

Development of microplastic remediation techniques from marine sediments

Michiel Van Melkebeke^{1,2}, Colin Janssen² and Steven De Meester¹

¹Department of Green Chemistry and Technology, Graaf Karel de Goedelaan 5, 8500 Kortrijk, Belgium

²Laboratory of Environmental Toxicology and Aquatic Ecology, B-9000 Ghent, Belgium

Presenting author email: Michiel.VanMelkebeke@UGent.be

Keywords: microplastics, microplastic remediation, sediment remediation, centrifuge, flotation

Introduction

Microplastic pollution of the aquatic environment has raised worldwide concerns over the past two decades. This is reflected in the number of scientific publications on the sources, occurrence and particularly the impact of microplastics [1]–[3]. Most commonly defined as plastic particles between 1 μm and 5 mm, microplastics pose a threat to the marine environment and potentially the public health. Various adverse effects such as the blockage of feeding appendages, oxidative stress and a reduction of growth rate have been found in many different marine species. Next to that, drinking water and ordinary human food products such as table salt and tea bags have been shown to contain significant amounts of microplastics [4]–[7]. While the direct and indirect risks associated with human consumption of microplastics are only marginally understood, it is clear that the continuous accumulation of microplastics in the marine environment is detrimental on a global scale. Despite this, there is hardly any work reported on the technological remediation of these persistent marine pollutants. As most microplastics accumulate in marine sediments [8]–[10], the development of cleaning/separation techniques that are able to isolate microplastics from these sediments seems imperative. To that end, a fundamental analysis of the characteristics, sinking behaviour and surface properties of typical microplastics is essential, as many separation techniques are based on these properties. In this work, proven separation technologies are evaluated against their predicted ability to isolate microplastics from marine sediments. Ultimately, an optimal design is proposed as the first microplastics remediation technique of marine sediments. Further improvements to this novel installation are currently ongoing on a laboratory scale.

Materials and methods

The first part of this work provides new fundamental insight into the characteristics and sinking behaviour of typical microplastics. For these analyses, microplastics were used originating from municipal plastic waste comprising six different polymer types. Each microplastic particle was characterized by mass, density and shape. As particle shape is a complex parameter to quantify, various dimensionless shape descriptors were used for this purpose. Important to note is that both spheres, fibers and films were included in the experimental design. Subsequent sinking rate measurements were then evaluated against 11 shape-dependent drag models. Next to that, the effect of biofouling is investigated. Biofouling is expected to change the density of the microplastic particle as well as its polarity. To quantify the polarity of the microplastics' surfaces, contact angle measurements were performed. This because a water contact angle smaller than 90° indicates a hydrophilic surface, while a water contact angle greater than 90° indicates a hydrophobic surface.

To evaluate proven separation technologies, (some of) the most important separation factors for the microplastics/sediment mixture are determined, namely density, shape and polarity. Note that particle size is not considered to be an important separation factor as their respective size ranges significantly overlap. From the theoretical evaluation, centrifugal sedimentation and (froth) flotation are considered to be the most promising for dealing with this solid-liquid mixture. Centrifugal sedimentation was further examined by simulating the grade efficiency curves corresponding to a decanter centrifuge with conventional dimensions. To account for the distinctive shapes of typical microplastics, the shape-dependent drag model that best fitted our data in the previous part was incorporated in these calculations. With respect to (froth) flotation, a series of explorative experiments were performed including dissolved air flotation (DAF), mechanical flotation and a pneumatic flotation column.

Results

From the first part of this work, it was found that particle shape is a particularly important parameter influencing the sinking behaviour of typical microplastics. For instance, terminal sinking velocities were shown to deviate up to 7 times from the reference law for spheres when evaluating film shaped particles. Therefore, identifying appropriate shape descriptor(s) to quantify the distinctive shapes of microplastics is concluded to be essential in order to make sensible predictions about their flow behaviour. Here, it is found

that the drag model by Dioguardi et al. [11] most accurately predicts the sinking behaviour of typical microplastic particles with an average error of 13,20 %. The corresponding shape-dependent drag model uses sphericity and circularity to quantify particle shape. With respect to biofouling, it is derived that floating film shaped particles are more likely to sink as a direct result of density modification caused by biofouling compared to floating spherical particles. Furthermore, the polarity of the plastic surfaces appears to alter from (near) hydrophobic to strong hydrophilic as a result of biofouling. The latter reduces the potential of polarity as a separation factor in microplastic/sediment mixtures as sediment particles are known to be predominantly hydrophilic [12], [13].

From the grade efficiency curves of the decanter centrifuge (Figure 1), it may be concluded that separation solely by centrifugal sedimentation is not sufficient for the remediation of sediment mixtures polluted with microplastics. The main reason for this appears to be the significant overlap in size of the particles to be separated when considering the strong correlation between the particle diameter and the grade efficiency. However, low-density microplastics that would intrinsically float in seawater (prior to biofouling) are predicted to be completely separated from the sediment particles. This is partially due to the expectation that the turbulent regime inside the decanter would readily detach the biofilm layers from the microplastics' surfaces. Consequently, particle density is shown to be a useful separation factor, unlike particle size.

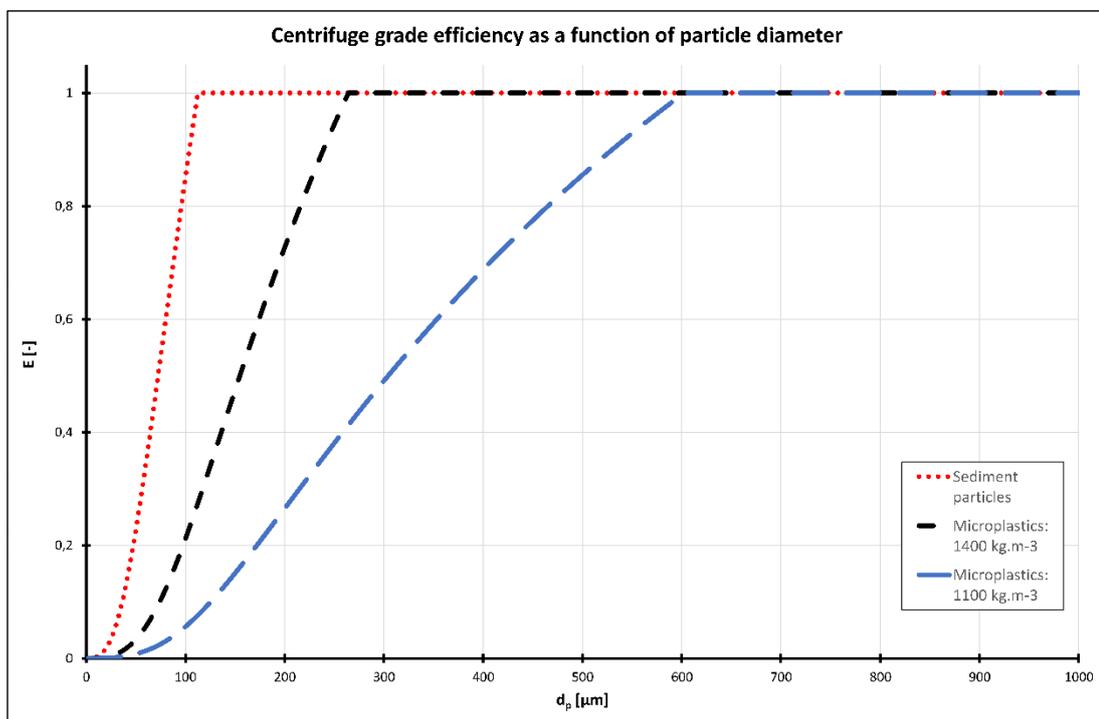


Figure 1. Overview of the theoretical grade efficiency curves associated with a conventional decanter centrifuge operating at optimal conditions. The simulations take into account the best-fitted drag model as developed by Dioguardi et al. [11]. An estimated average value is taken for the corresponding shape factors for the sediment and microplastic particles independently. Three curves are presented: (1) the grade efficiency of sediment particles (red dots), (2) the grade efficiency of microplastics with an average density of 1400 kg.m⁻³ (black short dashes), and (3) the grade efficiency of microplastics with an average density of 1100 kg.m⁻³ (blue long dashes).

The explorative (froth) flotation experiments revealed that strongly turbid flow regimes are unfavourable for the separation performance, that clogging of the air spargers by sediment particles is a critical impediment and that the air bubble size is a particularly important parameter. However, across all experiments polarity proved to be an effective separation factor for the microplastics/sediment mixture. By optimizing the froth flotation technique, we achieved an excellent separation performance when dealing with sediment mixtures with a sand particle size between 63 μm and 2 mm and a microplastic concentration of 1000 particles/kg sediment. For sediment mixtures with a sand particle size smaller than 63 μm , the sediment entrainment in the concentrate stream increased from approximately 0,1 m% to 5 m%. The microplastic recovery rate, however, remained nearly constant at 95 %.

Acknowledgement

This work was financially supported by the Moonshot SBO project PREFER.

References

- [1] M. Wagner and S. Lambert, *Freshwater Microplastics: Emerging Environmental Contaminants?* 2017.
- [2] P. Sundt, P. E. Schultze, and F. Syversen, "Sources of microplastic- pollution to the marine environment Project report," 2014.
- [3] L. G. A. Barboza and B. C. G. Gimenez, "Microplastics in the marine environment: Current trends and future perspectives," *Mar. Pollut. Bull.*, vol. 97, no. 1–2, pp. 5–12, Aug. 2015, doi: 10.1016/J.MARPOLBUL.2015.06.008.
- [4] H. S. Auta, C. U. Emenike, and S. H. Fauziah, "Distribution and importance of microplastics in the marine environment: A review of the sources, fate, effects, and potential solutions," *Environ. Int.*, vol. 102, pp. 165–176, May 2017, doi: 10.1016/J.ENVINT.2017.02.013.
- [5] Q. Zhang *et al.*, "A Review of Microplastics in Table Salt, Drinking Water, and Air: Direct Human Exposure," *Environ. Sci. Technol.*, vol. 54, no. 7, pp. 3740–3751, 2020, doi: 10.1021/acs.est.9b04535.
- [6] S. L. Wright, R. C. Thompson, and T. S. Galloway, "The physical impacts of microplastics on marine organisms: A review," *Environ. Pollut.*, vol. 178, pp. 483–492, Jul. 2013, doi: 10.1016/J.ENVPOL.2013.02.031.
- [7] L. G. A. Barboza, A. Dick Vethaak, B. R. B. O. Lavorante, A. K. Lundebye, and L. Guilhermino, "Marine microplastic debris: An emerging issue for food security, food safety and human health," *Mar. Pollut. Bull.*, vol. 133, pp. 336–348, Aug. 2018, doi: 10.1016/J.MARPOLBUL.2018.05.047.
- [8] L. Van Cauwenberghe, A. Vanreusel, J. Mees, and C. R. Janssen, "Microplastic pollution in deep-sea sediments," *Environ. Pollut.*, vol. 182, pp. 495–499, Nov. 2013, doi: 10.1016/J.ENVPOL.2013.08.013.
- [9] C. Sherrington, "Plastics in the Marine Environment," 2016.
- [10] A. A. Koelmans, M. Kooi, K. L. Law, and E. van Sebille, "All is not lost: deriving a top-down mass budget of plastic at sea," *Environ. Res. Lett.*, vol. 12, no. 11, p. 114028, Nov. 2017, doi: 10.1088/1748-9326/aa9500.
- [11] F. Dioguardi, D. Mele, and P. Dellino, "A New One-Equation Model of Fluid Drag for Irregularly Shaped Particles Valid Over a Wide Range of Reynolds Number," *J. Geophys. Res. Solid Earth*, vol. 123, no. 1, pp. 144–156, 2018, doi: 10.1002/2017JB014926.
- [12] E. Angu, J. Drelich, J. Laskowski, and K. Mittal, *Apparent and Microscopic Contact Angles*. Utrecht: CRC Press, 2000.
- [13] A. Borysenko *et al.*, "Experimental investigations of the wettability of clays and shales," *J. Geophys. Res. Solid Earth*, vol. 114, no. 7, pp. 1–11, 2009, doi: 10.1029/2008JB005928.