

Interaction, Adhesion and Aggregation of Microplastic/ Nanoplastic Particles: Effects of Plastic Polymer Type

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Received January 27, 2023; revised and accepted July 5, 2023

Abstract: Microplastics (MP; <5 mm) and nanoplastics (NP; <1000 nm) are now ubiquitous in nature as they have been released into inland water, lakes, rivers, and marine water through direct discharge, runoff from upland watersheds, and other terrestrial environments. To understand the aggregation of MP and NP in the last decade, surveys and analytical efforts were undertaken. The MP and NP particles in the soil and water environments interact with other natural and engineered nanoparticles, which in turn form aggregates. These plastic particles could form homoaggregates and/or heteroaggregates depending on pH, ionic type, ionic valence, salt concentration, shape and size of the plastic particles, and the polymer type of MP. Differences in MP aggregation due to differences in plastic polymer type are not yet well documented. This review is conducted to investigate the effect of polymer type on the aggregation of MP and NP particles in the presence of various aggregation conditions.

Key words: Aggregation, micro- and nano-plastics, polymer type, soil and water environment.

Introduction

Plastics are widely used in the modern world due to their low cost and availability (Alimi et al., 2018; Zhang et al., 2020). Upon environmental exposure, the plastic particles are degraded into tiny sizes called microplastics (MPs) and nanoplastics (NPs). The MP and NP micro- and nano-plastics are synthetic polymers that come in a wide variety of sizes, forms, and chemical compositions (Alimi et al., 2018; Wagner et al., 2014). There are different types of plastics, and the most common are polyethylene (PE), polystyrene (PS), polyvinyl chloride (PVC), nylon, polyethylene terephthalate (PET), polypropylene (PP), polytetrafluoroethylene (PTFE), and polyurethane (PUR). The classical Derjaguin-Landau-Verwey-Overbeek (DLVO) theory explains the aggregation of MP and NP particles (Cai et al., 2018; Li et al., 2018; Shams et al., 2020), whereas the

chemical composition of particles affects their surface behaviour and plays a role in aggregation. The MP and NP aggregations determine the transport and overall fate of these particles in aquatic environments (Alimi et al., 2018).

When two or more MP particles move in the same direction toward each other in order to collide, aggregation happens (Zhang, 2014). Both homoaggregation and heteroaggregation of MP of the same type and different types, respectively, may occur in laboratory conditions and natural aquatic systems (Alimi et al., 2018). MP particles form heteroaggregates with other solid components like natural minerals and organic colloids, inducing floatation, sedimentation, and/or resuspension in a natural aquatic system (Long et al., 2017; Singh, 2019). The homoaggregation and heteroaggregation of MP particles affect the transport and sedimentation of MP and are influenced by other

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natural colloids, suspended solids, solution pH, ionic strength, and organic matter concentration in aquatic systems (Cai et al., 2018; Li et al., 2018; Shams et al., 2020; Wang et al., 2021). The interparticle interactions that induce the homoaggregation and heteroaggregation of MP and NP cause particle sedimentation and affect particle transport in aqueous media and porous channels (Figure 1).

Many experimental investigations show the effect of pH, dissolved organic matter, particle concentration, and ionic strength on the adhesion and aggregation of MP and NP (Cai et al., 2018; Hakim and Kobayashi, 2021; Li et al., 2018; Shams et al., 2020). Some of the previous literature showed the heteroaggregation of MP polymers and polyethylene with clay (Wang et al., 2013). Very few experimental investigations demonstrate the comparative aggregation behaviour of different MP/NP under similar natural environmental and laboratory

conditions (Li et al., 2020; Shams et al., 2020). In this case, an extensive experimental investigation is required to investigate the effect of MP or NP polymer type on the aggregation and ultimately the MP transport. The aggregation behaviours of various MP and/or NP particles, i.e., polystyrene (PS), polyethylene (PE) and polypropylene (PP), have been discussed in this review to establish a comparative evaluation and to determine the effect of MP plastic polymer type on MP and NP aggregation.

Aggregation of Polystyrene, Polyethylene and Polypropylene Plastic Particles

Salt Induced Aggregation of MP and NP

Early investigation in a Coulter Counter-T model using a 30 μm orifice found the polystyrene latex of 1 μm diameter formed aggregates that were around 1600-fold larger than primary particles (Ives and Debouni,

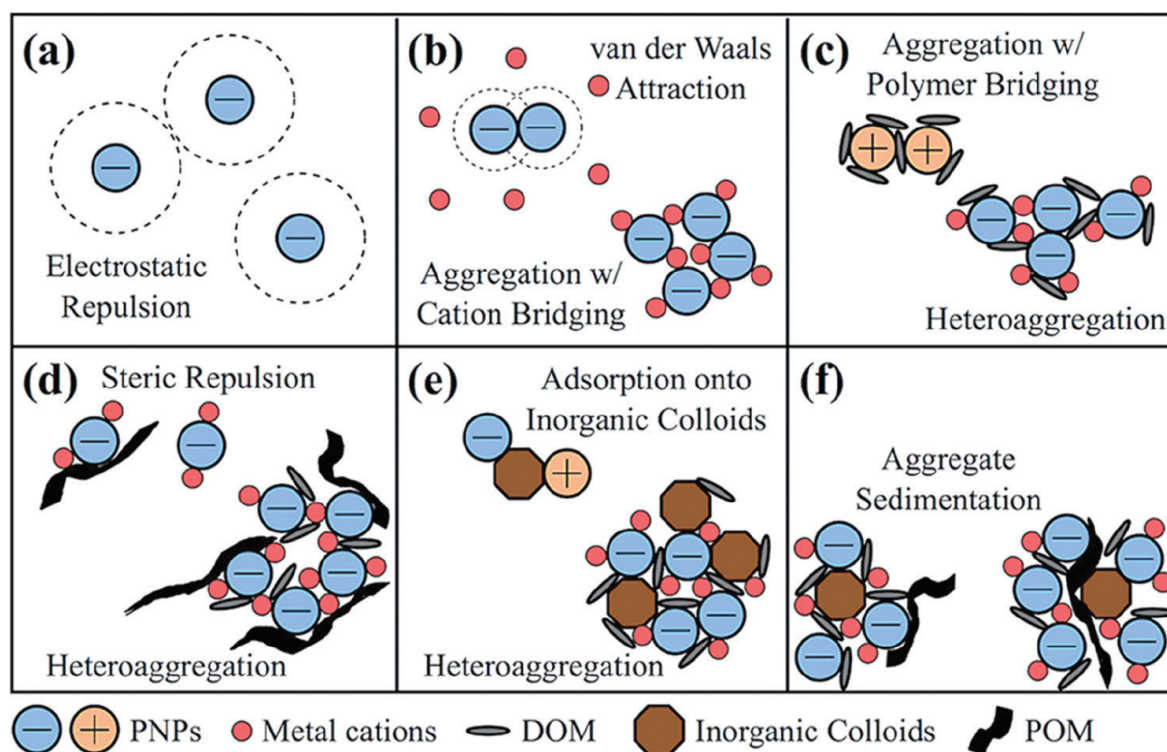


Figure 1: A schematic diagram shows the interactions between microplastic and/or plastic nanoparticles (PNPs) and other suspended or aqueous components. (a) The electrostatic repulsion of similarly charged plastic particles prevents particle aggregation (b) The metal cations in the suspension increase particle aggregation through the van der Waals attraction between particles and adsorption/cation bridging. (c) The natural polymer, dissolved organic matter (DOM), causes polymer bridging between the particles, leading to heteroaggregation. (d) Adsorption of particulate organic matter (POM) on the plastic micro and nanoparticles causes steric repulsion between micro- and nanoplastic particles, which reduces aggregation. (e) Adsorption of micro-nanoplastic particles onto inorganic colloids leads to heteroaggregation. (f) larger size aggregates and the higher aggregation rate increase the sedimentation of plastic particles (source: Brewer et al., 2021, *ACS EST Water* 1, 1, 48–57, reused with permission under the Creative Commons Attribution License).

1979). The aggregation of polystyrene latex spheres was confirmed using the light scattering technique (Lips et al., 1971), along with dynamic light scattering (DLS), transmission electron microscopy (TEM) and cryogenic scanning electron microscopy (Cai et al., 2018; Shams et al., 2020). Carpineti et al. (1990) observed the salt-induced aggregation of polystyrene latex particles. The shear-induced aggregation in a Couette-flow system of polystyrene latex spheres was reported in a highly electrolyte environment of fractal dimension 2.5 (Oles, 1991). This higher fractal character indicates compact aggregates of polystyrene latex particles. However, a relatively lower fractal dimension of 2.1 is confirmed for salt-induced polystyrene aggregates in the presence of hydrophobic polymers (Kjønksen et al., 1999). Moreover, Kobayashi et al. (2004) observed a floc strength of 2 nN for a polystyrene latex sphere in KCl solution through floc breakup by a converging flow. Hakim and Kobayashi (2021) confirmed the aggregate strength of 1.87 nN for polyethylene microspheres in the presence of humic acid and CaCl_2 solution. The higher aggregate strength for polystyrene particles than that of polyethylene particles indicates the effect of plastic polymer type on the MP/NP aggregation. The stability of aged PE particles is higher than that of pristine PE in a monovalent salt solution (Wang et al., 2023). The cation types affect the stability of PE microplastic particles and subsequently affect PE aggregation (Wang et al., 2023). Shams et al. (2020) found that polyethylene microplastics are more unstable than polystyrene in the aquatic environment, which means that the aggregation is triggered more in the presence of polyethylene than polystyrene. The possible mechanisms could be explained by the aromatic content in polystyrene

structures and their surface functionalisation, which leads to steric repulsions (Bastos and De las Nieves, 1994; Sastri, 2014).

According to Schmidtman et al. (2022), the MP particles in the natural environment mix with other micro- and nanoparticles to produce large-size heteroaggregates that cause the MP sedimentation and transport. In a mixing flow condition, the influence of salt concentration and particle surface charge on the heteroaggregation of polystyrene latex spheres was demonstrated (Sugimoto et al., 2022). Theoretical estimates of the attractive and repulsive heteroaggregation of polystyrene sulphate and amidine latex particles were established by Sugimoto and Kobayashi (2020). Additionally, recent work has reported on the anion-induced aggregation of polystyrene particles (Sugimoto et al., 2018). Additionally, charge regulation has a significant impact on the heteroaggregation of charged and neutral polystyrene nanoplastics (Trefalt et al., 2020).

Clay and Nanoparticles Induce MP and NP Aggregation

Kobayashi et al. (2013) exhibited the coagulation of polystyrene sulphate and amidine latex in the presence of aluminium silicate clay, Imogolite. Schmidtman et al. (2022) used scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) to confirm the heteroaggregation between PS particles and ferrihydrite. According to Schmidtman et al. (2022), the ferric (oxy)hydroxides stimulate heteroaggregation with PS and coat the negatively charged PS surface with positively charged ferrihydrite particles, causing charge reversal (Figure 2). Under various salt solutions, Wang et al. (2023) studied the homoaggregation of PE

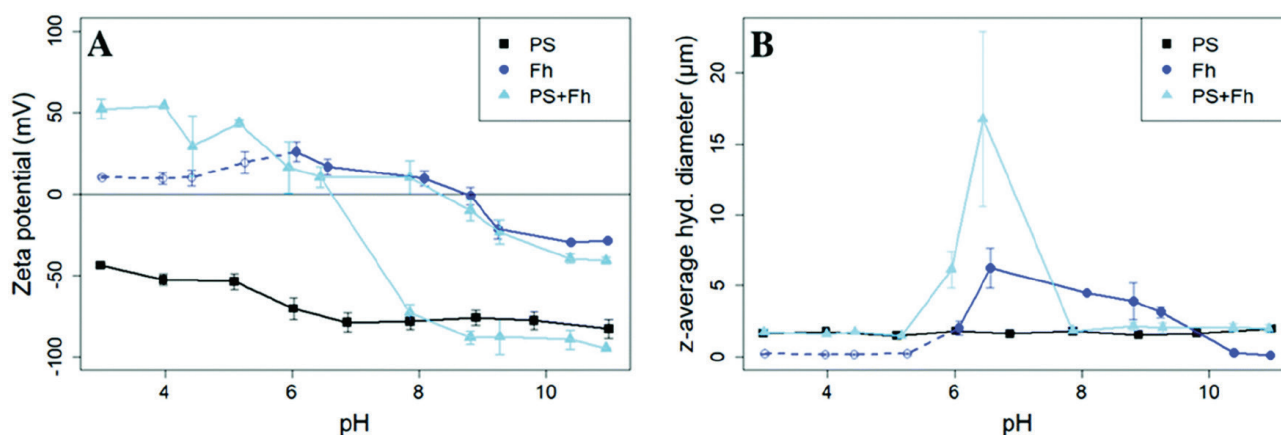


Figure 2: Zeta potential (A) and z-average hydrodynamic diameter (B) of PS (10 mg L^{-1} , $I = 10 \text{ mM}$, black squares), ferrihydrite (Fh, 10 mg L^{-1} , $I = 10 \text{ mM}$, dark blue circles), and samples with PS + ferrihydrite (PS + Fh, 10 mg L^{-1} PS, 10 mg L^{-1} Fh, $I = 10 \text{ mM}$, light blue triangles) (Source: Schmidtman et al., 2022, *Environ. Sci.: Processes Impacts*, 24, 1782, reused under the Creative Commons Attribution License)

microplastics and the heteroaggregation-settling of PE with distinctive clay minerals (kaolinite, illite, chlorite, and montmorillonite). In heteroaggregation systems, electrostatic repulsion dominates the interaction of PE with clay minerals. However, the settling ratio of PE determined by clay minerals is not significantly affected by the type of clay mineral (Wang et al., 2023).

According to a recent investigation, polyethylene (PE), polypropylene (PP), and polystyrene (PS) microplastic particles interact and aggregate with silver nanoparticles (AgNPs) in aquatic environments (Li et al., 2020). According to Li et al. (2020), there were no significant interactions between the AgNPs and the polyethylene (PE) and polypropylene (PP), but the AgNPs were effectively removed by the PS microplastics. Greater interaction and capture efficiency for AgNPs are provided by the electrostatic interactions and loose structure of PS caused by the phenyl group. None of the studies, however, decisively show the homoaggregation of different microplastic polymers under the same laboratory and natural conditions to evaluate the impacts of different microplastic polymers at constant pH, salt concentration, and ion type. That is why the effect of microplastic polymer type on the aggregation of microplastic is still vague.

Aggregation of Polyethylene and Polypropylene Micro- and Nano-Plastic

Polyethylene is one of the most commonly used model polymers and is frequently found in marine environments (Burns and Boxall, 2018). Very few investigations focussed on the homoaggregation of polyethylene and polypropylene (Shams et al., 2020; Hakim and Kobayashi, 2021), but heteroaggregation is discussed in some cases (Herbort et al., 2018; Li et al., 2020). Leiser et al. (2020) observed stable aggregates of polyethylene particles in the presence of iron colloids in a hypolimnion. In the presence of natural organic matter (NOM), polyethylene plastics are more stable due to the steric repulsion of NOM than the salt-induced aggregation of polyethylene (Shams et al., 2020). The calcium (Ca^{2+}) and magnesium (Mg^{2+}) ions affect the hydrodynamic diameter of polyethylene nanoplastics more than those of sodium (Na^+). While there was no significant interaction between AgNPs with PE and PP, and these microplastics coexist with AgNPs with no visible changes (Li et al., 2020), Hakim and Kobayashi (2021) illustrate the effect of ionic valence on the naked eye and microscopic aggregation of polyethylene microplastics.

However, adsorption-induced aggregation of colloidal particles is a well-established phenomenon supported by the screening of repulsive electrostatic interaction due to the adsorption of oppositely charged adsorbate on the particle surfaces (Petit et al., 1998; Szilagyi et al., 2014). According to the adsorption-induced aggregation mechanism, the polyethylene and polypropylene adsorption capacities have been discussed in previous investigations (Fu et al., 2021). The strong hydrophobic nature of PE and PP increases the adsorption of organic molecules (Fu et al., 2021), and ultimately, this adsorption could increase PE and PP aggregation through the screening of repulsive double layer interactions. A recent study demonstrates the effect of the polymer type of microplastic on soil aggregation (Lehmann et al., 2021). Polyethylene particles reduce the amount of new aggregation in soil, and polypropylene has a distinct negative impact on soil aggregation (Lehmann et al., 2021). To better understand the fate and transport of microplastics in soil and aquatic environments, future research should compare the aggregation behaviours of these three commonly used plastic particles.

Interaction and Aggregation of Micro/Nanoplastic Particles Depends on Polymer Type

Polyethylene (PE), polypropylene (PP) and polystyrene (PS), three different types of polymer nanoparticles, were investigated for their interactions with model lipid membranes (Bochicchio et al., 2017). However, at the membrane core, where PE has a tendency to aggregate, PP and PS behave differently. In this investigation, Bochicchio et al. (2017) looked at how plastic polymers interacted with heterogeneous membranes when ternary lipid mixtures were present. The three polymers exhibit fundamentally dissimilar behaviours: PE changes the geometry of the phase boundaries and depletes the liquid-ordered phase of cholesterol, whereas PP promotes lipid phase separation and PS stabilises it. In the simulation snapshot in Figure 3, it is clear that the PE aggregate engulfs some of the cholesterol molecules.

Moreover, another investigation on PE and PET oligomers' interactions with membrane bilayers found that the transport of plastic oligomers could be explained by passive diffusion. The simulation study suggests that large amounts of the PE and PET oligomers or longer-chain polymers can affect the membrane properties or show aggregation behaviour (Järvenpää et al., 2022).

The aggregation-dispersion of bacterial cells also depends on the polymer type (Kłodzinska et al., 2010; Zajac et al., 2023). A recent investigation manifests

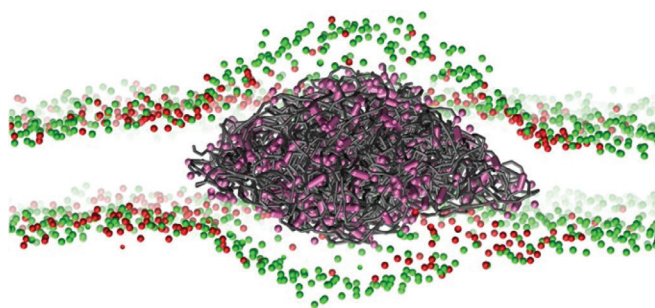


Figure 3: The ternary system+PE80 (9.7% in mass) shows cholesterol presence within the PE aggregate. Where the head groups are shown for the phospholipids: The DPPC (dipalmitoylphosphatidylcholine) is red, DLiPC(dilinoleoyl-phosphatidilcholine) is green. PE chains are shown in gray, and cholesterol beads within 0.5nm of PE are shown in pink colour. (Source: Bochicchio et al., 2017, *Sci Rep* 7, 6357, reused under the Creative Commons Attribution License).

the higher dispersion of bacterial cells (*Staphylococcus aureus* and *Klebsiella pneumoniae*) with the increase in PS particle concentration (Zajac et al., 2023). *Staphylococcus aureus* exhibits greater negative absolute zeta values than *K. pneumoniae* at any pH and PS particle concentration (Figure 4). The capacity for *S. aureus* to disperse in the presence of PS particles increases with increasing negative absolute zeta value, and vice versa, with decreasing aggregation potential. Under a constant pH condition, the absolute negative zeta potential value of bacterial cells was lower at higher sodium chloride concentrations. The results can be explained by the compression of the electric double layer by counterions like Na^+ , the reduction of the Stern potential, and the effective surface charge declining at high ionic strength (Zajac et al., 2023). Whereas there was an increase in the absolute negative value of the zeta potential for *Staphylococcus aureus*

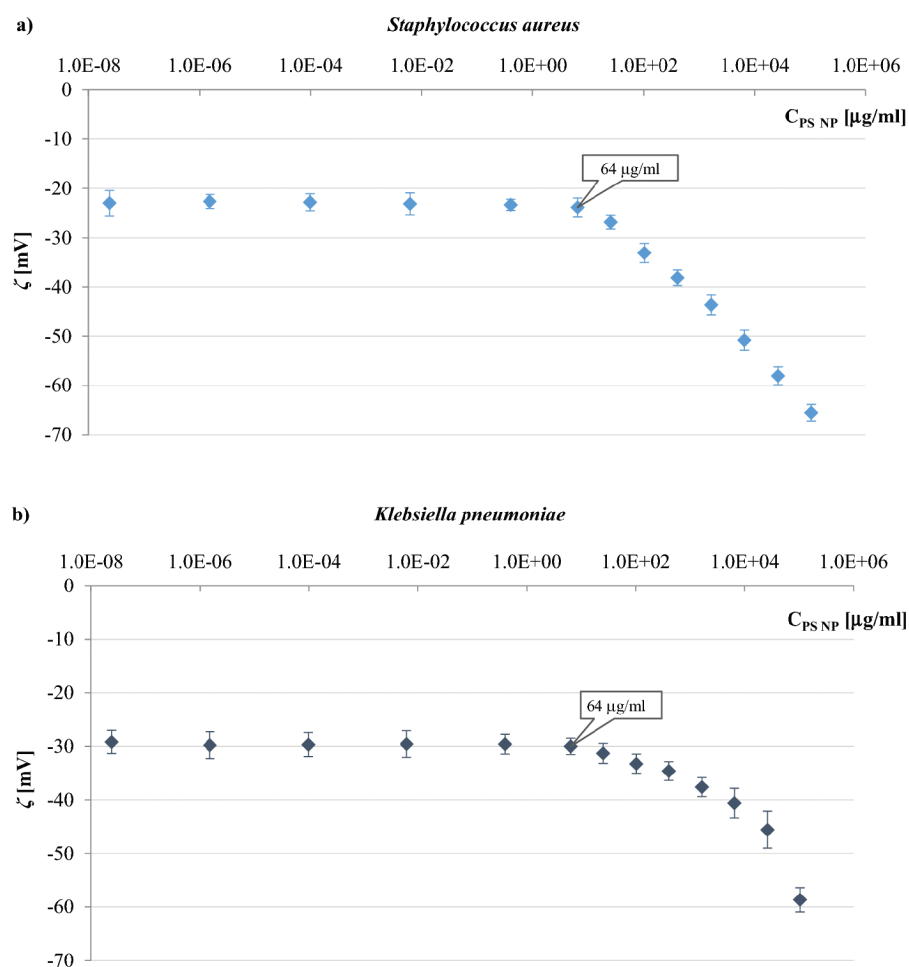


Figure 4: The zeta potential of *S. aureus* strain ATCC 6538 (a), and *K. pneumoniae* strain ATCC 4352 (b) obtained incubated overnight at different concentrations of PS nanoparticles in the presence of 0.3 mM NaCl (Source: Zajac et al., 2023, *Sci Rep* 13, 9552, reused under the Creative Commons Attribution License).

and *Escherichia coli* in the presence of polyethylene oxide (PEO). This higher absolute negative zeta value indicates lower adhesion and/or aggregation of *S. aureus* and *E. coli* in the presence of PEO (Kłodzinska et al., 2010). Moreover, the microbial biofilms could enhance the aggregation of microplastic and biogenic particles (Michels et al., 2018). In their experimental investigation of clean microplastic (polystyrene beads) with biogenic particles, Michels et al. (2018) found the first visible aggregates to the naked eye after about

1 day (Figure 5). The marine biogenic particles and microplastics form aggregate quickly, accelerating the formation of the gross aggregate and the incorporation of organic material in the natural water system.

Conclusion

The aggregation and fate-transport of micro- and nano-plastic particles is a serious environmental issue that affects public health. The aggregation of PE is more

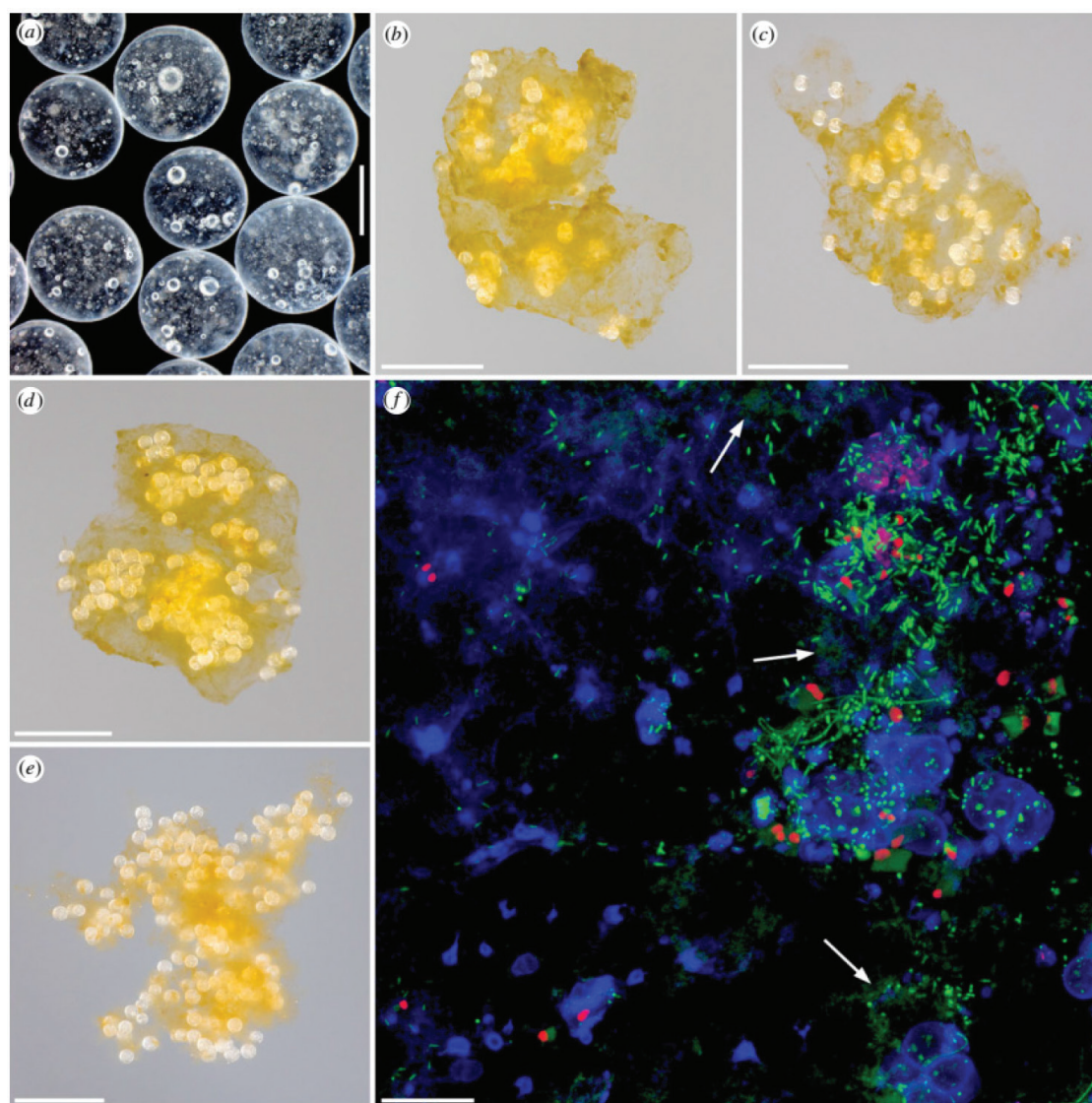


Figure 5: Aggregates consist of biogenic particles and microplastics. (a) Micrograph showing the polystyrene beads. (b-e) Aggregates of clean microplastics that are formed out of biogenic particles and microplastics (b) and biofilm-covered microplastics (c-e). (f) The Confocal laser scanning micrograph shows the biofilm on the surface of polystyrene beads within five weeks. Where the green color indicates structures containing nucleic acids, the blue color indicates structures containing polysaccharides, the red indicates the chlorophyll-containing structures, and the arrows in image f indicate exemplary structures that contain extracellular DNA. Scale bars: 500 μm (a), 5 mm (b-e) and 20 μm (f) (Source: Michels et al., 2018, *Proc. R. Soc. B* 285: 20181203, reused under the Creative Commons Attribution License).

prevalent than that of PS in aquatic conditions in the presence of natural organic matter. The aggregation of AgNPs is triggered more by PS than PE or PP. Regardless of the kind of polymer present in the MP/NP mixture, an increase in counter-ion valence causes an increase in MP/NP particle aggregation. The fate and transport of plastic pollutants in the environment may be revealed by a comparative analysis of MP/NP aggregation taking into account the MP/NP polymer types.

Conflicts of Interest

The authors declare no conflict of interest.

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