



The impact of nanoplastics on marine dissolved organic matter assembly

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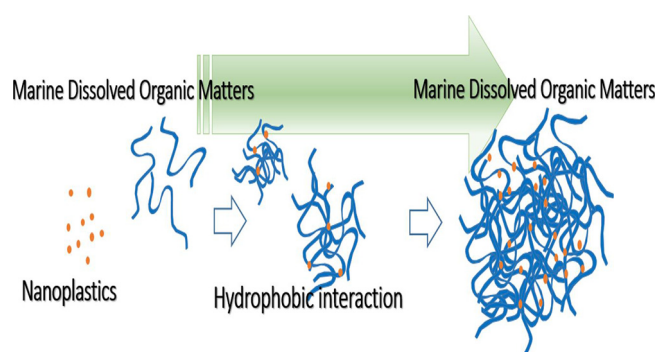
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HIGHLIGHTS

- In laboratory setting, 10 ppb nanoplastics (25 nm) can alter the DOM-POM transition in seawater samples.
- Hydrophobic interactions contribute to the facilitation of NP-DOM assembly.
- Results suggest that casual disposal of plastic waste potentially poses unforeseen disturbance to the marine carbon-cycle.

GRAPHICAL ABSTRACT



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ABSTRACT

The environmental impact of nanoplastics (NPs) released into natural aquatic surroundings is an increasing concern. NPs are widely generated from our daily waste disposal and eventually reach the ocean, wherein consequent influences on aquatic environments remain unclear. In this regard, there are few studies investigating NP-related ecological impacts. Comprising one of Earth's major carbon pools, marine dissolved organic matter (DOM) serves an essential role in global carbon dynamics. The spontaneous assembly of DOM into particulate organic matter (POM) plays important roles in the marine carbon cycle, and is involved in homeostasis of various ecological communities. Here, we report that 10 ppb NPs (polystyrene/polymethyl methacrylate, 25 nm) appeared in a water column accelerate the kinetic assembly rate of DOM-POM transition. NPs with various characteristics show similar influences on DOM assembly, and seawater samples collected from disparate sites were used to further confirm this unanticipated phenomenon. In this study, we demonstrated that hydrophobic interactions contribute to the facilitation of NP-DOM aggregations. Our results illustrate that NPs alter DOM-POM assembly, which may potentiate unanticipated perturbation to the largest marine carbon pool. Such effects would warrant increased vigilance on current practices of plastic usage and disposal.

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1. Introduction

Population and economic growth over previous decades have generated an ever-growing concern regarding ocean-pollution by anthropogenic plastic waste. Widely used in products ranging from packaging

and electronics to automotive and construction applications, this material has dominated the consumer market, and often winds up in the ocean as waste. Considering >5 million metric tons of plastic waste estimated to enter the ocean annually, the mass of marine plastic waste has been increasing remarkable (Jambeck et al., 2015; van Sebille et al., 2015; Siegfried et al., 2017). This plastic debris is distributed over coasts, sea surfaces and even accumulates on sea floors (Barnes et al., 2009; Hidalgo-Ruz et al., 2012). Alongside accumulated large plastic debris,

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plastic waste can potentially degrade and fragment into micron and nano-sized plastic dispersals (Barnes et al., 2009; da Costa et al., 2016). Based on oceanographic model prediction and expedition data, there are >250,000 tons of plastic debris disposed in the ocean, whereby most plastic waste was shredded into microplastics (Eriksen et al., 2014). In addition to gyres abrasion and light exposure, photo-oxidation comprises a major degradation pathway to convert plastic into nanosized particles in the natural environment (Lambert et al., 2013; Mattsson et al., 2015). Additional sources of nanoplastics (NPs) in the aquatic system stem from a wide spectrum of daily consumption, such as biomedical products and raw material used in industrial manufacturing (Koelmans et al., 2015). Comparative to debris in large size (>5 mm), smaller plastic waste becomes difficult to trace and may lead to unforeseen environmental consequences (da Costa et al., 2016). While numerous studies agree that plastic waste influences the marine ecosystem significantly, opinions differ as to component pathways (Gross, 2015). For instance, various organic pollutants and chemicals adsorbed onto dispersed plastic fragments can enter the marine food web through ingestion and biomagnification (Cole et al., 2011; do Sul and Costa, 2014). Nanosized polystyrene particles, were found to directly permeate into biological lipid membranes leading to undesired responses such as inflammation and development defects (Della Torre et al., 2014; Rossi et al., 2014). In aquatic environment, the fate of NPs is still under exploration. For instance, 30 nm polystyrene NPs aggregates rapid in seawater (Wegner et al., 2012), and hydrophobic interactions and high surface area contribute to nonlinear adsorption of nanoplastics (Velzeboer et al., 2014a). The transportation of NPs will be changed due to hetero-aggregation with natural organic matter (Besseling et al., 2017). Few studies investigated the correlation between NPs and dissolved organic matter (DOM) concentration in aquatic environments, and current findings propose DOMs may change the transportation and distribution of NPs in aquatic environments. (Lambert et al., 2013; Besseling et al., 2017)

Marine DOM assembly plays essential roles in mediating oceanic carbon cycles, which are among the largest carbon sinks on Earth, whereby organic colloids can grow over time and reach several centimeters in size. Sinking process of marine particulate organic carbon (POC), the marine colloidal pump, drains dissolved organic carbon (DOC) from the ocean surface and transports this content into the deep ocean (Ducklow et al., 2001; Najjar, 2009). Sinking colloids serve as critical vehicles in vertical carbon flux (Verdugo et al., 2004), and 80% of current anthropogenic CO₂ is expected to be transported into ocean on a 1000 year timescale (Herzog, 2001). Degrading phytoplankton materials collide with various smaller particles such as DOM and transparent extracellular polysaccharides (TEP), then are able to grow into organic aggregations such as marine snow (Azam and Malfatti, 2007). Such aggregations serve as important nutrient sources in aphotic zones. Moreover, recent studies also identified the marine organic gels as a parameter for regulating rainfall over the ocean, especially in the open ocean (Orellana and Leck, 2015).

Considering increased concern toward NP-induced alterations in the marine carbon cycle, several important models have been investigated. For example, microplastic ingested by zooplankton have been shown to reduce the sinking rate of fecal pellets, potentially perturbing the biological pump which transports carbon from ocean surfaces to the deep sea (Cole et al., 2016). Organic matter such as humic and exopolymeric substances adsorbed onto plastic debris have been potentiated to change the bioavailability and sinking rate of plastic debris (Rummel et al., 2017). In the current study, we aim to understand the influences of NPs released in ocean on marine DOM-POM transition, and the possible underlying mechanisms. In our laboratory study, several nanosized plastics were utilized to investigate induced alterations of marine DOM-POM assembly. These assemblages, in turn, are closely associated with carbon flux and are highly interconnected with carbon dynamics in the ocean by potentially interfering with the colloidal pump and nutrient bioavailability in respective microbial communities.

2. Materials and methods

2.1. Water sampling and storage

Seawater samples were collected at the Puget Sound (WA, USA) near Friday Harbor Marine Laboratories (48.54619, -123.00761). Specific permission was not required to collect seawater sample < 5 L at this site). In this study, majority of experiments were conducted with Friday Harbor coastal seawater, particularly if no specific address was mentioned. Seawater samples from the Gulf of Mexico were collected at 2 and 1500 m depths, respectively. All samples were filtered through a 0.22- μ m fiberglass membrane (prewashed with 0.1 N HCl), treated with 0.02% sodium azide—a microbial biocide, then were stored in sterile and sealed bottles in the dark at 4 °C before use. DOC concentration (2.57 mg L⁻¹) of seawater from Friday Harbor Marine Laboratories was measured using a total organic carbon analyzer (Shimadzu TOC-VCSH).

2.2. DLS measurement and temperature perturbation

Microgel assembly was monitored with dynamic laser scattering (DLS) as described previously (Chin et al., 1998). Seawater aliquots (10 mL) were filtered through a 0.22- μ m membrane (low-protein binding Durapore®, Millipore) and poured directly into scintillation vials. Scintillation cells were placed in the goniometer of a Brookhaven laser spectrometer (Brookhaven Instruments, NY) and scattered fluctuation signals were detected at a 45° scattering angle. The autocorrelation function of scattering intensity fluctuations was averaged over a 12-minute sampling time. Hydrodynamic diameters of microgels were analyzed by CONTIN method (Provencher, 1982), and the average aggregation size were presented. Seawater samples with NPs were monitored at specified time-points to identify assembly size changes. In order to mimic the environmental parameters and probe the hydrophobic interaction of NP-DOM assembly, we experimented over a range of temperatures. Samples in sealed scintillation vials were incubated between 30 and 40 °C for 24 h before equilibrium microgel sizes were measured.

2.3. Nanoparticle preparation

Polystyrene and polymethyl methacrylate nanoparticles (Bangs Laboratories, IN, USA) were used in our study as model NPs. The primary size and surface area of these nanoparticles were 25 nm and 2.48×10^{14} μ m²/g (certificate provided by vendor). The size of these NPs were independently confirmed by DLS. In order to avoid undesired nanoparticle aggregations, thorough sonication was applied to nanoparticle stock solutions before experiments, as described in our previous study (Chen et al., 2010). Nanoparticles with different surface modifications (amine and carboxyl functional groups, respectively) were used to investigate the potential effect of surface charges on DOM assembly kinetics.

2.4. Zeta potential measurement

Zeta potentials of NPs were measured to evaluate the influence of surface electrostatic force on NP-DOM assembly. NPs were dispersed in artificial seawater (40 g/L, Sigma Aldrich, USA) and sonicated thoroughly before measuring. Samples were injected into the standard zeta cells and measured using Zetasizer Nano ZS (Malvern, USA). Three replicates were performed for each type of NP.

3. Results and discussion

The assembly kinetics of DOM-POM transition were monitored using DLS wherein particle size measurements were recorded as a function of time (Chin et al., 1998; Vergugo, 2012). Results indicate DOM

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