Microplastics and Nanoplastics in Aquatic Environments: Aggregation, Deposition, and Enhanced Contaminant Transport

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Abstract

Plastic litter is widely acknowledged as a global environmental threat and poor management and disposal lead to increasing levels in the environment. Of recent concern is the degradation of plastics from macro- to micro- and even to nanosized particles smaller than 100 nm in size. At the nanoscale, plastics are difficult to detect and can be transported in air, soil and water compartments. While the impact of plastic debris on marine and fresh waters and organisms has been studied, the loads, transformations, transport, and fate of plastics in terrestrial and subsurface environments are largely overlooked. In this review, we first present estimated loads of plastics in different environmental compartments. We also provide a critical review of the current knowledge vis-à-vis nanoplastic (NP) and microplastic (MP) aggregation, deposition, and contaminant co-transport in the environment. Important factors that affect aggregation and deposition in natural subsurface environments are identified and critically analyzed. Factors affecting contaminant sorption onto plastic debris are discussed, and we show how polyethylene generally exhibits a greater sorption capacity than other plastic types. Finally, we highlight key knowledge gaps that need to be addressed to improve our ability to predict the risks associated with these ubiquitous contaminants in the environment by understanding their mobility, aggregation behavior and their potential to enhance the transport of other pollutants.

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1.0 Introduction

Currently, the global production of plastics exceeds 320 million tons (Mt) per year,[1] with production expected to double in the next 20 years.[2] Of this, only 6-14% is recycled, meaning up to 86% ends up either in landfills (21–42%) or released into the environment due to mismanagement through a variety of pathways (Figure 1).[2-6] Indeed, with the widespread use of different plastics, the current era has been referred to as the *Plasticene*.[7] Plastic debris has been detected in air,[8] oceans,[5, 9, 10] soils,[11-13] sediments,[14, 15] and surface waters worldwide.[16] It is estimated that in Europe and North America, the amount of microplastics (MPs) transferred every year from wastewater treatment plants (WWTPs) to agricultural soils as biosolids is greater than the total burden of MPs currently present in ocean water.[3]

Plastics are produced through the polymerization of various monomers and additives resulting in a spectrum of characteristics such as polarity and "glassiness".[17, 18] These differences in composition will impact their affinity for other pollutants and potential risks associated with them.[19] The most commonly detected plastics in the environment are polyethylene (PE), polyvinyl chloride (PVC), polypropylene (PP), polyethylene terephthalate (PET) and polystyrene (PS).[20]

The presence of MPs in the environment had been largely overlooked until recently; however, the number of studies is now growing rapidly due to the global ubiquity of plastic and its potential threat to human health and biota. Large plastic debris breaks down to form macroplastics (herein defined as >25 mm in size), mesoplastics (5-25 mm), MPs (<5 mm) and nanoplastics (NPs) (<100 nm).[21, 22] There is no clear consensus on the definition of MP and NP size in the literature;[5, 23-27] in this review, we define MPs and NPs as plastic debris with diameters of 100 nm-5 mm and <100 nm, respectively.

MPs/NPs are either primary or secondary in origin. One example of primary MPs is the plastic beads used as exfoliants in personal care products.[28] Primary NPs have also been detected in facial cleansers.[29] When these consumer goods are used, MPs and NPs are released into waste streams, with 95–99% partitioning into biosolids or removed in oil skimming in WWTPs.[3, 11, 30, 31] Nonetheless, an estimated 8 trillion pieces of MP (including microfibers) enter the aquatic environment via WWTP effluents.[32] Other sources of primary MPs in the environment include industrial abrasives and accidental spills.[33, 34]

Secondary MPs are the unintentional product of larger plastics degrading in the environment due to weathering processes (e.g., hydrolysis, UV photodegradation, mechanical abrasion, biodegradation). Sources include car tires, agricultural plastic mulch, microfibers from textiles, and larger plastics in the ocean (e.g., plastic bags, bottles, ropes, nets).[33, 35-37] Secondary MPs can often be identified by their irregular shapes and changes to the chemical structure.[38]

The increasing load of plastic in the ocean has received considerable attention.[1, 10, 25, 33] It is estimated that there will be over 250 Mt of plastic accumulated in the ocean by 2025.[1, 39] Though the processes may be slow, plastic will inevitably break down into MPs and NPs due to exposure to UV radiation, mechanical abrasion, and wave action. The combination of primary and secondary sources leads to estimates that 5.25 trillion plastic particles contaminate the global sea surface,[40] the majority of which are less than 10 mm in size.[36]

In contrast to marine plastics, there is limited information on the behavior, fate, and loading of MPs and NPs in the terrestrial environment. MPs and NPs may be introduced to soils as a result of landfill leachate, agricultural mulches, application of wastewater biosolids to agricultural land, or by direct releases of secondary MPs and NPs from abrasion or maintenance of outdoor plastic goods and coated surfaces.[2, 13, 41-45] Following release into the environment, MPs and NPs may undergo various transformations commonly associated with natural or anthropogenic colloids;[46] namely, homo- and heteroaggregation, interactions with microorganisms and macromolecules (e.g., adsorption of proteins, natural organic matter) and biodegradation. These processes, as well as the mobility of the particles in natural soils and ground waters, are strongly influenced by porewater chemistry (e.g., pH, ionic strength, natural organic matter content) as well as the properties of the plastics (e.g., size, shape, chemical composition) and soil/aquifer sediments (e.g., composition, size distribution). While a large number of laboratory studies have examined the transformations and transport of natural colloids and engineered nanomaterials such as metals and metal oxides in terrestrial environments,[46-50] there is little data on how these environmental processes and conditions affect different types of NPs and MPs that are present in soils.

Natural colloids such as iron oxides and clays have been shown to affect the transport of contaminants such as radionuclides, pesticides, and polycyclic aromatic hydrocarbons in the subsurface environment.[51-56] Likewise, MPs and NPs can act as vectors for the transport of contaminants such as heavy metals[57-59] and persistent organic pollutants (POPs).[20, 60-65] Yet, the extent to which sorption of contaminants onto different types of MPs and NPs enhances or mitigates the environmental and health impacts of these pollutants remains unclear.

This paper provides a critical review of the existing scientific literature examining the aggregation and transport of NPs and MPs in soil and groundwater systems. First, we estimate the loads of plastics in different environmental compartments. Next, we critically assess existing studies on the aggregation, transport and contaminant sorption behavior of NPs and MPs in terrestrial and subsurface environments. Finally, the current gaps in knowledge precluding a comprehensive understanding of MP and NP fate and impacts are discussed.

1.1. From Macro to Micro to…Nano?

Macroplastics can degrade to form MPs through stresses that impact the structure and reactivity of the plastic polymer.[66, 67] Degradation of plastics can occur by multiple processes including hydrolysis, photodegradation due to UV exposure, mechanical abrasion by sand or wave action, and biodegradation.[68, 69] These processes may also act synergistically. For example, UV exposure leads to carbonyl group formation, rendering the materials more brittle and increasing the likelihood of mechanical degradation.[36] Degradation mechanisms are not uniform for all plastics; for instance, PS and PE are more prone to weathering by UV radiation than other plastics.[70] While the occurrence of MPs in the natural environment is increasingly well documented,[16, 68, 71, 72] to date, no study reports on the presence of NPs in aquatic or soil environments. This is mainly due to methodological challenges associated with detection and recovery of such small, carbon-based particles in complex natural matrices. Although there is no data available on environmental loads of NPs, weathering of macroplastics and MPs is expected to yield secondary NPs.[67] Indeed, a controlled laboratory study[66] shows that NPs and MPs (ranging in size from 30-2000 nm) are released during weathering of macroplastic (a polystyrene coffee cup lid) in a simulated marine environment. A recent study further reveals that consumer products such as facial scrubs can act as an unintentional yet important source of primary NPs to natural waters and soils.[29] Thus, although environmental levels of NPs are yet to be quantified, plastic nano-litter is expected to be as ubiquitous as its bulk counterparts.

2. Living in the Plasticene: Plastic in Every Corner of the Earth

Plastics can be found throughout the globe. Despite their ubiquity, the global cycling of MPs and NPs is not well understood. Figure 1 synthesizes the existing knowledge of global plastic cycling. Significant transport pathways are identified and loads in selected environmental compartments are reported; however, the relative flux of plastics from one compartment to another is often unknown or associated with large uncertainty. Despite numerous studies to date, wide ranges in reported concentrations are observed, representing both spatial variation and measurement uncertainty. For example, estimates of plastic loads in the oceans range six orders of magnitude,[14, 25, 73] while no comprehensive data exist for MPs in soils despite considerable agricultural use.[74, 75] Finally, atmospheric deposition of MPs and NPs is expected, but largely unexamined[76] except for few studies.[16, 77]

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2.1 Freshwaters Are Key Vectors for Microplastic Transport

Rivers are estimated to transport 70-80% of plastics that eventually arrive in the oceans, with primary inputs from mishandled debris either during manufacture and use, from agriculture and land, and WWTP effluent (Figure 1).[30] Concentrations of plastics in freshwater can rival marine levels, though there is great spatial variation, depending on proximity to urban or industrial sites, or the presence of WWTPs.[68, 78-80] Freshwaters are generally closer to plastics sources than marine waters and have more shoreline to retain particles, facilitating accumulation and mechanical degradation.[71] Branches, logs, and dams have been identified as local hotspots of plastics.[81, 82]

Several studies have focused on large rivers, such as the Chicago River, Rhine-Main, Danube, and Thames.[34, 79, 80, 83] In general, significant differences are seen in concentrations of MPs upstream of a point source versus downstream.[79] However, the presence of multiple sources along the length of a river makes identifying the origin of specific particles difficult.[80] Additionally, changes in flow due to bends, particularly deep or shallow sections, etc. can cause particle buildup and influence transport. Pulsed, accidental releases have been identified as a primary source of peak loading events.[34] Furthermore, periods of high flow are capable of both re-suspending particles that may have settled to the sediments and depositing MPs onto adjacent shorelines.[30]

WWTPs, ubiquitous along populated waterways, act as significant point sources of MPs to freshwaters. McCormick et al. demonstrated a 10-fold increase in plastic fibers downstream of a WWTP in the Chicago River[79] despite the fact that 95-99% of plastics partition into the wastewater biosolids.[16, 84, 85] Likewise, in a WWTP with tertiary treatment, as little as 0.1% of the incoming MPs and microfibers were released in the effluent water.[31] Although a relatively large fraction of MPs/NPs are expected to be trapped in wastewater biosolids, it is estimated that 520,000 tons/year of plastic waste is still released in wastewater effluents in Europe alone.[30] It is noteworthy that application of sludge can represent a significant source of MPs (and very likely NPs) to agricultural lands.

Lakes can act as temporary or long-term sinks of MPs. Areas of the Laurentian Great Lakes are as polluted as ocean gyres, but there are large spatial variations, both within a single lake and between lakes.[78] Transport in lakes is driven by currents, similar to rivers and streams, but also by wind patterns that can produce areas of seasonally high local MP concentrations.[86] Hoffman and Hittinger estimate plastic introduction into the Great Lakes at 10,000 metric tons/year.[87]

Estimates of plastic loading in rivers and lakes range from 10^{-5} to 10s of pieces/L (Figure 1).[30, 73, 79] Making precise estimates of plastic loading in lakes is difficult however, as sampling generally takes place at the water surface, while large concentrations of MPs can also exist below the surface, depending on the biological, physicochemical, and hydrodynamic conditions (e.g., plastic density, mixing of water column and aggregation/attachment of bacteria/algae).[81]

2.2 Aquatic Species Are at High Risk

With the high loads of MPs (and likely NPs) in natural waters, aquatic species are expected to experience chronic exposure and to potentially bioaccumulate the plastic particles.[88-90]

Invertebrates such as crustaceans, barnacles, polychaete worms, mussels, and amphipods have ingested MP fragments in controlled studies.[15, 91, 92] These tests often employ high MP concentrations, limiting their environmental significance.[1] A few researchers investigated plastic accumulation in organisms in their natural habitat confirming MP ingestion and accumulation in the gut and stomach of various species of fish,[90, 93] shellfish,[94] and fur seals.[95] Although there is a growing number of studies on MP accumulation in aquatic organisms, the (likely) biouptake of NPs has not been examined. The small size and organic composition of NPs present significant methodological challenges to their detection and quantification in complex biological samples. Techniques that may hold promise for future studies in this area include pyrolysis combined with chromatography, mass spectrometry, infrared spectroscopy, scanning electron microscopy with energy dispersive x-ray spectroscopy, field flow fractionation with pyrolysis and multi-angle light scattering.[96]

2.3 Plastic Loads in Soils and Sediments Are Less Understood

In comparison to aquatic environments, relatively few studies have investigated plastics levels in soils and sediments.[5, 68] Significant sources of MPs and NPs to soils are likely land application of biosolids, plastic agricultural products, and litter. Moreover, a considerable fraction of the global production of plastic waste (21-42%) (Figure 1) is stored in landfills,[11] often under poorly managed conditions that can result in release to soils. Not surprisingly, MPs and synthetic polymer fibers found in sewage sludge were still detectable up to fifteen years after being applied to soils.[12, 13] Indeed, the US market for agricultural plastic was estimated at $5.8 billion in 2012, including products such as plastic seed casings, ground covers, crop mulch, greenhouse coverings, labels, and wraps.[97] Despite this, little is known about the retention and fate of agricultural plastics after their use. Bioturbation by earthworms can increase soil plastic retention; particles were observed to move downward in the soil profile, with smaller particles being transported to a greater extent.[75, 98] Modeling the fate of plastics in biosolids applied to soils, Nizzetto et al. estimate that only 16-38% of deposited MPs are retained in soils.[3]

Analysis of river sediment cores suggest plastic accumulation over the past four decades.[99] In the Rein-Main river, plastics <5 mm were found in all sediments sampled, with loads up to 1 g/kg (4,000 particles/kg).[80] In general, less plastic mass but similar particle numbers – highlighting the fact that small particles are generally more mobile – were found in areas of lower population density and nature preserves. Despite this, no clear correlation was established between sediment MP levels and population density, industrial proximity, or WWTPs, illustrating the complex influences of a river system. Lastly, Browne et al.[68] found polyester and acrylic fibers used in clothing were present at over 250% greater concentrations in coastal sediments at historical sewage discharge locations versus reference sites.

Although soils – particularly agricultural lands – are expected to be important sinks for MPs and NPs, significant research is needed to better understand their loads, fate and potential for biouptake in these complex heterogeneous environments. Plastic laden soils also pose a risk for contamination of natural subsurface environments, including groundwaters that may be used as drinking water sources. Thus, there is also an urgent need to characterize the behavior and mobility of MPs and NPs in natural soils and subsurface environments such as groundwater aquifers.

2.4 Modeling with Incomplete Data

Because the data on MP and especially NP loads are limited and the uncertainty is high, there are few reliable models for MP (or NP) transport.[11] Jambeck et al.[39] calculated broad estimates of future land-based (terrestrial and freshwater) plastics entering the ocean based on waste management data, suggesting that between 100 and 250 million tons of plastics will be released into the ocean by 2025. On a finer scale, Nizzetto et al. developed the first mathematical study of MP fate in terrestrial environments and rivers using an integrated catchment model.[11] This work focused on the mechanisms of MP particle storage, entrainment and deposition in soils and streams to calculate the retention efficiency of soils and river sediments during MP transport toward the sea.[3, 11] One of the primary shortcomings of many models is the lack of 3-dimensional resolution, with models assuming that all particles exist at the water surface which does not account for the variability of plastic concentrations with depth.[83] Furthermore, validating models becomes difficult as not all plastics are captured due to limitations in sampling methods that are unable to detect the smallest particles.[87] To overcome these shortcomings, it could be of interest to implement transport and fate models that have recently been developed for engineered nanoparticles (ENPs).[100] Several models that take aggregation and heteroaggregation into account have been developed for ENPs that could be applicable to NPs.[101-103] Currently, the lack of robust models prohibits a comprehensive understanding of the risks posed by MPs and NPs and reflects the shortcomings in existing data sets of

MP and NP behavior and fate in environmental systems. To address this, considerable fundamental research is needed to characterize the aggregation and deposition kinetics of MPs and NPs over a broad range of environmental conditions.

3. Current State of Knowledge on Micro- and Nanoplastic Aggregation and Deposition

NPs and MPs in natural soils and waters will undergo various transformations (e.g., degradation, coating with environmental macromolecules) that will influence their aggregation, deposition, and transport. These processes will depend largely on the aquatic chemistry of the water column, aquifer porewaters, and sediment properties and will directly influence the environmental fate of the plastic particles.

Aggregation involves the transport of two particles towards each other to collide, followed by attachment. This can occur with two particles of the same type (homoaggregation), or two different particles (heteroaggregation). Deposition is the process of a particle attaching to a larger, immobile collector surface, such as an aquifer/sediment grain.[104] The fundamental mechanisms governing particle aggregation and deposition have been extensively described.[47, 105, 106] Once particles collide with each other (aggregation) or with a collector grain surface (deposition), the likelihood of attachment is controlled by van der Waals and electrical double layer forces described by the Derjaguin-Landau-Verwey-Overbeek (DLVO) theory,[107, 108] as well as non-DLVO interactions (including steric forces).[109, 110] The likelihood of attachment or "attachment efficiency" (α) is the ratio of collisions that result in attachment to the number of total collisions. If attractive forces dominate, the process is considered diffusion-limited and α approaches 1. Conversely, if $\alpha < 1$, repulsive forces influence the likelihood of attachment, and the process is considered reaction-limited. According to DLVO theory, increasing the ionic strength (IS) of a solution compresses the electrical double layer and decreases repulsive forces, resulting in a higher rate of aggregation or deposition. The minimum electrolyte concentration needed to completely destabilize a particle suspension is the critical coagulation concentration (CCC). The CCC represents the point at which α reaches unity, beyond which the aggregation rate is insensitive to an increase in IS.[111] Additionally, according to the Schulze-Hardy rule, multivalent electrolytes will have lower CCC values.

Besides water chemistry, particle-specific properties (e.g., size, density, shape, chemical composition, surface charge, surface coating), hydrodynamic conditions, and soil/sediment properties (e.g., grain size distribution, organic matter content)[104, 111, 112] also influence the potential mobility of particles in natural aquatic environments. Below, the existing literature on NP and MP aggregation and deposition has been summarized and critically analyzed.

3.1. Laboratory Studies Investigating the Aggregation of NPs and MPs.

Aggregation largely determines the fate, mobility, persistence, and bioavailability of particles in the environment. It is generally controlled by the IS and valence of the electrolytes in the surrounding medium; however, aggregation can also be impaired for NPs/MPs that are polymer-coated, either intentionally or incidentally.[46] Heteroaggregation, in which two or more different types of particles form aggregates, is more likely to take place than homoaggregation for MPs and NPs due to the overwhelmingly greater number of natural colloids.[46] While this has been shown for ENPs and natural colloids,[113] the impact of heteroaggregation on the state or fate of MPs and NPs is unknown. Table 1 summarizes the laboratory studies that have investigated the aggregation rates, CCCs and general aggregation behavior of MPs and NPs.

A large number of studies have examined the homoaggregation behavior of PS NPs and MPs. In general, the data in Figure 2 show that the particle-particle attachment efficiency (α_{pp}) increases with increasing IS due to compression of the electrical double layer, in agreement with the DLVO theory. For example, Wegner et al[88] report that carboxylated 30 nm PS particles in seawater rapidly aggregated to over 1000 nm in less than 30 minutes. Data in Figure 2 show how the electrolyte valence impacts the aggregation rates of PS NPs and MPs. For instance, the CCC for sulfonated PS particles was an order of magnitude greater in monovalent electrolytes compared to divalent electrolytes.[114] The importance of valence is further highlighted in Figure 2a, where trivalent cations destabilized carboxylate- and sulfate-modified PS at lower concentrations than divalent cations.[115-117]

A study by Ruiz Cabello et al.[118] shows that sulfate-functionalized PS behaves similarly in two different monovalent electrolytes at pH 4 (in NaCl and KCl, Figure 2b). Interestingly, Oncsick et al. also examined sulfate-functionalized PS, at pH 4 in NaCl, but report different behavior. The 530 nm sulfate-functionalized PS used by Oncsick et al.[117] (Figure 2b) is less stable in NaCl when compared to a 960 nm PS employed by Ruiz Cabello et al. The observed difference in particle stability may arise from differing surface charge densities and/or disparities in diffusion kinetics due to particle size.

Changes to the particles resulting from engineered functionalization or incidental coatings will also impact NP aggregation. Hierrezuelo et al.[119] show that a 270 nm sulfate-modified PS is more stable than a 200 nm amidine-modified PS under the same experimental conditions (open red squares and circles). Della Torre et al. observed that 40 nm carboxylated PS NPs rapidly formed aggregates of ~1700 nm in natural seawater, while 50 nm amino-modified PS NPs remained temporarily stable at ~90 nm, but moderately aggregated at longer times.[120] Sakota and Okaya demonstrated that increasing the extent to which PS particles are carboxylated increased stability, due to an increase in surface charge.[121] Similarly, the stability of sulfonated PS latex was observed to depend on surface charge density.[114] Thus, particle aggregation will not only depend on water chemistry but also on the particle surface functionalization.

Few studies have examined the effect of coatings on the stability of NPs and MPs. In Figure 2c, solid symbols represent coated NPs and MPs while open symbols represent uncoated particles. Although polymer coatings are generally observed to stabilize ENP suspensions due to steric or electrosteric stabilization,[47] their impact on NP and MP aggregation is not clear. For example, Hierrezuelo et al. report that 200 nm uncoated amidine-modified PS particles are less stable than those coated with either polyacryclic acid (PAA) or polystyrene sulfonate (PSS) polymers (Figure 2c, circles). However, they also show that a 270 nm linear polyethyleneimine (LPEI)- and polydiallyldimethylammonium chloride

(PDAPMAC)-coated sulfate-modified PS have a comparable stability to the bare particles (Figure 2c, squares). [119]

Natural organic matter (NOM), which is ubiquitous in natural waters, is expected to adsorb on the surface of plastic particles, as has been demonstrated for natural colloids and ENPs.[122-127] Studies on plastic aggregation in the presence of NOM are sparse,[128, 129] precluding generalizations of the impact of these diverse environmental molecules. Two studies[128, 129] show that PS NP aggregation is reduced in the presence of polysaccharides, humic and fulvic acids; however, additional research is needed to develop a more comprehensive understanding of the effects of these ubiquitous and heterogeneous environmental macromolecules on plastic stability.

The existing literature on the aggregation of spherical PS particles is generally in qualitative agreement with the DLVO theory of colloidal stability. Namely, the data in Figure 2 show that α_{pp} increases with increasing solution IS until the mass transport-limited aggregation rate is reached (where $\alpha_{\mathrm{pp}}=1$). In all studies, spherical particles are used; however, since a large proportion of MPs and NPs in the environment is expected to have variable and non-spherical shapes, the aggregation behavior of different-shaped plastics warrants further investigation. Moreover, plastic debris in the environment is not restricted to PS. While the aggregation behavior of PVC latex and polyurethane (PU) particles have been observed to generally follow the DLVO theory and Schulze-Hardy rule,[130, 131] little attention has been paid to these materials. Thus, future research should take into account the diversity of plastics to better understand environmental fate and associated risks.

3.2 Laboratory Studies Investigating the Deposition of NPs and MPs.

Different experimental approaches have been used to investigate NP and MP deposition kinetics in systems representative of the unsaturated (vadose) and water saturated zones of the subsurface environment.[111, 132] Fully or partially water-saturated columns filled with well-characterized granular media (*e.g.*, glass beads, sand, or soil) are commonly used to study particle transport and deposition by monitoring changes in the column effluent particle concentration as a function of time.[129, 132-163] Alternatively, the quartz crystal microbalance with dissipation monitoring (QCM-D) has been used to characterize NPs deposition onto model aquifer grain surfaces.[159] Nearly all studies on the deposition of NPs or MPs have focused on spherical PS particles modified with sulfate, amine or carboxyl groups (Table 2). Figure 3 summarizes data from some of these studies. As noted in the case of particle aggregation, the particle-collector attachment efficiency (α_{pc}) also increases with IS due to electrical double layer compression.

The porewater flowrate in subsurface environments has been shown to affect the transport of NPs and MPs.[160, 162] Generally, decreased plastic deposition is observed at high porewater velocities, in agreement with studies involving other types of colloids (*e.g.* titanium dioxide, fullerenes).[48, 162, 164, 165] Tong and Johnson observed a decrease in retention of PS MPs in columns packed with quartz sand as flow velocity increased.[164] This behavior was observed by others when the plastic particle and collector have the same charge (unfavorable condition).[166, 167]

The type of granular media will play a large role in determining the fate of NPs and MPs in the environment. Despite this, most studies have used clean glass beads and high purity quartz sand that poorly represent natural environments. Researchers that investigated the behavior of plastic particles in realistic media other than quartz sand and glass beads suggest that retention is much higher under 'dirty' conditions. Bouchard et al. showed that PS MP retention was greater in sediments from a creek in Georgia, USA than in pure Iota quartz sand.[137] The observed retention was attributed to the high aluminum hydroxide content and rougher surface of the sediment that can provide positive charge and localized sites for deposition, respectively. Additionally, using comparable media grain sizes, Quevedo and Tufenkji showed that PS NP retention was greater in an agricultural loamy sand than high purity quartz sand.[159] These studies suggest that retention is higher in more heterogeneous granular media, although further investigations using environmentally relevant granular materials are needed to establish a comprehensive understanding of the effect of geochemistry and grain size distribution on the transport of NPs and MPs.

Microorganisms and biofilms are ubiquitous in natural aquatic environments; yet, their impact on the mobility of NPs and MPs has not been well studied. Tripathi et al observed that PS NPs and MPs with different surface functionalities (carboxylate and sulfate) exhibit increased retention in columns packed with quartz sand coated with *Pseudomonas aeruginosa* biofilm.[158] Mitzel et al also reported higher retention of sulfate and carboxylate functionalized PS NPs when sand is coated with *Pseudomonas aeruginosa* biofilm. This trend is observed in the presence of other types of biofilms (*e.g.*, *Pseudomonas fluorescens, Stenotrophomonas maltophilia, Lactococcus lactis* etc.).[168, 169] The hydrophobicity of biofilm-coated sand can also influence the transport behavior of PS. For example, Mitzel et al observed dynamic NP transport behavior in sand coated with a hydrophilic biofilm, whereas NP transport was constant with time in the sand coated with a more hydrophobic biofilm.[163]

Few studies have examined the impact of NOM such as fulvic and humic acids on the transport and deposition of NPs and MPs in model subsurface environments.[150, 152, 156] Particles coated with NOM (Figure 3a, solid symbols) were generally observed to have lower attachment efficiencies than uncoated particles (open symbols) with few exceptions. Franchi and O'Melia showed that negatively charged sulfate functionalized PS particles coated with Suwannee River humic acid exhibit reduced retention (except at IS below 10 mM NaCl) in columns packed with glass beads (Figure 3a, star symbol). This was one of the first studies to report on the role of the secondary energy minimum in the reversible attachment of NPs or MPs.[152] In examining the effect of varying NOM concentrations, Amirbahman and Olson observed little difference in stability of negatively charged PS MPs when the concentration of peat humic acid increased from 1 to 10 ppm (Figure 3b, black and purple solid downward triangle).[149] Deshiikan et al looked at the effect of increasing Georgetown NOM concentration on two PS MPs. The stability of the positively charged PS MPs increased significantly as NOM concentration increased from 5 to 20 ppm. This was associated with a reversal of MP surface charge from positive to negative. However, no substantial difference in stability was observed for the negatively charged particles in the presence versus absence of NOM, a result attributed to less NOM having sorbed to the negatively-charged particles.[150] As humic substances are commonly negatively charged at environmentally relevant pH, adsorption of NOM onto positively charged MPs will reduce the magnitude of the surface charge.[48] The type of NOM sorbed to the surface also influences particle stability. In comparing adsorption of two NOMs having differing average molecular sizes (Georgetown fulvic and peat humic acid, both at 1 ppm) onto positively and negatively charged PS MPs, particles coated with the lower molecular-sized Georgetown fulvic acid were more likely to deposit onto quartz sand (Figure 3b, solid diamonds), despite similar electrophoretic mobilities.[149] This was attributed to the reduced steric-stabilizing effect of the smaller organic molecule.

A significant fraction of NPs and MPs is expected to enter groundwater via the unsaturated (vadose) zone, where particle mobility is often reduced compared to water saturated environments due to the role of the air-water interface and film straining.[132, 135, 148, 154] For example, Wan and Wilson attributed increased retention of PS MPs in sand-packed columns with increasing gas content to the air-water interface;[148] however, Torkzaban et al. suggest that for similarly charged surfaces, straining is the predominant retention mechanism.[132] Colloid hydrophobicity also plays an increased role in particle retention in the vadose zone, resulting in greater partitioning to the air-water interface.[148] Thus, the transport of hydrophobic plastics is expected to be significantly mitigated when moving through the vadose zone.

A review of the existing literature (Table 2) shows that few studies have examined the transport and deposition behavior of NPs and MPs in environmentally relevant systems. The subsurface environment is heterogeneous and complex, and laboratory studies using pristine granular media are likely to underestimate NP and MP deposition. As is also the case for plastic aggregation, most of the studies on plastic deposition were performed using spherical primary plastics that are less likely to be encountered in the environment. The transport behavior of more environmentally relevant secondary plastics comprised of fragments, films, rods, etc. requires investigation. Furthermore, studies have largely been limited to PS; thus, there is a need for additional research to understand the impacts of factors such as water chemistry, microbial biofilms, and soil type on the mobility of different plastics.

4. Plastics May Act as Vectors for Other Contaminants

4.1 Plastics as Contaminant Source and Sink

NPs and MPs can serve as both sources and sinks for contaminants in the environment (Figure 4a). Chemical byproducts, monomers, and additives (*e.g.*, bisphenol A, triclosan, bisphenone, flame retardants, phthalates, organotins) are added during the manufacturing of plastics. Several of these additives, that may leach from the plastic into the environment,[170] are of significant concern (endocrine disrupting, carcinogenic and/or mutagenic).[171] On the other hand, plastics can also sorb inorganic and organic contaminants,[20, 58, 59, 63, 172] as MPs in the aquatic environment were found to be contaminated with POPs and heavy metals. For example, MPs were reported to exhibit concentrations of POPs up to six orders of magnitude greater than the background concentration in the surrounding seawater.[62, 173-175] These interactions with contaminants are increasingly being studied to better understand the risks associated with plastics in the environment.[18, 57, 62, 64, 173, 176] Furthermore, the bioavailability of these sorbed contaminants to aquatic organisms may be considerable, with significant rates of desorption for DDT and phenanthrene observed across a range of salinities.[60]

Table S2 summarizes existing studies on the sorption of contaminants on plastics, many of which have focused on micrometer and millimeter-sized particles. Sorption of contaminants onto plastic debris depends on several factors such as the physicochemical properties of the polymer, solution chemistry of the immediate environment, the degree of weathering, and temperature.[18, 61, 62, 65, 174-177]

When plastics are released into the environment, several environmental factors will lead to fouling and weathering.[61, 177] Degradation and breakup of pristine plastics can increase the exposed surface area, resulting in increased sorption capacity. For example, Napper et al. investigated PE microbeads extracted from personal care products and found that rough MPs adsorbed more DDT and phenanthrene than smooth ones.[64] Similar trends were observed with heavy metal contaminants; aged PE pellets adsorbed more cationic metals than pristine pellets.[59] Brennecke et al. observed that higher levels of Zn^{2+} and Cu^{2+} sorbed onto aged PVC compared with pristine PS particles, despite the fact that PS generally shows a greater sorption capacity than PVC (Figure 4b).[57] On the other hand, weathering via photodegradation oxidizes plastics (adding *e.g.* carbonyl groups) and increases their polarity which can decrease their sorption capacity. Fouling, sometimes indicated by discoloration, is also expected to affect the adsorption of contaminants onto plastics. Discolored plastic particles have been shown to adsorb more PCBs than non-discolored ones.[61]

Increasing temperatures will generally reduce the "glassiness" of a polymer, increasing its affinity for contaminants; however, increasing the temperature of the aqueous phase also leads to increased solubility of organic and inorganic contaminants.[178] Crawford and Quinn observed greater affinities for PE MP with 33 different PAHs at 21 °C versus 10 °C.[18] Hu et al. also reported an increase in sorption with temperature of lubricating oil on PE NP and PS MP.[65] In contrast, Zhan et al. showed that sorption of PCBs onto PP decreased as temperature increased from 19 to 27 °C.[179] These conflicting data highlight the difficulty in predicting changes in contaminant sorption, because temperature will impact both the properties of the plastic and the contaminant.

The salinity of the surrounding aqueous environment can also influence the sorption behavior of plastics by affecting the water solubility of organic compounds. Generally, an increase in salinity decreases the solubility of non-polar and weakly polar organic contaminants in water,[178] known as the *salting-out effect*. Hence, high salt levels can increase the availability of certain hydrophobic contaminants for adsorption onto plastics. Indeed, the adsorption of phenanthrene on PP MPs increased with salinity.[180] Hu et al reported that lubricating oil adsorbed more to PE NPs and PS MPs when salinity increased. This was attributed to the salts promoting outer-sphere surface complexation between the particles and the oil.[65] Increased salinity levels also resulted in greater sorption capacities of PCBs onto PE and PS.[181] However, this trend does not appear to be universal for plastics. As salinity increased from freshwater to seawater, sorption of heavy metals by PE was observed to decrease considerably, with the exceptions of Cu^{2+} and Cr^{2+}.[182] Decreased sorption of DDT onto either PVC or PE was noted when salinity increased from river to seawater (no effect was noted on the sorption of phenanthrene).[60] Given the limited studies and contradictory reports, the effects of salinity on the sorption of inorganic and organic contaminants onto plastic merit further investigations.

Beyond environmental factors influencing sorption, the type of plastic also plays an important role. Figure 4b provides an integrated comparison of polymer sorption capacities to determine the expected relative contaminant association as a function of plastic type. For each study, the sorption capacity of different plastics was ranked by assigning a score of 1 to those that exhibited the highest sorption capacity and increasing values for plastics that exhibited lower sorption capacities. The average ranking for each type of plastic across the different studies was then calculated and reported alongside their glass transition temperatures (T_g). In general, rubbery polymers such as PE and PP are expected to allow greater diffusion of contaminants into the polymer than glassy polymers such as PET and PVC.[17] At room temperature, rubbery polymers exist above their T_g which results in greater flexibility and facilitates contaminant sorption.[17] Indeed, the rubbery polymer PE commonly shows a greater affinity for contaminants than other types of plastics (i.e., in Figure 4b, PE most often receives a score of 1).[4, 17, 20, 177, 183] Conversely, PET and PVC generally exhibit lower sorption capacities (i.e., they receive higher scores of 4 or 5).[20, 63] However, this generalization does not hold for all types of contaminants (Figure 4b). For instance, PS appears to be an exception to the rule, whereby its average sorption capacity ranking is greater than would be predicted by its T_g. Five studies have compared the sorption of contaminants by PS and other plastics, and twice it ranked highest despite PS being a glassy polymer at room temperature.[184] A possible explanation for this is the presence of benzene in the PS monomer rather than, for example, hydrogen in PE (Figure S1). This benzene ring increases the distance between the polymer chains and can facilitate contaminant attachment and integration into the polymer.[17] Other exceptions to the general trend include the accumulation of less metal by high density PE than low density PE, PET, PVC and PP,[58] and sorption of more alkylbenzenes by PVC than PE.[185] Thus, whereas general sorption trends appear to be well correlated to T_g, these latter observations are likely related to the chemistries of a specific contaminant and plastic.

Contaminants are unlikely to exist in isolation in the environment; however, studies investigating the potential for competitive sorption onto plastics are nearly nonexistent. Competitive sorption between phenanthrene and DDT was demonstrated for PE and PVC, which sorbed more DDT than phenanthrene.[64, 186] The observed trend for DDT could be due to several factors, including its greater hydrophobicity. There is a need to investigate the sorption capacity of plastics in environmentally relevant heterogeneous systems to further understand the mechanisms by which plastics preferentially interact with different organic and inorganic contaminants.

Figure 4a and Table S2 show that several investigations have focused on the association of plastic particles with persistent, bio-accumulative, and toxic compounds (*e.g.*, metals, PAHs, PCBs and DDT).[60, 61, 64, 173, 181] Pharmaceuticals and other endocrine-disrupting compounds which are contaminants of emerging concern are less studied in this context. Wu et al investigated the effect of salinity and presence of NOM on the sorption capacity of four pharmaceutical contaminants onto PE MPs.[187] They reported that sorption to PE MPs depends on contaminant hydrophobicity and that the presence of NOM decreased the affinity of all but one contaminant (carbamazepine).[187] Since the interaction of plastics with POPs can differ from pharmaceutical contaminants (some of which can be ionic), there is need for more research to understand the mechanisms by which pharmaceutical contaminants and NOM interact with plastic particles in aquatic environments.

4.2 NPs and MPs Can Facilitate the Transport of Contaminants

The mobility of organic and inorganic contaminants can be enhanced by association with colloids in soils, surface waters, and groundwaters.[52, 188, 189] Natural colloids (*e.g.* iron oxides and clays) have been reported to increase the transport of metals such as copper, zinc, lead, cadmium, arsenic and nickel up to 50 times the rates observed in non-colloid associated tests.[51, 53, 54, 190-194] Colloids have also demonstrated the potential for promoting the transport of organic pollutants, such as prochloraz, glyphosate, and atrazine.[195-197] The movement of colloids can be faster than that of the porewater due to the size exclusion effect, in which colloids are excluded from small pores.[198]

Studies on the facilitated transport of contaminants by plastics (PS and polyurethane, PU) in model or natural subsurface systems are sparse,[199, 200] though they are in general agreement with the literature on natural colloids. PU used within a remediation paradigm improved the removal of phenanthrene from soil by facilitating the contaminant's mobility in porous media and increasing the bioavailability to microbial populations that can degrade the contaminant.[199, 200] Jaradat et al. showed that phenanthrene in leaf compost had a greater affinity for sulfate- and carboxylate-modified PS MPs than the compost materials.[201] Laboratory-scale columns packed with leaf compost media revealed that phenanthrene levels in the column effluent were significantly higher in the presence of more hydrophobic sulfidated PS MPs than carboxylated PS. The potential for 76 and 301 nm sulfate-modified PS plastics to facilitate the transport of pyrene and phenanthrene has also been investigated in columns packed with glass beads or quartz sand.[145] At low IS, both pyrene and phenanthrene showed an earlier breakthrough in the presence of PS particles compared to that without particles. In contrast, at high IS, increased retention of PS particles in the granular medium resulted in increased retardation of contaminant compared to experiments with no particles. This suggests that the ability of plastics to facilitate the transport of contaminants is linked with the stability of the plastic itself. In their study with PS MPs, Roy and Dzombak identified slow desorption of contaminants from particles as an important prerequisite for significant colloid-facilitated transport.[53] Taken together, these studies show that MPs and NPs have the potential to facilitate the transport of contaminants but a great deal more research is needed to understand the scope of this problem.

5. Regulatory Policy

The increasing evidence of MP's potential for harm - either directly or indirectly, has led to numerous calls for regulations and bans on MP use in consumer products and release into the environment.[28, 34, 89, 202-205] In the U.S., most plastics are grandfathered into the Toxic Substances Control Act of 1977 and therefore are considered safe until proven otherwise.[206] MPs are considered non-hazardous solid waste from a regulatory standpoint, and governmental agencies have been hesitant to include MPs in water quality regulations such as turbidity or particulate matter, which would largely impact WWTPs.[89, 202, 206] In Austria, limits on plastic discharge into freshwater rivers and streams do exist, although the limit, at 30 mg L^{-1} day^{-1}, is so high as to be ineffective.[34] The European Marine Strategy Framework Directive, which requires member states to establish strategies for maintaining marine waters, includes MPs as marine litter but does not specify how countries should keep MPs from reaching their coastal waters (*e.g.*, improvements to WWTPs or MP bans).[204]

Amidst this backdrop, several governmental organizations have enacted legislation, primarily focused on MPs in single use cosmetics, to specifically ban microbeads or MPs. In 1999, the Canadian government classified microbeads as a toxin, which was coupled with the intent to prohibit importation, manufacture, or sale of some microbeads. Similar to other legislation however, this ban primarily covers personal care products and does not include abrasives, cleaning products, and other household uses. Nine US states have enacted legislation banning the use of microbeads, with Illinois the first to do so in 2014.[205] These acts primarily ban either the manufacture or sale of personal care products containing microbeads, though significant loopholes exist. For example, California law does not apply to products containing less than 1 ppm plastic by weight.[205] Furthermore, the US government passed the Microbead Free Waters Act (MFWA) in 2015, which amends the Food Drug and Cosmetic Act to ban the sale or distribution of MBs under a tiered timeline. The primary shortcoming of regulations that exist is that the scope is narrow and sufficient loopholes exist such that microbeads/MPs continue to be introduced into the environment.

The characterization of microbeads and MPs as non-biodegradable entities is commonly included in regulatory definitions. This suggests that any change in particle size – either incidental or engineered – would permit the incorporation of MPs in products despite the existing bans. Furthermore, penalties for circumventing plastic waste regulations either do not exist (Maryland and Maine)[205] or are not enforced.[203]

6. Environmental Implications and Outlook

We have presented estimated loads of plastics in different environmental compartments and an overview of the key factors that govern the degradation, aggregation, and transport of NPs and MPs in aquatic and terrestrial environments. The fate and transport of NPs and MPs strongly depend on the physicochemical properties of the plastics and water and soil chemistries. A significant concern regarding MPs and NPs is their demonstrated ability to act as transport vectors for environmental contaminants. The rubbery polymer PE has shown a higher sorption capacity compared to other plastics for most contaminants reviewed. PE is also the most produced and frequently detected plastic in the environment widely used in packaging. As such, regulatory bodies would do well to consider PE in policy making. Additionally, regulations should not only consider the ability of plastics to act as sinks for environmental contaminants, but also the contaminants that originate from the plastics. For example, while PVC generally accumulates lower amounts of contaminants from its surroundings than other plastics (Figure 4b), it is composed of a high content of carcinogenic phthalates (~50%).[207]

Despite the considerable body of plastics research, important questions remain unanswered:

- How do we define NPs and MPs? And how can we make this definition uniform within literature? There is a need to improve detection and characterization techniques, as there is currently no rigorous methodology to detect NPs in the environment. How can we develop new techniques or improve existing ones to push the resolution towards detection at the nanoscale?
- Does the transport of other commonly detected plastics (PE, PP, PVC, PET, etc.,) differ from that of PS in the subsurface environment? Do model primary plastics behave differently from environmentally relevant secondary plastics? Changes in the physicochemical properties of a particle will impact both aggregation and deposition behavior. Will plastic types of similar size/surface areas behave differently?
- Could plastic debris contamination in groundwaters be an important concern? What tools exist for the accurate detection of NPs/MPs in groundwaters?
- How do NPs and MPs interact with pharmaceuticals and other emerging contaminants? The effect of salinity on contaminant sorption remains unclear. How do complex aquatic environments containing natural organic matter, microorganisms, mixtures of contaminants, etc. affect sorption capacity? Does the formation of biofilms affect sorption/desorption capacities?

Answering these and other questions will significantly improve our understanding of the fate, transport, and risks associated with MPs and NPs that are already ubiquitous in the environment. Although policy makers are starting to acknowledge the potential risks and implications of MPs and NPs, which is leading to the ban of some products, these are important concerns, as MPs and NPs have been accumulating in the environment for decades. Understanding the behavior and prevalence of MPs and NPs in the environment is the first step towards mitigating the impacts of these contaminants.

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ASSOCIATED CONTENT

Supporting Information Available:

Molecular structures of commonly detected plastics (Figure S1); Data of plastic transport and concentrations in the environment (Table S1); Summary of contaminant sorption studies (Table S2).

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Figure Captions

Figure 1. Estimates of plastic loading and transport pathways in the environment aggregated from reports in the literature. Percentages indicate the fraction of plastics in a given compartment moving to a subsequent compartment, with wider arrows representing greater plastic transfer. Ranges of concentrations measured, either in number of particles per liter or per area, are given where reliable values were observed. Values indicated are for macro and microplastics. *corresponds to estimates for microplastics only. †corresponds to values divided between two compartments. ‡corresponds to best estimates in the absence of data in the literature. Data and references are summarized in Table S1.

Figure 2. Aggregation stability curves of selected PS NPs and MPs in **(A)** multivalent salts **(B)** monovalent salts without coating and **(C)** monovalent salts with coatings from studies summarized in Table 1.[116-119] Here, α_{pp} = particle-particle attachment efficiency and SMP, CMP and AMP = sulfate-, carboxyl- and amidine-modified plastics, respectively. Solid symbols indicate addition of polyelectrolyte to the background solution.

Figure 1. Deposition stability curves of PS NPs and MPs from studies in Table 2.[133, 149, 152, 153, 156, 159] **(A)** SMP with and without SRHA and **(B)** SMP, AMP, and CMP with and without various types of NOM Here, α_{pc} = particle-collector attachment efficiency and solid symbols indicate addition of NOM. PHA = peat humic acid, GFA = Georgetown fulvic acid, SRHA = Suwannee river humic acid and ChMP = chloromethyl-modified plastic.

Figure 4. (A) Contaminants that have been found to associate with plastic debris in the environment.[17, 57, 59, 62] Schematic adapted from [176] **(B)** Relative ranking of sorption capacity as a function of plastic type. In a given study, a score of 1 indicates the highest sorption capacity and increasing values indicate plastics that exhibit lower sorption capacities. HDPE = high-density polyethylene; LDPE = low-density polyethylene; POM = polyoxymethylene; PA = polyamide; PET = polyethylene terephthalate. *Glass transition temperature from [184].

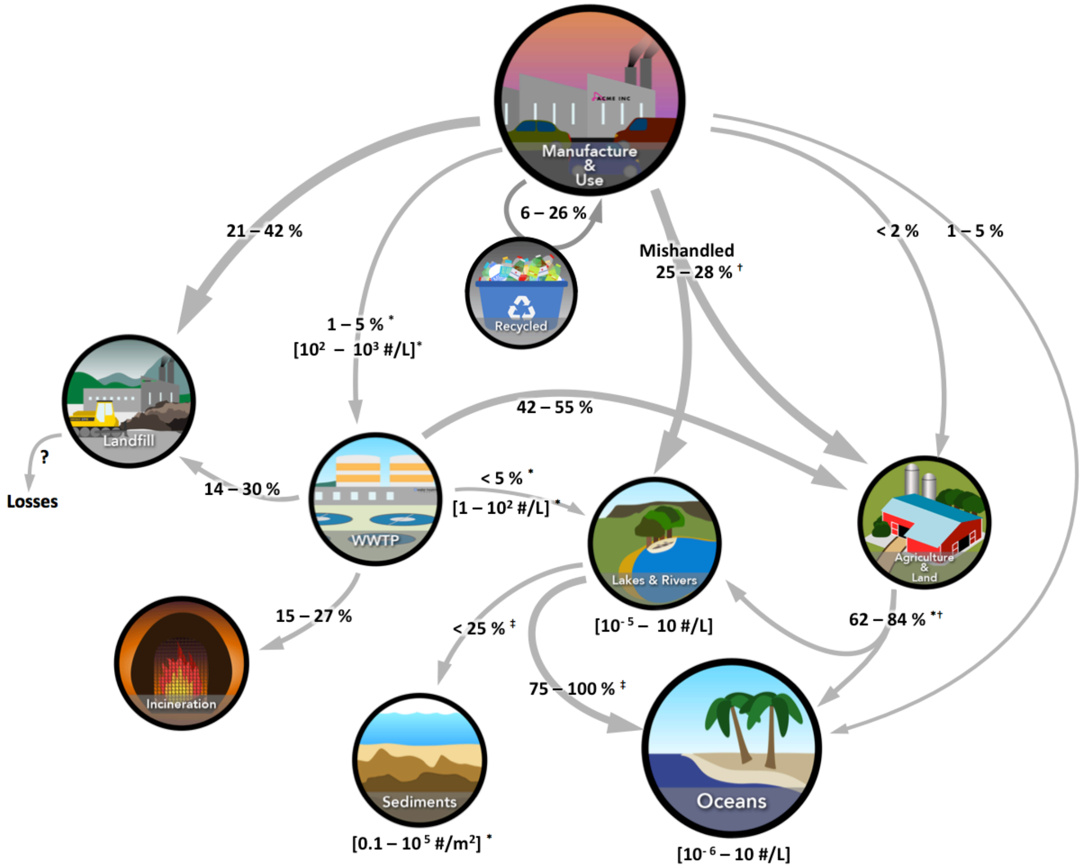
Manufacture
&
Use
6 – 26 %
Recycled
21 – 42 %
1 – 5 % *
[10^2 – 10^3 #/L]*
Mishandled
25 – 28 % †
< 2 %
1 – 5 %
Landfill
?
Losses
42 – 55 %
14 – 30 %
WWTP
< 5 % *
[1 – 10^2 #/L] *
Lakes & Rivers
Agriculture
&
Land
[10^{-5} – 10 #/L]
62 – 84 % *†
15 – 27 %
Incineration
< 25 % ‡
75 – 100 % ‡
Sediments
[0.1 – 10^5 #/m^2] *
Oceans
[10^{-6} – 10 #/L]

Figure 1.

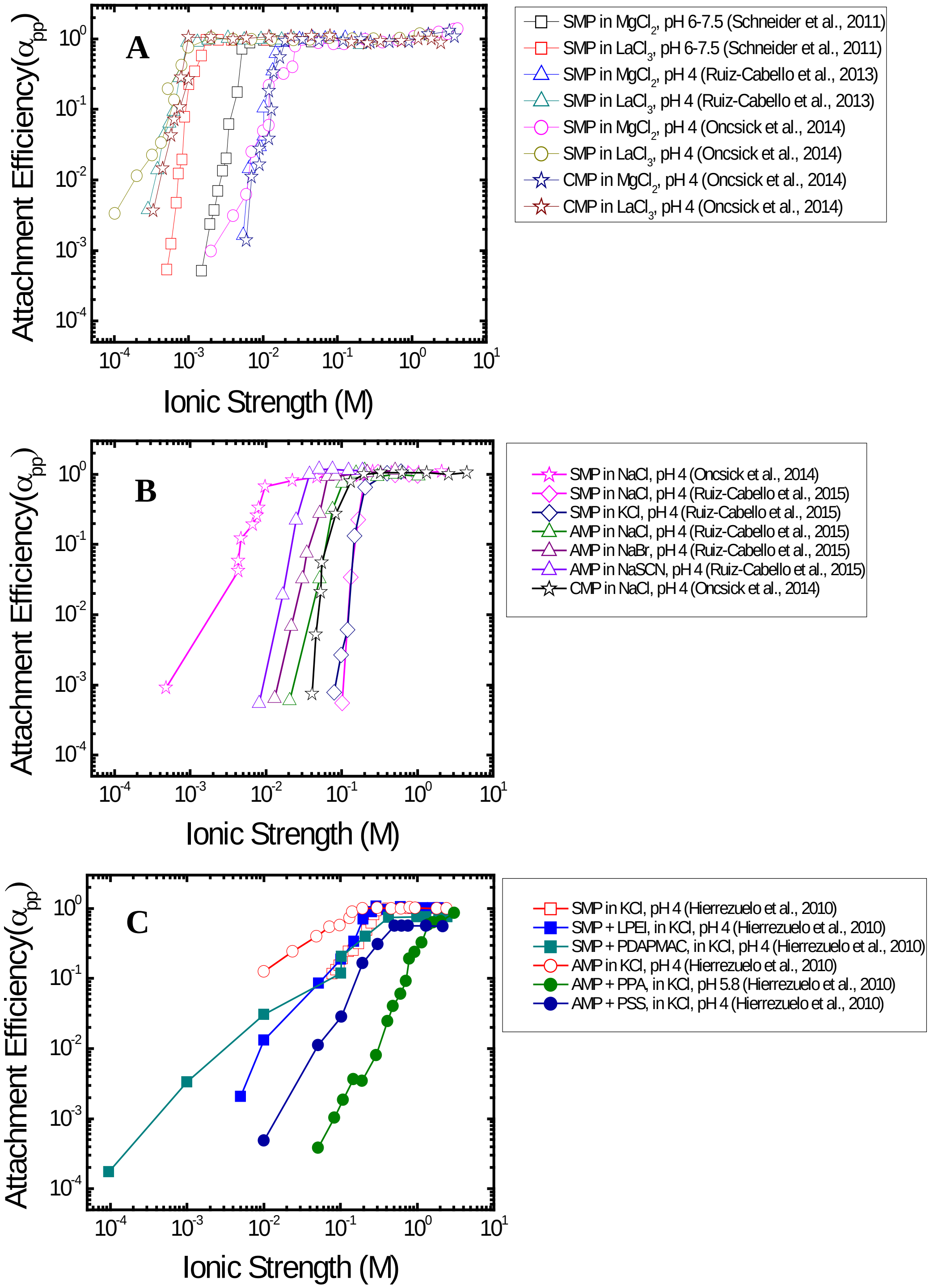
A
Attachment Efficiency(α_{pp})
10^{0}
10^{-1}
10^{-2}
10^{-3}
10^{-4}
Ionic Strength (M)
10^{-4}
10^{-3}
10^{-2}
10^{-1}
10^{0}
10^{1}
SMP in $MgCl_2$, pH 6-7.5 (Schneider et al., 2011)
SMP in $LaCl_3$, pH 6-7.5 (Schneider et al., 2011)
SMP in $MgCl_2$, pH 4 (Ruiz-Cabello et al., 2013)
SMP in $LaCl_3$, pH 4 (Ruiz-Cabello et al., 2013)
SMP in $MgCl_2$, pH 4 (Oncsick et al., 2014)
SMP in $LaCl_3$, pH 4 (Oncsick et al., 2014)
CMP in $MgCl_2$, pH 4 (Oncsick et al., 2014)
CMP in $LaCl_3$, pH 4 (Oncsick et al., 2014)
B
Attachment Efficiency(α_{pp})
Ionic Strength (M)
SMP in NaCl, pH 4 (Oncsick et al., 2014)
SMP in NaCl, pH 4 (Ruiz-Cabello et al., 2015)
SMP in KCl, pH 4 (Ruiz-Cabello et al., 2015)
AMP in NaCl, pH 4 (Ruiz-Cabello et al., 2015)
AMP in NaBr, pH 4 (Ruiz-Cabello et al., 2015)
AMP in NaSCN, pH 4 (Ruiz-Cabello et al., 2015)
CMP in NaCl, pH 4 (Oncsick et al., 2014)
C
Attachment Efficiency(α_{pp})
Ionic Strength (M)
SMP in KCl, pH 4 (Hierrezuelo et al., 2010)
SMP + LPEI, in KCl, pH 4 (Hierrezuelo et al., 2010)
SMP + PDAPMAC, in KCl, pH 4 (Hierrezuelo et al., 2010)
AMP in KCl, pH 4 (Hierrezuelo et al., 2010)
AMP + PPA, in KCl, pH 5.8 (Hierrezuelo et al., 2010)
AMP + PSS, in KCl, pH 4 (Hierrezuelo et al., 2010)

Figure 2

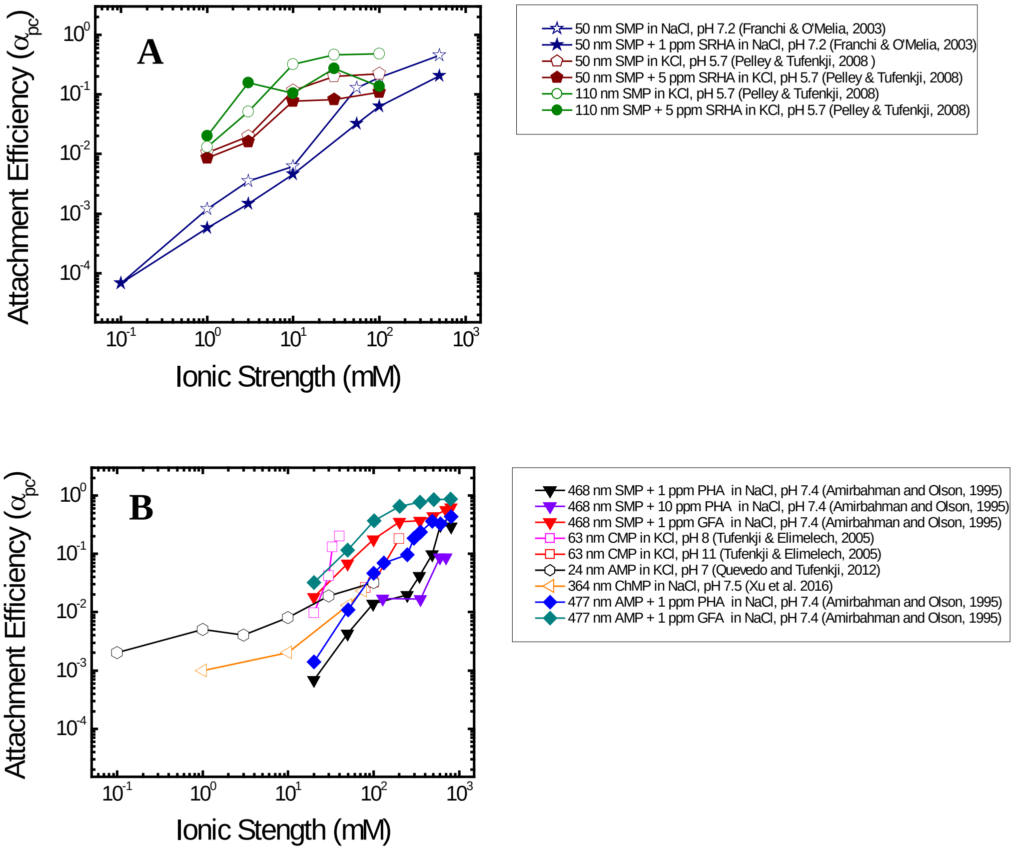
A
Attachment Efficiency (α_{pc})
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-1}
10^0
10^1
10^2
10^3
Ionic Strength (mM)
50 nm SMP in NaCl, pH 7.2 (Franchi & O'Melia, 2003)
50 nm SMP + 1 ppm SRHA in NaCl, pH 7.2 (Franchi & O'Melia, 2003)
50 nm SMP in KCl, pH 5.7 (Pelley & Tufenkji, 2008)
50 nm SMP + 5 ppm SRHA in KCl, pH 5.7 (Pelley & Tufenkji, 2008)
110 nm SMP in KCl, pH 5.7 (Pelley & Tufenkji, 2008)
110 nm SMP + 5 ppm SRHA in KCl, pH 5.7 (Pelley & Tufenkji, 2008)
B
Attachment Efficiency (α_{pc})
10^0
10^{-1}
10^{-2}
10^{-3}
10^{-4}
10^{-1}
10^0
10^1
10^2
10^3
Ionic Stength (mM)
468 nm SMP + 1 ppm PHA in NaCl, pH 7.4 (Amirbahman and Olson, 1995)
468 nm SMP + 10 ppm PHA in NaCl, pH 7.4 (Amirbahman and Olson, 1995)
468 nm SMP + 1 ppm GFA in NaCl, pH 7.4 (Amirbahman and Olson, 1995)
63 nm CMP in KCl, pH 8 (Tufenkji & Elimelech, 2005)
63 nm CMP in KCl, pH 11 (Tufenkji & Elimelech, 2005)
24 nm AMP in KCl, pH 7 (Quevedo and Tufenkji, 2012)
364 nm ChMP in NaCl, pH 7.5 (Xu et al. 2016)
477 nm AMP + 1 ppm PHA in NaCl, pH 7.4 (Amirbahman and Olson, 1995)
477 nm AMP + 1 ppm GFA in NaCl, pH 7.4 (Amirbahman and Olson, 1995)

Figure 2

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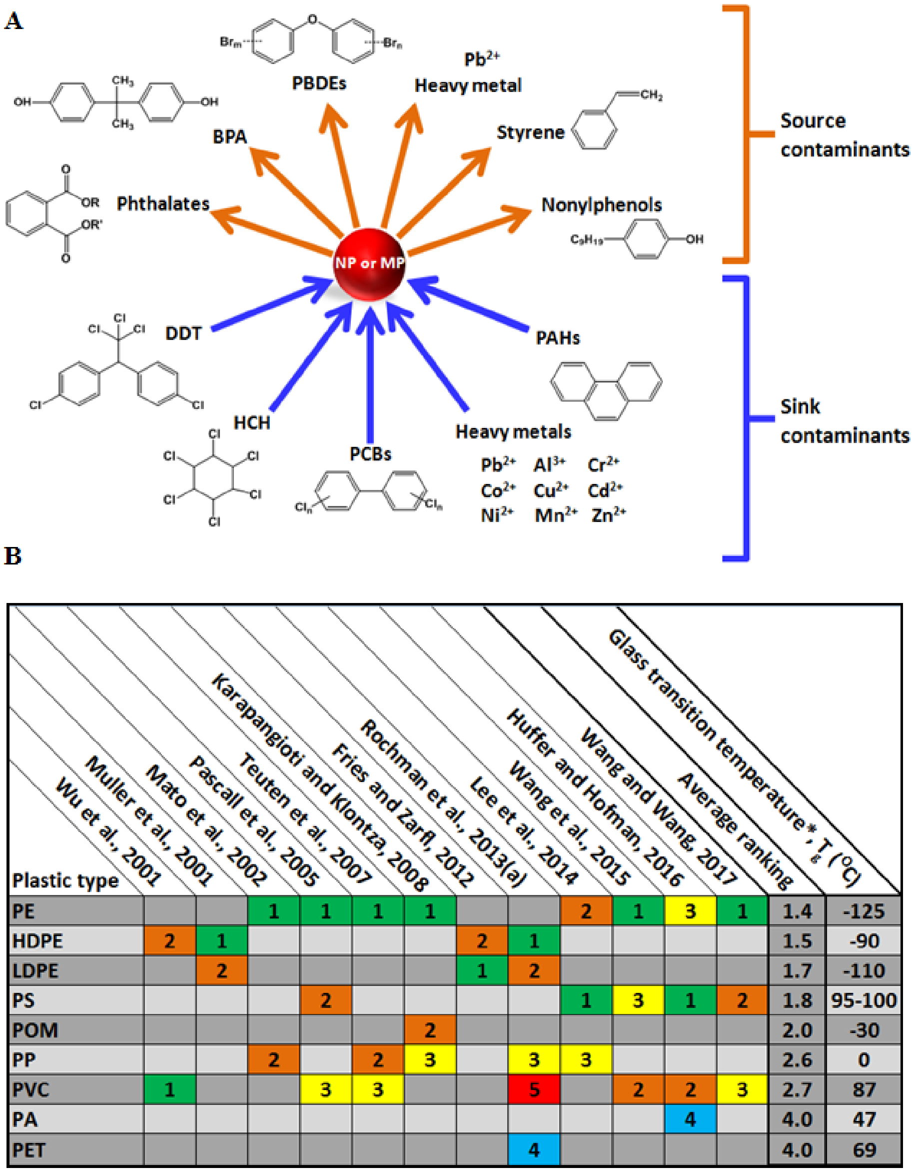


Plastic type	Wu et al., 2001	Muller et al., 2001	Mato et al., 2002	Pascall et al., 2005	Teuten et al., 2007	Karapangioti and Klontza, 2008	Fries and Zarfl, 2012	Rochman et al., 2013(a)	Lee et al., 2014	Wang et al., 2015	Huffer and Hofman, 2016	Wang and Wang, 2017	Average ranking	Glass transition temperature*, T_g (°C)
PE			1	1	1	1			2	1	3	1	1.4	-125
HDPE	2	1					2	1					1.5	-90
LDPE		2					1	2					1.7	-110
PS				2					1	3	1	2	1.8	95-100
POM						2							2.0	-30
PP			2		2	3		3	3				2.6	0
PVC	1			3	3			5		2	2	3	2.7	87
PA											4		4.0	47
PET								4					4.0	69

Figure 4

Table 1. Laboratory studies investigating NP and MP aggregation

Plastic type and surface functionality	**Nominal size and concentration**	**Solution chemistry**	**Experimental approach**	**Main findings and conclusion**	**References**
PVC latex	d= 480 nm C_0=36 – 1800 mg/L	Na^+ and Mg^{2+}	spectrophotometry	• Aggregation kinetics in agreement with DLVO theory • Aggregation follows Schulze–Hardy rule	Bibeau and Matijević, 1973
PVC latex	d= 338 , 510 nm $C_0 = 3 \times 10^8$ particles/mL	$NaNO_3$, $Ca(NO_3)$ and $La(NO)_3$	DLS	• Qualitative but not quantitative agreement with DLVO theory	Bleier and Matijević, 1976
PS latex (carboxylate)	d = 90 - 244 nm C_0 = 0.001- 0.005 wt %	NaCl,$BaCl_2$, $LaCl_3$	photoelectric colorimetery	• Aggregation follows Schulze–Hardy rule • Stability increases with addition of surface carboxyl groups	Sakota et al., 1977
PS latex (casein coating)	d = 348 ± 4 nm $C_0 = 2.1 \times 10^9$ particles/ mL	100 – 1000 mM NaCl pH 7.3	DLS	• Casein coating imparts stability • Stability increases with casein coverage	Dickinson et al., 1983
PS latex (sulfate)	d = 297 ± 3 nm $C_0 = 3 \times 10^{-4}$ (solid volume fraction)	KCl, $CaCl_2$ and $La(NO_3)_3$	spectrophotometry	• Good agreement between theory and experiment for all electrolytes studied • Aggregation follows Schulze–Hardy rule	Carrique et al., 1991
PS latex (sulfate)	d = 179 ± 12 nm $C = 10^{10}$ particles/mL	KBr , $MgSO_4$ pH 3 - 9	spectrophotometry	• Aggregation follows Schulze–Hardy rule	Bastos and de las Nieves, 1993
PS latex (sulfate)	d= 156 nm $C_0 = 7\times10^7$ particles/mL	15 – 100 mM NaCl 1 mg/L organic matter pH 7.4	DLS	• Coated NPs stable over 24 hours • NPs more stable with larger size fraction of humic acid • Steric interactions dominate stability	Amirbahman and Olson , 1993
PS latex	d = 789 ± 3 nm C_0 = 3.5% m/m solids	250 mM NaCl + polyelectrolytes (HEC, SCMC, SPSS)	DLS	• Polymer depletion led to NP aggregation • Stabilization by polyelectrolytes dependent on molecular size and osmotic pressure	Smith and Williams, 1995
PS latex (sulfate and amidine)	d = 110, 120 nm	500 mM NaCl 20 mg/L humic acid and polysaccharides pH 7	DLS	• Smaller humic acid fractions impart greater stability on smaller aggregates than polysaccharides. Impact is less significant for larger aggregates • Stability increases with adsorbed organic matter; positively charged particles adsorb more humic acid than the negatively charged counterpart • Stability of aggregates inversely proportional to aggregate size	Walker and Bob, 2001
Synthetic amphiphilic polyurethane (APU)	d = 17- 97 nm $C_0 = 1.5\times10^4$ mg/L	0.2, 5 mM $CaSO_4$ 0.02% NaN_3	DLS	• Stability decreased with increasing ionic strength for all except particles with poly(ethylene glycol)-modified urethane acrylate precursor chains	Tungittiplakorn et al., 2004

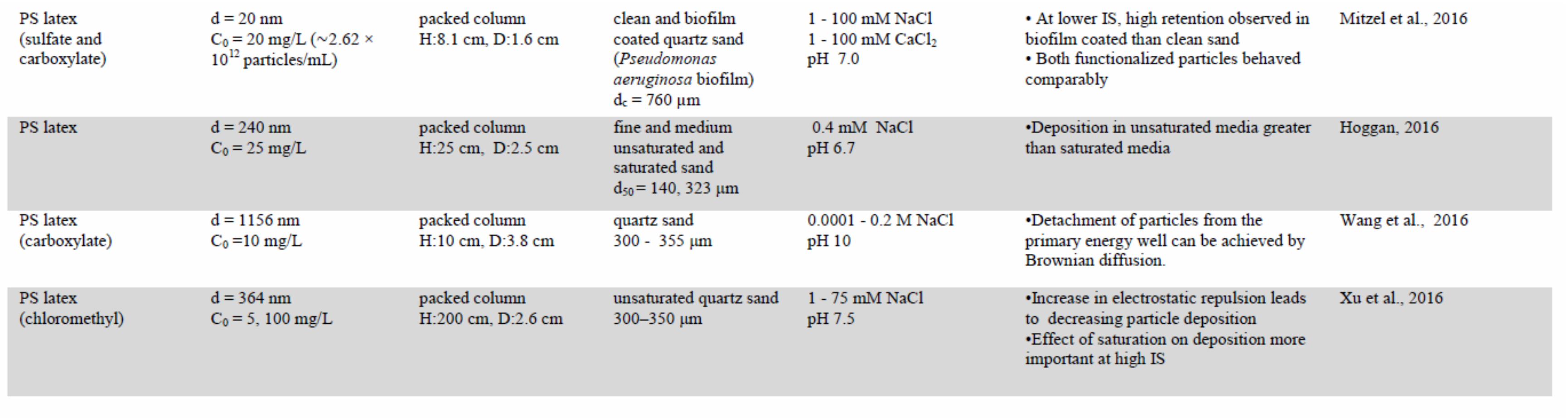
PS latex (carboxylate and amphoteric)	$d_{TEM} = 364 \pm 13$, 320 ± 15 nm C_0 = 110, 180 mg/L	NaCl pH 5,7,9	Low-angle light-scattering	• Surfactant-complexed particles more stable than bare particles • Electrostatic repulsion imparts stability • At low electrolyte concentrations, colloids remain stable after addition of surfactants	Jódar-Reyes et al., 2006
PS latex (sulfate and amidine)	d = 200, 270 nm C_0 = n. s.	KCl + polyelectrolytes pH 4, 5.8	DLS	•Both particles behave similarly in the presence of oppositely charged polyelectrolytes •In the presence of polyelectrolyte coatings, EDL forces responsible for stabilization	Hierrezuelo et al., 2010
PS latex (sulfate)	d = 115 ± 7 nm $C_0 = 1.3\times10^{8}$, 1×10^{10} particles/mL	0.1 – 1000 mM KCl, $MgCl_2$, $LaCl_3$ pH 6- 7.5	DLS	•Aggregation follows Schulze–Hardy rule •Multivalent counterion concentration determines counterion adsorption •DLVO theory assumption of constant charge is in agreement for monovalent salt.	Schneider et al., 2011
PS latex (carboxylate)	d = 30 nm C_0 = 100, 200, 300 mg/L	sea water	DLS	• NP destabilized shortly after being introduced to seawater (~1000 nm aggregates)	Wegner et al., 2012
PS latex (carboxylate)	d = 1000 nm C_0 = 80 mg/L	KCl, $MgCl_2$, $LaCl_3$, $ZrCl_4$ pH 4	DLS	• Aggregation follows Schulze–Hardy rule	Ruiz-Cabello et al., 2013
PS latex (carboxylate and amidine)	d = 40, 50 nm $C_0 = 1\times10^{4}$ mg/ L	natural seawater	DLS	• Carboxylate modified NP formed large aggregates of ~1700 nm while amine modified ones was dispersed at ~90 nm	Della Torre et al., 2014
PS latex (carboxylate and sulfate)	d = 530, 1000 nm C_0 = 4.5 mg/L	5 – 1000 mM NaCl, KCl, CsCl, $MgCl_2$, $CaCl_2$, $LaCl_3$ pH 4	DLS	• Aggregation follows Schulze–Hardy rule • Aggregation highly dependent on valence of counter ion • Aggregation insensitive to ion type for the same valence	Oncsik et al., 2014
PS latex (sulfate and amidine)	d_{TEM} = 960, 980 nm C_0 = 50–200 mg/L	10 - 200 mM NaCl 8 - 100 mM NaSCN 9 - 180 mM NaBr 10 - 500 mM KCl 10 - 500 mM CsCl pH 4	DLS	• Both MPs behave similarly in all electrolytes	Ruiz-Cabello et al., 2015
PS latex	d = 70, 1050 nm C_0 = 50 mg/L	Natural fresh water	DLS	• Heteroaggregation observed with kaolin or bentonite clays in natural freshwater	Besseling et al., 2016
PS latex (carboxylate)	d = 24 - 495 nm $C_0 = 1.5\times10^{8}$ - 1×10^{12} particles/mL	500 mM NaCl 2000 mM $CaCl_2$ pH 7	DLS	• Aggregation rate directly proportional to particle concentration	Henry et al., 2016
PS latex (sulfate and amidine)	d_{TEM} = 110, 265 nm C_0 = 2–10 and 50–200 mg/L; 0.3 – 2×10^{9} particles/mL	NaCl, NaBr, NaSCN, $NaN(CN)_2$ pH 4	DLS TEM	• Surface charge and aggregation rate both sensitive to the type of ion • Aggregation follows Schulze–Hardy rule	Oncsik et al., 2016

Table 2. Laboratory studies investigating NP and MP deposition

Plastic type and surface functionality	Particle size and concentration	Deposition system	Collector surfaces	Solution chemistry	Main findings and conclusions	References
PS latex (sulfate)	d = 46,378,753 nm C_0 = 1 - 4 mg/L	packed column H:15, 20 cm, D:n.s.	glass beads d= 200,400 µm	3-300 mM KCl, $CaCl_2$ pH 6.7	• Deposition rate increases with increasing IS until 0.1M KCl and 0.01M $CaCl_2$ (above which deposition rate decreases)	Elimelech & O'Melia, 1990a
PS latex (sulfate)	d= 46, 121,378,753 nm C_0 = 0.5 - 4 mg/L	packed column H: 20 cm, D:n.s.	glass beads d= 200,400 µm	3-300 mM KCl pH 6.7	• Deposition rate increases with IS • Stability curve slope independent of particle size	Elimelech & O'Melia, 1990b
PS latex (sulfate)	d = 156 nm C_0= 7×10^7 particles/mL	packed column H:n.s, D:n.s	quartz sand d= 275 µm	15 – 100 mM NaCl 1 mg/L humic matter pH = 7.4	•Steric repulsion responsible for stability at high ionic strengths •Peat humic acid imparts more stability than Georgetown fulvic acid	Amirbahman and Olson, 1993
PS latex (sulfate and carboxylate)	d = 190, 220 nm C_0 = 5 × 10^7 particles/mL	packed column H:30 cm, D:2.5 cm	unsaturated quartz sand d_{50} =212 - 315 µm	1 mM $NaNO_3$ pH 7.0	• Presence of air at collector surface increases retention • Retention greater for hydrophobic particles	Wan and Wilson, 1994
PS latex (sulfate and amidine)	d = 468, 477 nm C_0 = 10^7 particles/mL	packed column H:n/a	quartz sand d_{50} =275 µm	20 - 800 mM NaCl pH 7.4 1, 10 mg/L Georgetown fulvic acid, peat humic acid	• Negatively charged particles more stable than positively charged when coated due to greater magnitude of repulsive force • Larger molecular sized NOM shows less retention due to steric contribution	Amirbahman and Olson, 1995
PS latex	d = 76, 301 nm C_0= n/a	packed column H:15 cm, D:3.2 mm	glass beads 100, 200 µm quartz sand 74, 149 µm	0 - 1 mM KCl	• At higher IS, particles deposited on porous media	Sojitra, 1995
PS latex (sulfate and amidine)	d = 2500 nm C_0 = 2.3 × 10^9 particles/mL	packed column H:10 cm, D:2.5 cm	Ottawa sand d_{50} =580 µm	10 mM NaCl 0 - 20 mg/L Georgetown NOM, pH 5.4	• Presence of NOM imparts stability due to contribution of steric and electrostatic repulsion	Deshiikan et al., 1998
PS latex	d = 53 – 1960 nm C_0= n.s.	packed column H:20 cm, D:10 cm	Munich gravel d= 250 µm Sengethal sand d= 100 µm	Milli- Q water 1, 10 mM NaCl, $CaCl_2$ pH = n/a	•Deposition in agreement with DVLO: particle transport increases with decreasing IS •Impact of counterion valence more apparent in sand gravel	Huber et al., 2000
PS latex (fluorescent) (carboxylate)	d = 450, 1000, 2000, 3200 nm C_0= n.s.	packed column H:15 cm, D: 4.8 cm H:10 cm, D: 5 cm	Ottawa sand d_{50} = 150 – 710 µm glass beads d_{50} = 260 µm	1 mM NaCl pH 7	• Model included; particle and grain size influence retention due to straining • Retention increases with decreasing grain size and increasing colloid size	Bradford et al., 2002
PS latex (sulfate)	d = 98 nm C_0= 1 mg/L	packed column H: 250 cm ,D: 2.5 cm	glass beads d= 200 µm	0-500 mM NaCl 0,1 mg/L SRHA 13-960 mM KCl, $CaCl_2$ pH 6.7, 7.2	• Deposition increases with IS; particle depositing at lower IS more prone to reentainment • Deposition decreases with SRHA (except at low IS)	Franchi and O'Melia, 2003

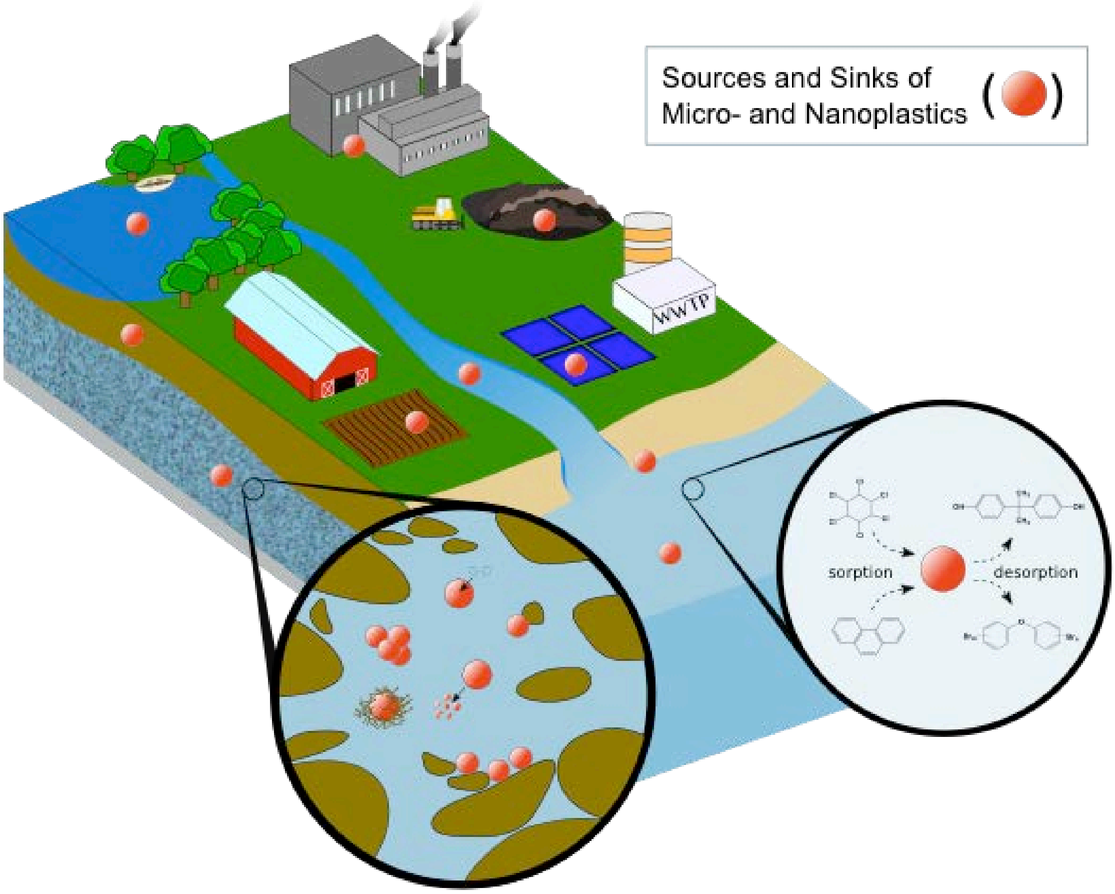
PS latex (sulfate)	d = 308 nm C_0 = 1 mg/L	packed column H:20 cm, D:2.5 cm	glass beads d= 200, 400 µm	10 - 100 mM KCl 200 mM $CaCl_2$ pH 5, 6.7-6.8	•Latex particle exhibit little affinity for glass beads	Hahn et al., 2004
PS latex (carboxylate)	d = 63, 320, 3000 nm $C_0 = 1\times 10^7$ - 3.6×10^9 particles/mL	packed column H: 12.6 cm, D:1.6 cm	glass beads d_{50} = 330µm	3 - 300 mM KCl 0.06 mM SDS pH 8, 11	• Retention increases with IS • Deviations from colloid filtration theory investigated	Tufenkji and Elimelech, 2005
PS latex (sulfate)	d =20 - 420 nm C_0 = 100 mg/L	packed column H:10 cm, D:4.5 cm	quartz sand d= 300 - 355 µm	1 mM (NaCl + $NaHCO_3$) pH 7.5	• Particle retention lower in saturated flow conditions • Deposition decreases reaching a minimum then increases as plastic size increases to MP	Zhuang et al., 2005
PS latex (fluorescent) (carboxylate)	d = 1000, 3200 nm $C_0 = 0.25, 0.5, 1, 2 \times C_T$ $C_T = 3.68\times 10^7$, 1.18×10^6 particles/mL	packed column H:15 cm, D: 4.8 cm H:10 cm, D: 5 cm	Ottawa quartz sand d= 150, 240, 360 µm	1 mM NaCl pH 7	•Retention increase with increasing MPs size and decreasing sand size •Less deposition with increase in C_0	Bradford and Bettahar, 2006
PS latex (carboxylate and amidine)	d = 100 - 2000 nm $C_0 = 1\times 10^6$, 1×10^7 particles/mL	packed column H:20 cm, D:3.8 cm	glass beads and quartz sand 417 – 600 µm	1 – 50 mM NaCl pH 2, 6.7	•Particle retention increases with increasing velocity	Tong and Johnson, 2006
PS latex (sulfate)	d = 30, 66, 1156, 3000 nm C_0 = 10, 40 mg/L	packed column H:10 cm, D:3.8 cm	glass beads (3 fraction sizes) d= 88 - 125 µm d=180 - 250 µm d= 590 - 840 µm	DI water 200 mM NaCl pH 10	•Using DI water, 0% particle deposition observed except for 3 µm PS •High IS imparts a lower critical straining ratio	Shen et al., 2008
PS latex (carboxylate)	d = 1100 nm $C_0 = 2.5$ - 2.8×10^7 particles/mL	packed column H:10 cm, D:5 cm	unsaturated Ottawa sand d_{50} =240, 360 µm	6, 30 and 60 mM KCl pH 10	• Retention decrease with level of saturation	Torkzaban et al., 2008
PS latex (sulfate)	d = 2000 nm C_0 = 7.5 mg/L	packed column H:6.7 cm, D:1.8 cm	Zirconia beads d_{50} = 326 µm	0.1 - 100 mM KCl pH 3 - 11	• Retention increases with IS • pH effects on transport not significant	Kobayashi et al., 2008
PS latex (sulfate)	d = 50, 110, 1500 nm $C_0 = 1.2\times 10^{11}$, 1.2×10^{10}, 8.6×10^6 particles/mL	packed column H: 15 - 16.5 cm, D:1cm	quartz sand d_{50} = 256 µm	1 – 100 mM KCl 0, 5 mg/L SRHA pH 5.7	• Deposition increases with IS • Addition of SRHA generally decreases deposition due to steric contribution	Pelley and Tufenkji, 2008
PS latex (fluorescent) (carboxylate)	d = 20, 200, 1000 nm C_0 = n.s.	packed column H:20 cm, D:5.4 cm	Dune sand d_{50}= 310 - 320 µm	3-4 mM artificial rainwater pH 7.8	• Particle transport: acid washed sand > distilled water washed > natural sand	Shani et al., 2008

PS latex (fluorescent)	d = 980 nm C_0= n/a	packed column H:30 cm, D:2.05 cm	unsaturated and saturated sand d_{50} = 250 µm	2 - 50 mM $NaNO_3$ pH 7	•Saturated flow: decreasing solution surface tension and IS, decreases deposition •Unsaturated flow: deposition decreases exponentially with travel distance.	Zhuang et al., 2010
PS latex (carboxylate)	d = 40, 500 nm C_0= 7.8 × 10^{11} / 1.2 × 10^{8} particles/mL	packed column H:30 cm, D:1cm	saturated quartz sand d = 270µm	0.05 – 100 mM NaCl pH 6.7, 9.6	•Deposition is IS dependent, and increases with increasing IS	Qing, 2011
PS latex (fluorescent) (carboxylate)	d = 1000 nm C_0 = 3.64 × 10^{7} particles/mL	packed column H:7 cm, D:1.4 cm H:30 cm, D:5.5 cm	natural quartz sand d = 800 µm	Dead sea water 8.5 M	•Particle deposition accelerates at high IS	Magal et al., 2011
PS latex (sulfate and carboxylate)	d = 20, 1000 nm C_0= 8 × 10^{11}, 10^{7} particles/mL	packed column H:8cm, D:1.6 cm	clean and biofilm coated quartz sand (*Pseudomonas aeruginosa* biofilm) d_{50} = 763 µm	10 mM KCl pH 7.2	• Retention increased in biofilm coated sand vs. clean sand • Higher retention with sulfate functionalized particles than carboxylate	Tripathi et al., 2011
PS latex (sulfonated)	d = 800 nm C_0= n.s.	n.s.	hematite covered mica monolayer	IS = 0.1- 10 mM pH 3.5	• Deposition deviates from the mean-field DLVO theory but agrees with DLVO theory at low IS	Nattich-Rak et al., 2012
PS latex (carboxylate)	d= 24 nm C_0 = 10^{12} particles/mL	packed column H :10 cm, D:1cm	loamy and quartz sand d = 225 µm, loamy d = 256 µm, quartz	0.1 -100 mM KCl 0.1 -10 mM $CaCl_2$ pH 7	• Retention increases with IS • Higher deposition with divalent electrolyte vs. monovalent in quartz sand • Retention greater in loamy than quartz sand	Quevedo and Tufenkji, 2012
PS latex (fluorescent) (carboxylate)	d = 100, 1050 nm C_0= 10.5 mg/L	chamber bed L×W×H: 20×10×10 cm	glass rod d = 5000 µm	1- 100 mM KCl pH 7	• Retention decreases with increasing flow velocity and increases with increasing IS	Wu et al., 2012
PS latex	d = 60 nm C_0 = 50 mg/L	deep well plate columns H:3.4 cm	Iota quartz sand and Calls Creek sediment d = 200, 361 µm	0.001–0.1% sodium dodecyl sulfate (SDS)	•Greater deposition in the sediment than in Iota quartz sand.	Bouchard et al., 2013
PS latex (fluorescent) (carboxylate)	d = 75, 300 and 2100 nm C_0 = 4.31 × 10^{10} and 1.35 × 10^{8} particles/mL	packed column H:15.2 cm, D:2.61 cm	fine and medium quartz sand d_{50} = 181, 513 µm	0.1 - 1000 mM NaCl 83-95% saturation	• Less attachment onto fine than medium quartz sand • Deposition slighter higher with large particles	Mitropoulou et al., 2013
PS latex (carboxylate)	d = 100, 500, 2000 nm C_0 = 1.1 × 10^{6} , 7.3 × 10^{7}, 2.3 × 10^{10} particles/mL	packed column H:11 cm, D:2 cm	quartz sand d_{50} = 250 µm	0 - 800 mM NaCl, pH 5.6 - 5.8	• Deposition behavior in column experiments not consistent with batch experiments	Treumann et al., 2014
PS latex (carboxylate)	d= 300 nm C_0 = 5.8 × 10^{7} particles/mL	physical micromodel	polydimethylsiloxane (PDMS)	DI water IS = 0.0012 mM pH 7.0	• Retention decreases with flow velocity	Zhang et al., 2015



PS latex (sulfate and carboxylate)	d = 20 nm C_0 = 20 mg/L (~2.62 × 10^{12} particles/mL)	packed column H:8.1 cm, D:1.6 cm	clean and biofilm coated quartz sand (*Pseudomonas aeruginosa* biofilm) d_c = 760 µm	1 - 100 mM NaCl 1 - 100 mM $CaCl_2$ pH 7.0	• At lower IS, high retention observed in biofilm coated than clean sand • Both functionalized particles behaved comparably	Mitzel et al., 2016
PS latex	d = 240 nm C_0 = 25 mg/L	packed column H:25 cm, D:2.5 cm	fine and medium unsaturated and saturated sand d_{50} = 140, 323 µm	0.4 mM NaCl pH 6.7	•Deposition in unsaturated media greater than saturated media	Hoggan, 2016
PS latex (carboxylate)	d = 1156 nm C_0 =10 mg/L	packed column H:10 cm, D:3.8 cm	quartz sand 300 - 355 µm	0.0001 - 0.2 M NaCl pH 10	•Detachment of particles from the primary energy well can be achieved by Brownian diffusion.	Wang et al., 2016
PS latex (chloromethyl)	d = 364 nm C_0 = 5, 100 mg/L	packed column H:200 cm, D:2.6 cm	unsaturated quartz sand 300–350 µm	1 - 75 mM NaCl pH 7.5	•Increase in electrostatic repulsion leads to decreasing particle deposition •Effect of saturation on deposition more important at high IS	Xu et al., 2016

TOC Graphic


Sources and Sinks of
Micro- and Nanoplastics (●)
WWTP
sorption
desorption