 5. The coagulation time was 60 s with 400 rpm, flocculation time was 400 s with 50 rpm, and sedimentation time was 30 min.

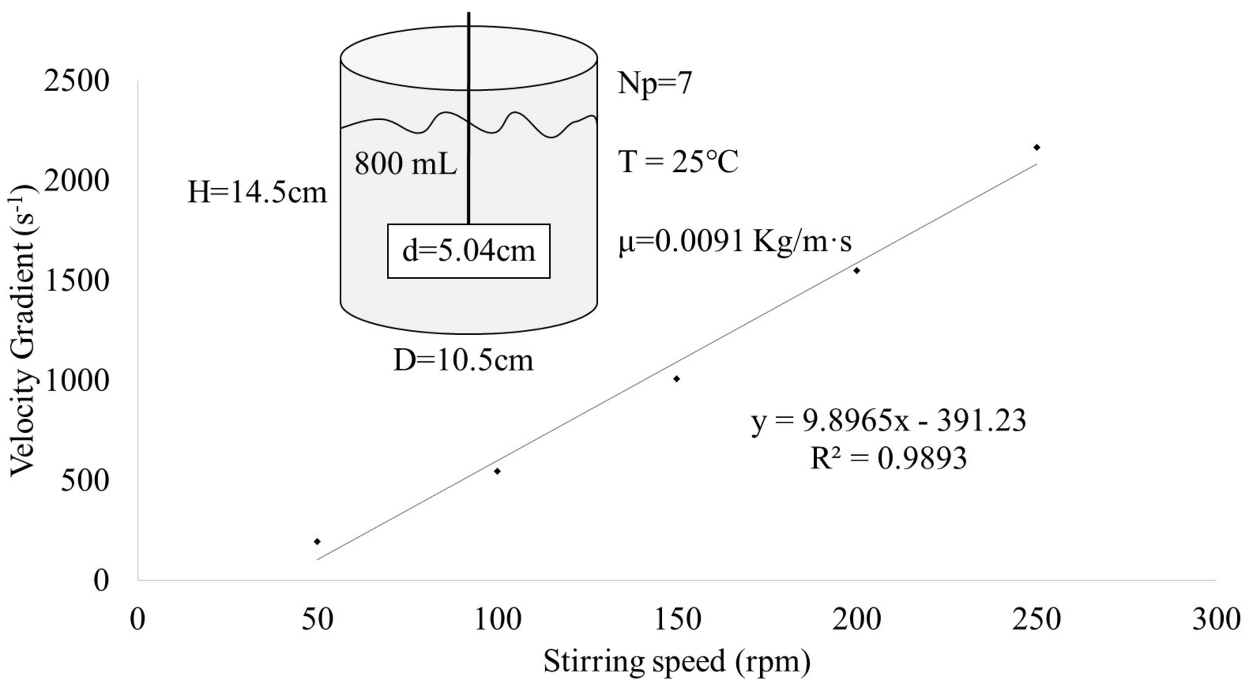

Fig. S3 - Conversion between stirring speed (in rpm) to velocity gradient (in s^{-1}) with the configuration and conditions used. Note: H = jar depth; D = jar diameter; d = blade diameter; Np = power number; T = water temperature; μ = water viscosity.