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Interactions between microplastics and phytoplankton aggregates: Impact on their respective fates

Marc Long, Brivaëla Moriceau *, Morgane Gallinari, Christophe Lambert, Arnaud Huvet, Jean Raffray, Philippe Soudant

Laboratoire des Sciences de l'Environnement MARin (LEMAR), UMR 6539 CNRS/Ifremer/IRD/UBO, Institut Universitaire Européen de la Mer (IUEM), Technopôle Brest-Iroise, 29280 Plouzané, France

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ABSTRACT

Plastic debris are resistant to degradation, and therefore tend to accumulate in marine environment. Nevertheless recent estimations of plastic concentrations at the surface of the ocean were lower than expected leading the communities to seek new sinks. Among the different processes suggested we chose to focus on the transport of microplastics from the surface to deeper layers of the ocean via phytoplankton aggregates that constitute most of the sinking flux. Interactions between microplastics and aggregates were studied by building a new device: the flow-through roller tank that mimics the behaviour of laboratory made aggregates sinking through a dense layer of microplastics. Three types of aggregates formed from two different algae species (the diatom *Chaetoceros neogracile*, the cryptophyte *Rhodomonas salina* and a mix) were used as model. With their frustule made of biogenic silica which is denser than the organic matter, diatom aggregates sunk faster than *R. salina* aggregates. Diatom aggregates were on average bigger and stickier while aggregates from *R. salina* were smaller and more fragile. With higher concentrations measured in *R. salina* aggregates, all model-aggregates incorporated and concentrated microplastics, substantially increasing the microplastic sinking rates from tenths to hundreds of metres per day. Our results clearly show that marine aggregates can be an efficient sink for microplastics by influencing their vertical distribution in the water column. Furthermore, despite the high plastic concentrations tested, our study opens new questions regarding the impact of plastics on sedimentation fluxes in oceans. As an effect of microplastic incorporation, the sinking rates of diatom aggregates strongly decreased meanwhile the sinking rates of cryptophyte aggregates increased.

1. Introduction

Plastics are highly persistent materials that tend to accumulate in the environment. As the plastic industry grows, plastic debris are becoming more and more abundant and can be found in every ocean. For example, an increase in microplastic concentrations has been reported in the Pacific Subtropical Gyre over the last 30 years (Wright et al., 2013). Microplastics, defined by the US *National Oceanic and Atmospheric Administration* (Wright et al., 2013) as particles smaller than 5 mm, are a large but mainly ignored portion of the plastic debris. Two main categories of microplastics (MPs) have been defined depending on their origin. Primary MPs originate from cosmetics, paints, textiles in household wastewaters or as pellets from plastic industry. Secondary MPs result from macroplastic (size bigger than 5 mm) fragmentation mainly caused by UV, waves or physical abrasion (Andrady, 2011). Like other plastic debris, MPs are observed in all parts of the ocean, from the surface layer to the sediment (Claessens et al., 2011; Graham and Thompson, 2009; Mohamed Nor and Obbard, 2014; Thompson et al., 2004), as deep as 4844 m in Porcupine abyssal plain (Van Cauwenberghe et al., 2013) and also in diverse levels of the trophic web (Dantas et al., 2012; Eriksson and Burton, 2003; Jantz et al., 2013). Common techniques and mesh size used for MP sampling at sea limit measurements to debris bigger than 333 μm (Deltares, 2011), however data on smaller MP particles are becoming available (Desforges et al., 2014; Mohamed Nor and Obbard, 2014). Concentrations of MP have been found as high as 10,000 particles m^{-3} on the Belgian coast (Van Cauwenberghe et al., 2014) and even 102,000 particles m^{-3} in Swedish waters (Norén, 2007). MP repartition depends on various anthropogenic parameters, such as plastic inputs and human activity (marine transport, plastic industry, tourism, wastewater…), as well as environmental parameters like biofouling, hydrodynamics, wind, currents, local climate conditions and even seasonal variations (Barnes et al., 2009; Lima et al., 2014). Models based on MP data and on hydrodynamics have attempted to explain MP distribution and transport in the surface (Law et al., 2010) or in the bottom (Ballent

Abbreviations: ESD, equivalent spherical diameter; MP, microplastic; SE, Standard Error; TEP, transparent exopolymer particles.

* Corresponding author at: UMR 6539 CNRS/IRD/UBO/Ifremer, Laboratoire des Sciences de l'Environnement Marin (LEMAR), Institut Universitaire Européen de la Mer (IUEM), Technopôle Brest-Iroise, 29280 Plouzané, France.
E-mail address: moriceau@univ-brest.fr (B. Moriceau).

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et al., 2013) layers and have shown significant seasonal changes linked to climatic conditions. Recent studies also revealed low plastic concentrations at sea in comparison with expectations (Cózar et al., 2014; Eriksen et al., 2014) and suggested that deep sea may be a major sink (Woodall et al., 2014). Our understanding of the MP would clearly benefit from a better comprehension of the fate of these particles.

Vertical fluxes of plastics are not only dependent on the density of the particles, for example hydrodynamics can re-suspend MP from the benthos or mix particles from the surface into the water column (Collignon et al., 2012). But most of the time sinking is linked to density and MP particles with high densities are transported downward, while buoyant ones (46% of plastic particles according to US EPA, Environmental Protection Agency, 2006) mostly stay at the surface (Barnes et al., 2009). Processes like biofouling can modify the density of particles and buoyant particles may sink to the bottom because of an increase in density due to colonization by microorganisms (Barnes and Milner, 2005). Among these are microalgae, which have been found attached to MP particles (Zettler et al., 2013). This sticking ability has been demonstrated by Bhattacharya et al. (2010) and Long et al. (2014). Many algae excrete polysaccharides especially at high cell concentrations or when they are stressed for example by light and nutrient limitations (Staats et al., 2000; Passow, 2002; Underwood et al., 2004). Exopolysaccharides may coagulate due to turbulence to form sticky particles named transparent exopolymer particles (TEPs) (Engel, 2000; Passow, 2002) and with sufficient stickiness, collisions between microalgae and TEP result in cell aggregation. While some large microalgae can sink as free cells, most of the time aggregates are the main vessel for vertical transport of phytoplankton cells and detritus to the ocean floor (Moriceau et al., 2007; Thornton, 2002; Turner, 2002; Kranck and Milligan, 1988). MPs could potentially be incorporated into marine aggregates (Wright et al., 2013), which would constitute a vertical pathway for MPs through the water column. In addition, marine aggregates are an important source of food for phytoplankton grazers and higher trophic levels and the incorporation of MP in aggregates and their transfer to the sea floor may have a significant impact on marine biota. Such ingestion of plastics by marine organisms has been demonstrated at different levels of the food chain (Boerger et al., 2010; Carson, 2013; Cole et al., 2013; Farrell and Nelson, 2013; Fossi et al., 2012; Graham and Thompson, 2009).

This study aims to evaluate the impact of algae aggregation on MP repartition/distribution and vertical fluxes as well as the impact of MP on aggregate sinking rates. Two species of microalgae were selected for exposure to MP: the diatom *Chaetoceros neogracile* and the cryptophyte *Rhodomonas salina*. The first is known to form aggregates (Alldredge and Silver, 1988) and both are known to stick on large MP (Zettler et al., 2013). A specially designed flow-through roller tank, derived from the traditional roller tanks used for aggregation experiments (Shanks and Edmondson, 1989), was constructed for this study.

2. Material and methods

2.1. Flow-through roller tank

The flow-through roller tank is a hybrid between the roller tank commonly used to promote aggregation since Shanks and Edmondson (1989) and a flow through reactor often used in dissolution studies (Chou and Wollast, 1984; Rickert et al., 2002; Van Cappellen and Qiu, 1997). The flow-through roller tank (Fig. 1) was built by modifying the roller tank as described by Shanks and Edmondson (1989). The tank was drilled on both flat sides. The holes were diametrically opposed to avoid the inflow solution to be immediately pumped out, allowing the best mixing possible of the inflow with the tank water. Each opening was connected with a compact non-spill valve (1/8 hose barb valved panel mount coupling insert in Acetal, provided by Colder Product Company). Each valve was connected via a Tygon® tube, to another connexion made of two different parts: a 1/8 hose barb valved panel mount coupling insert stuck on the center of each side that turns with the tank. The latter is eventually coupled (not shown in Fig. 1) to a 1/8 hose barb valved panel mount coupling body (in Acetal, provided by Colder Product Company). This last valve is fixed to the roller table allowing the free rotation of the tank while the Tygon® tube connected to the peristaltic pump is kept immobile. Through this last Tygon® tube, the peristaltic pump injects a constant flow of solution with a known composition and flow rate. Using this experimental system, aggregates are continuously sinking in a medium that can be progressively renewed and controlled. This technique also allows sampling of the medium without creating turbulence or bubbles that may disrupt aggregates and their sinking. Aggregates were formed in these tanks in a first step without flow. Later in the experiment, a very low flow rate that renews the tank water in 6 h (4 L/6 h) was used and the sinking dynamic of aggregates maintained in the flow-through roller tank was not changed. In particular no changes in trajectory or deflection, even close to the input/output of flow, were observed.

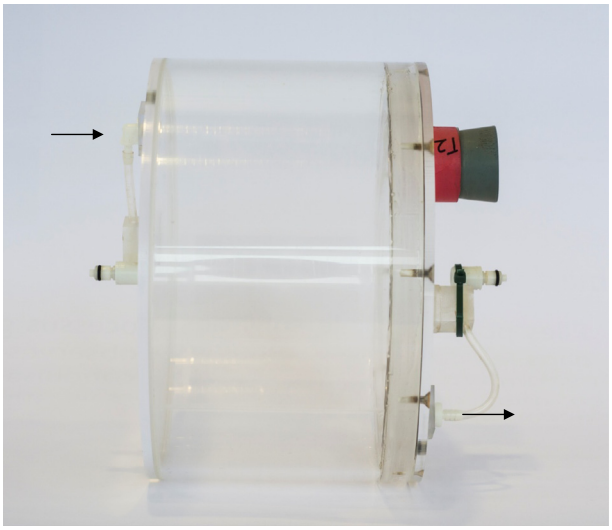

Fig. 1. Photograph of a flow-through roller tank. Input and output are diametrically opposed for a better mixing. The flows are illustrated by the two arrows.

2.2. Experimental protocol

2.2.1. Phytoplankton cultures

R. salina (cryptophyceae of 12 μm) and *C. neogracile* (bacillariophyceae of 5 μm) obtained from the Scottish Marine Institute were grown in f/2 medium (Guillard, 1975) prepared in filtered (0.22 μm) seawater from the bay of Brest (Brittany, France) with a density of 1.025 kg L^{-1}. Cells were grown in 6 L glassware round bottom flasks at 16 °C, under a 12 h/12 h photoperiod and an irradiance of 92 ± 13 μmol photons m^{-2} s^{-1}. Once cultures reached the stationary phase, i.e. when the cell concentration remained stable, the cells were maintained in the dark for few days to provoke light limitation stress, expected to promote TEP excretion. Three different mixtures were prepared. The first two were monocultures: one with *C. neogracile* at a cell concentration of 400,000 cells mL^{-1} and the other with *R. salina* at 100,000 cells mL^{-1}. The third was a combination of the two cultures with *C. neogracile* at 160,000 cells mL^{-1} and *R. salina* at 300,000 cells mL^{-1}.

2.2.2. Aggregate formation

In three sequential experiments, each of the three mixtures of algae was transferred to two 5460 mL cylindrical tanks for aggregation trials. One was the flow-through roller tank and the other a non-modified roller tank (Shanks and Edmondson, 1989). During aggregation, the two tanks were kept in the dark at 16 °C. Motorized roller tables, as

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