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# Micoplastic removal from natural aquatic environments through dissolved air flotation

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**Abstract.** In the current study dissolved air flotation has been applied to verify the removal characteristics in drinking water treatment processes. Various aspects of microplastic, and water quality conditions were tested using dissolved air flotation to determine the removal efficiency and mechanism of microplastics. To simulate DAF processes under practical conditions, the type and size of microplastics were set based on the water quality conditions of a river, and the experiments were conducted under similar water quality conditions. For the microplastics used in the experiments various size, and densities were tested to verify its removal characteristics. Different pollutant type and concentrations were also tested with microplastic to verify the removal rate and mechanism during the flotation process. To better understand the removal of microplastics, different type and concentration of coagulants were also used in the system to better understand the removal mechanism. Finally the transport of various heavy metals were also verified to better understand the effects of organic pollutant's effect on heavy metal transport in the system. Results showed a correlation with the microplastic flotation capacity and removal rates. Effects of organic matter showed a high impact on the removal mechanism, where the increased amount of organic matter resulted in lower removal rates. The amount of organic matter also influenced the ate and transport of heavy metal sorption and transport on microplastic surfaces, indicating a higher hindrance of sorption capacity due to chelating effects.
**Keywords:** dissolved air flotation; fate and transport; heavy metal sorption; micropastic; microplastic coagulation

## 1. Introduction

Plastic products have been widely utilized in our everyday lives including house hold, and commercial products. However, with the utilization of plastic products, primary and secondary micro plastics have become a rising issue/ For household products, primary microplastics are those that leach directly from within the product, such as functional cosmetics (Napper *et al*. 2015) and toothpaste (Ustabasi and Baysal 2019), while secondary microplastics are those that leach from items such as tea bags (Hernandez *et al*. 2019) and food containers (Du *et al*. 2020). The microplastics produced, are known to travel into the environment through various channels, and eventually travel into various drinking and wastewater treatment plants (Carr *et al*. 2016, Liu *et al*. 2021, Pivokonsky *et al*. 2018).

Research conducted in various regions in the world show a wide variety of different microplastic removal characteristics in drinking water treatment plants. Micropastic removal through conventional drinking water treatment plants composed of coagulation/flocculation and sand filtration systems were investigated in Czech and India, showing an overall removal of 40~85.3% micro plastics (Pivokonsky *et al*. 2018, 2020, Sarkar *et al*. 2021). Additional investigations with advanced treatment processes such as ozonation and activated carbon systems showed higher removal in general with 82~99.9% removal efficiencies. (Wang *et al*. 2020, Zhang *et al*. 2020) Additional to the treatment processes, variables such as coagulants have also been reported to influence the removal of microplastics in the treatment systems, where the type and amount of coagulants also showed different removal efficiencies (Na *et al*. 2021, Zhou *et al*. 2021).

With various types of feed waters, a variety of microplastics will also flow into the drinking water treatment plants. The effect of the material characteristics, size, and shape on the effects of microplastic removal in drinking water treatment plants have been investigated. Results showed that plastics with higher densities, such as polyethylene terephthalate (PET) and, polyethersulfone (PES) with densities of 1.38 and 1.46 g/cm3 respectively, are discovered in higher abundancies, compared to the less denser polyethylene (PE), and polystyrene (PS), which both have densities lower than 1.0 g/cm3, in the treatment effluent (Dalmau-Soler *et al*. 2021, Barbier *et al*. 2022, Dronjak *et al*. 2022). The size of the microplastics also differed during the treatment process, where the size composition if the microplastics in the effluent were highly dependent on the size composition of the feed water (Kankanige and Babel 2021, Chu *et al*. 2022, Xu *et al*.

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2024). However, studies have also shown that during the treatment process, microplastic smaller than 50 μm were mostly removed due to the conventional systems, leaving microplastic's larger than 50 μm as the majority size in the effluent (Jung *et al*. 2022, Dronjak *et al*. 2022).

Through various studies, DAF has been proven to be an effective method in removing microplastics in drinking water treatment processes. Due to the low density of microplastics, various flotation methods have been applied for the removal of the materials. Methods such as hydro cyclone treatment (Yuan *et al*. 2024), batch air flotation (Seart *et al*. 2022), froth flotation (Jiang *et al*. 2023), and dissolved air flotation (Wang *et al*. 2021) have been researched for the removal of microplastics in various feed waters. Microplastic removal has been tested for various wastewater and grey water systems, where high concentrations of organic mad microbial matter exists (Esfandiari *et al*. 2021, Chai *et al*. 2023). Due to the difficulty of individual microplastic removal in water, various attempts have also been conducted to add modifying agents in the systems (Feilin and Mingwei 2022, Xu *et al*. 2024, Jiang *et al*. 2021). However although various research has been conducted on microplastic removal through dissolved air flotation, there is a lack in the application of dissolved air flotation for drinking water treatment processes.

In the current research, various conditions were tested to understand the fate and transport of microplastics in drinking water treatment plants. Microplastic of different particle size, and density were tested to understand the characteristic effects on removal efficiency. Different coagulants as well as pollutants were also tested under various combinations to understand the effects of flocculation characteristics on the removal mechanism of microplastics in the treatment process. Additionally, heavy metal and organic matter were tested for sorption characteristics and its transport potential. By understanding the removal mechanism of microplastics in dissolved air flotation processes, the research seeks to improve microastic removal efficiency and contribute to the development of future field applications.

## 2. Materials and methods

### 2.1 Microplastic sample preparation

To acquire the microplastics, commercial products composed of polystyrene (PS), nylon, polyvinylchloride (PVC)and polyurethane(PU). The polymers were selected based on the different densities (PS;1.02~1.08, Nylon; 1.13-1.15, PU;1.2, PVC;1.3-1.6) to verify its removal characteristics through dissolved air flotation. The base material for each of the micropalstics were fabricated from petri dishes (PS), sponges (PU), Cable ties (Nylon), and pipes (PVC). The material was cut with steel scissors or crushed with a hammer to initially break the material to small fragments. The fragments were then inserted into a commercial blender (Tefal optimo glass BL3131KR), where the plastic fragments were added with cold water and mixed under medium and high intensities. The fragmented plastic after blending, was then sieved for size separation. The sieves were composed of sizes including 1 mm, 500 μm, 250 μm, and 150 μm. After sieving, the size-sorted samples were dried at room temperature and stored in a temperature controlled desiccator.

### 2.2 Dissolved air flotation

A lab scale multi-unit dissolved air flotation unit was manufactured for the research. The unit was composed of an air compressor (Gunpoong power tool, EWS24, Korea) with a capacity of 0.116 m3/min, a 2 L saturation vessel composed of stainless steel. The saturator was connected to a water and pressure distributer to conduct various experiments simultaneously. The flotation cell was composed of glass to prevent residual plastic to contaminate the samples. A cylindrical rim was installed on the flotation cells for collection of the separated microplastics. The collection of micoplastics after the DAF experiment was conducted by continuously pumping water into the flotation cell with a peristaltic pump (LEADFLUID, BT100L, Korea) until the separated scum overflows into the cylindrical rims. After the floated scum is removed, the remaining microplstics in the flotation cells are then collected and analyzed for separation calculation. The DAF units were operated at pressure 4 bar, with an inlet water volume 300 ml. The coagulation process was conducted under 180 rpm high speed stirring for 5 minutes, followed by a flocculation process of 50 rpm stirring for 15 minutes, and a settling time of 10 minutes prior to bubble introduction.

### 2.3 Fluorescent Microscopy Analysis

Images were captured and analyzed using a Confocal Microscope (Leica EZ4 HD, Leica systems, Korea), with a magnification range up to X30 ~ X800,000. Flocculation samples were collected after jar tests and stationed in a petri dish. The sampler were magnified where the size was analyzed with the software (eXcope, Leica systems) for the size and shape of the microplastics embedded in the flocs.

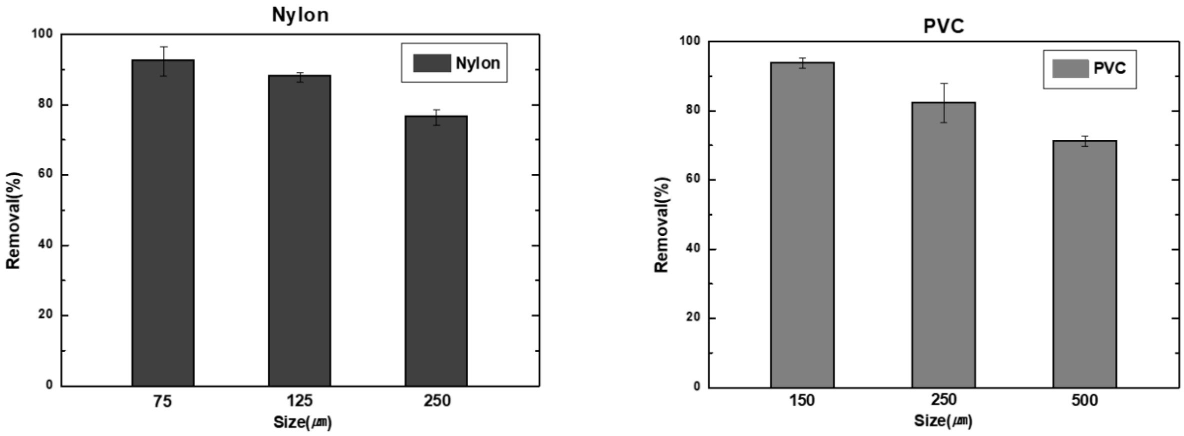
### 2.4 Heavy metal sorption experiments

For the current experiment, polystyrene of 125 μm in size, was used. Humic acid (Sigma Aldrich, USA) was used for organic matter, and copper (Cu), nickel (Ni), and zinc (Zn) (Sigma Aldrich, USA), which are commonly found in water supplies, were used for heavy metals. For the adsorption experiments, 1000 ppm of polystyrene microplastics were added to 10 ppm of humic acid and 50 ppb of heavy metals and stirred at 70 rpm on a shaking plate for 1, 3, 5, 7, and 9 days, respectively. After the adsorption experiment, the residual amount of organic matter was measured by TOC analyzer for organic matter and ICP-MS for heavy metals.

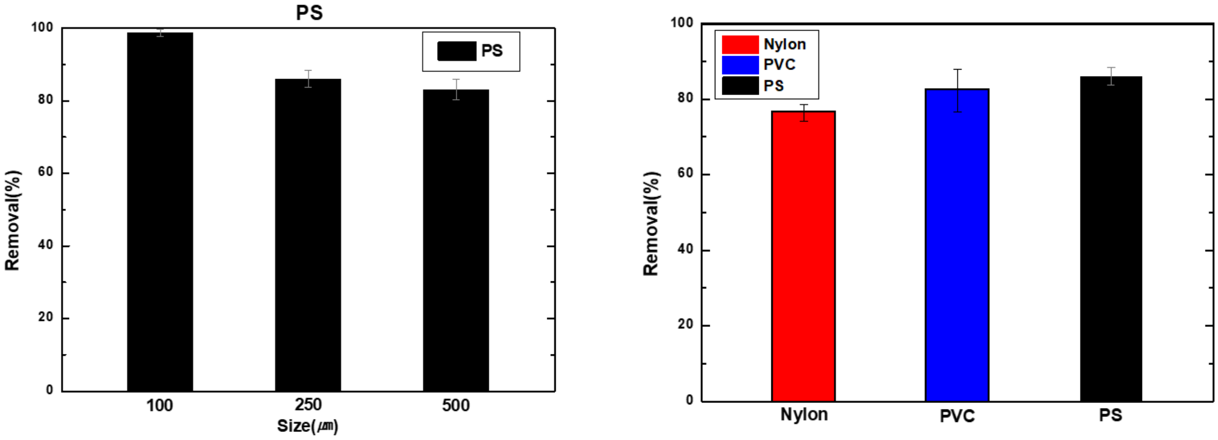
## 3. Results

### 3.1 Effect of MP size and density

Figs. 1(a)-1(d) shows the removal of microplastics based



(a) Nylon (b) PVC



(c) PS (d) comparison of the removal of different MPs

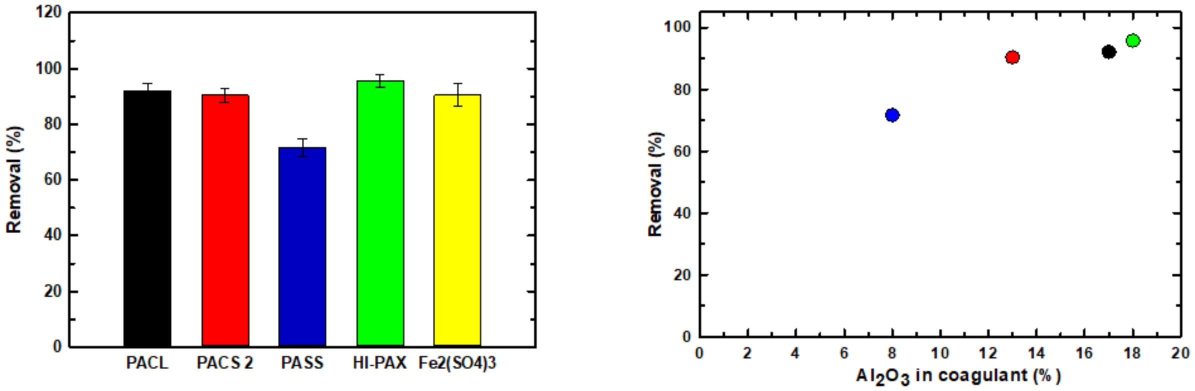
Fig. 1 ANN model output training data for upstream typhoon wind field coming from N direction with exponent 0.22

on the size and density of the particles. In general, the degree of removal decreased based on the density of the plastics where the highest density PVC particles(1.3~1.6) showed the lowest removal, and the lowest density PS particles(1.05~1.08) showed the highest removal. With the increase of particle size, results also showed a higher decrease in the removal rates with higher density materials. The highest density microplastics which were nylon, showed a removal decrease of 96% to 76%, where PVC shows a removal of 35% to 70%, and PS with the lowest density showed a removals of 98% to 80% with increasing size. The results correlated well with the stokes law, where the higher density and particle sizes resulted in higher sedimentation speeds and hence, lower removal rates. However, regardless of the size and density, the decrease in removal show larger differences compared to conventional particular matter (Kwak *et al.* 2018, Kwon *et al.* 2023), which may be due to the lack of coagulation among the microplastic particles. With conventional particular matter, such as kaolinite, floc formation increases the interaction with air bubbles, where plastic do not form flocs, resulting in a removal rate influenced strongly with its size and density of the microplastics. The size may also influance the attachment due to the higher diffusion of the smaller particles, where the smaller particle show higher diffusion within the treatment process resulting in higher degrees of attachment to bubbles and flocs.

### *3.2 Effect of coagulants on MP removal*

To further analyze the relationship between microplastic removal and coagulation, various coagulants were tested for the removal of microplastics. Five different alum and ferric coagulants currently used in drinking water treatment plants were tested for its coagulation and air flotation removal properties. The removal rates under the different coagulants were compared in Fig. 2. Results showed maximum removal using Hi-PAX with a 95.73±2.24% removal, and minimum using PASS resulting in microplastic removal of 71.7± 3.41%. Aside from the coagulant PASS, the majority of the removal of microplastes in the range of 90% to 95%. The removal rates were related to the coagulation capacity of each of the coagulants where the larger the flocs, higher removal rates were observed. To verify the relationship between the coagulation and microparticle removal, the amount of alum dosage in each coagulant was correlated with the removal rate. Results showed a positive correlation with the percent of alum content in the coagulant chemicals and the removal rate. Considering the fact that the composition of the coagulating agents are mainly alum and the additional substances in the coagulants are for additives for pH control and coagulation aids including silica dioxide, sodium silicate, and calcium ions, the main removal mechanism was due to the alum.

To verify the removal of microplastics during the



(a) Removal by coagulants (b) removal by amount of alum in coagulants

Fig. 2 Removal of MP under various coagulants

**(a) PACL** (b) PACS 2 (c) PASS (d) HI-PAX

Fig. 3 Floc formation under various coagulants

coagulation process, images of the flocs after the removal process are shown in Fig. 3. The coagulation experiments were conducted with polyurethane microplastics, and imaged with fluorence microscopy. Results show that with the coagulants PACL and PACS 2 which show larger sizes of flocs compared to the coagulant PASS.

Images show microplastics surrounded by the flocs indicating the attachment of the plastics in the flocculation process. Results from Fig. 3 show microplastics attached or entraped in different size of flocs, where with larger size flocs a higher number of microplastics entrapped. This is due to the surface are of the flocs having a higher attachment with larger sizes, making it easier to interact with microplastics.

### *3.3 Effect of particle and organic matter on micro-plastic removal*

#### *3.3.1 Microplastic removal with particular and organic pollutants*

The effect of pollutant composition on the removal of microplastics were tested under various feed water conditions. Experiments were initially conducted with particular matter, where humic acids of various concentrations were added with the particular matter. The different composition of pollutants were tested to understand the effects of different water pollutant compositions and its effect on microplastic removal through dissolved air flotation systems.

Under conditions with only particular matter, the results show 90% of microplastic removal under the concentraitons tested. With the increase of humic acid concentrations, the results showed lower microplastic removal. This can be seenwith humic acids of 2ppm and 4ppm conditions where the increase of humic acids showed a decrease in the removal rate. With 2ppm of humic acid the removal rate of microplastics decreased from 90% to 65%, and with humic acids of 4ppm the removal rates decreased from 87% to 24%. The decrease of the removal rate with the increase of organic matter indicates the role of organic matter in the coagulation process. The increase of organic matter results in the reduction of the effectiveness of coagulants reducing the size of the flocs, and hence the lower rate of interaction between microplastics and the flocs. Smaller sizes of flocs result in the decreased rate of microplastic entrapment in the flocs, where the flocs aid in increasing the surface area for the microbubbles to interact with. However with only particular matter as pollutants, the flocculation will result in larger flocs due to the lack of organic matter leading to higher entrapment rates. Additionally the hydrophobic surface of kaolinite may also aid in the attachment of particular matter to the microplastics. Results indicate that under conditions with high organic pollution, the effectiveness of microplastic removal decreases.

#### *3.3.2 Cogalulant dosage for microplastic removal*

To further verify the effect of organic matter on the removal of microplastics, the optimal coagulant doses for the coagulation process for dissolved organic matter was tested. various coagulant dosages were tested under different pollutant concentrations, where the maximum removal of microplastics were summarized in Fig. 5. Results showed that the requirement of coagulants increased with the amount of particulate and organic matter. And with the addition of organic matter the required dosage increased with a higher rate. When the particular matter was increased from 25mg/L to 100 mg/L, the amount of coagulants needed was increased by approximately 263 %,

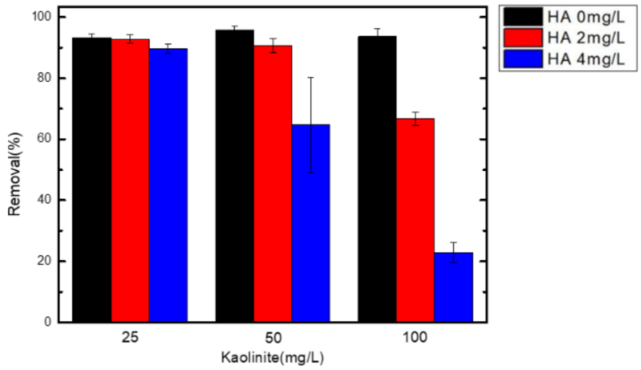


Fig. 4 Removal of microplastics under various particle and humic acid concentrations

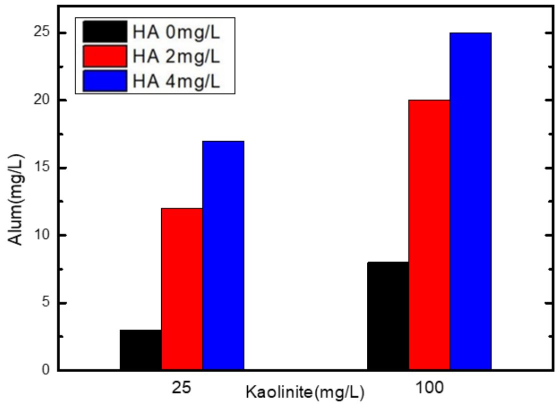
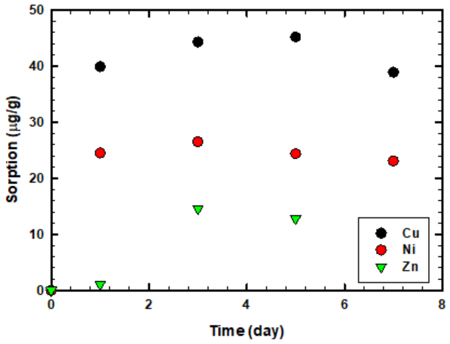
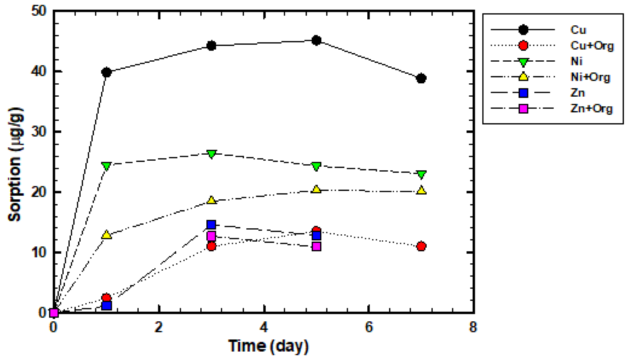


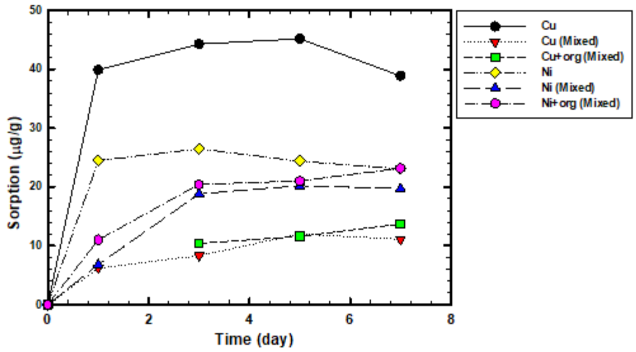
Fig. 5 Optimal coagulant dosage for various pollutant contents



(a) sorption of metallic ions



(b) sorption of metallic ions and organic matter



(c) sorption of organic matter with various heavy metal combinations

Fig. 6 Sorption of metallic species on microplastics

while with the increase with organic matter, the optimum dosage of coagulants increased by 250~400 % with humic acids of 2ppm and 310~600% with humic acids of 4ppm. The findings with the optimal coagulant dosage indicates that organic matter highly interacts with the coagulants that hinder the efficiency of the floc formation and hence the removal of microplastics through dissolved air flotation.

### *3.3.4 Organic attachment on microplastic surfaces*

Previous reports have shown that microplastics in drinking and waste water treatment plants may be contaminated with heavy metals (Islam *et al*. 2023, Ghergel *et al*. 2019, Hatinoglu *et al*. 2021). To better understand the phenomena interaction between microplastics, organic matter and heavy metals. Three different experiments were conducted to verify the potential of heavy metal transport through the treatment process. Sorption kinetics were tested with, and without organic to verify the fate and transport of heavy metals in water treatment systems. As shown in the Fig. 6(a), aside from zinc (Zn), heavy metal sorption occurred within 24 hours suggesting that if heavy metals are present, they are likely to be adsorbed on the microplastics. Copper showed the highest amount of sorption, followed by Nickel and Zinc.

Fig. 6(b) is an experiment on the effect of the presence of organic matter on the adsorption of heavy metals. Experiments were conducted where organic matter was added after the heavy metal sorption occurred to verify the effects of organic matter. Results showed that for all heavy metals tested, the amount of adsorption decreased in the presence of organic matter. This is expected to be caused by the binding of metal ions to the organic matter through the chelate reaction. Suspended organic matters interacted with the heavy metals detaching it from the microplastic surface. As shown in the results, copper, which is the most strongly chelated with organic matter, showed the greatest reduction in adsorption, while less chelated heavy metals such as Zn showed less reduction in adsorption. From the results, it can be expected that under field conditions, when higher concentrations of organic matter is introduced to the feed water, detachment of heavy metals may occur from the microplastics, increasing the amount of dissolved matter. Additionally, the amount of detachment of heavy metals rely on the chelating strength of the organic matter and heavy metals

In the case of Fig. 6(c), various heavy metals were adsorbed with organic matter simultaneously. In addition, microplastics were added to a copper and nickel mixture to understand the competitive sorption of the heavy metals. In the case of copper, the amount of sorption decreased when various heavy metals were present. Copper sorption decreased significantly, which is due to the more sensitive electro= magnetic reactions, where copper has a larger induced dipole than nickel. In contrast, with of nickel, adsorption occurs regardless of the charge. It was also confirmed that the amount of adsorption onto microplastics decreased when organic matter was present along with various heavy metals. The decreased amount of heavy metal sorption due to organic matter chelation was lower than metal detachment due to organic matter shown in Fig. 6(b). Compared to the organic matter detachment in Fig. 6(b) the attachment of the organic and heavy metals showed lower results. Results indicate that with various pollutants, the sorption of heavy metals will decrease indicating a higher amount of suspended metals in ionic or chelated forms.

## 4. Conclusions

In the current study, the effects of size, density, and pollutant configuration on microplastic removal through dissolved air flotation was verified. Various plastic composed of different sizes and densities was tested for the effect of buoyancy on the removal mechanism. Additionally the configuration and concentration of various pollutants were treated with microplastics to verify the effects of floc formation on the removal mechanism. Results showed the effects of density and size of microplastics on the removal through dissolved air flotation. With plastics of higher density, removal decreased with the increase of the particle sizes. Plastics with densities higher than water showed a higher decrease in the removal with the increase of size. Decrease in the removal rate was higher than other conventional particular matter due to the low coagulation of the microplastics. Microscopy of the floc formation and coagulant analysis indicated the removal mechanism of microplastics through the entrapment of the plastics during the floc formation. Microplastics were shown to be entrapped during the formation of the flocs or would be attached to the surface of the flocs, which in both cases rely on the formation and interaction of the plastics and flocs. To verify the effects of pollutants on the removal of microplastics, different pollutant configuration and concentrations were tested. Results showed that organic matter highly impacted the removal of plastics due to the interaction with coagulants. Organic matter showed a high affinity with coagulants, indicating a higher concentration of coagulants needed for microplastic removal. The amount of particular matter showed lower impacts on the removal, however with the optimal dosage of coagulants, the removal rates increased with particular matter concentrations. Additionally the attachment of heavy metals due to organic matter was also verified to understand secondary pollution in the water treatment system. The presence of organic matter with heavy metals showed a decreased attachment of the heavy metals to the microplastic surface indicating the increase in suspended forms. The current study various different effect and characteristics of microplastic removal through dissolved air floatation processes. With the findings in the research with hopefully aid to optimize plant operations and expand the understanding of microplastic in dissolved air floatation systems.

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