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Deposition of engineered nanoparticles (ENPs) on surfaces in aquatic systems: a review of interaction forces, experimental approaches, and influencing factors

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Abstract

The growing development of nanotechnology has promoted the wide application of engineered nanomaterials, raising immense concern over the toxicological impacts of nanoparticles on the ecological environment during their transport processes. Nanoparticles in aquatic systems may undergo deposition onto environmental surfaces, which affects the corresponding interactions of engineered nanoparticles (ENPs) with other contaminants and their environmental fate to a certain extent. In this review, the most common ENPs, i.e., carbonaceous, metallic, and nonmetallic nanoparticles, and their potential ecotoxicological impacts on the environment are summarized. Colloidal interactions, including Derjaguin-Landau-Verwey-Overbeek (DLVO) and non-DLVO forces, involved in governing the depositional behavior of these nanoparticles in aquatic systems are outlined in this work. Moreover, laboratory approaches for examining the deposition of ENPs on collector surfaces, such as the packed-bed column and quartz crystal microbalance (QCM) method, and the limitations of their applications are outlined. In addition, the deposition kinetics of nanoparticles on different types of surfaces are critically discussed as well, with emphasis on other influencing factors, including particle-specific properties, particle aggregation, ionic strength, pH, and natural organic matter. Finally, the future outlook and challenges of estimating the environmental transport of ENPs are presented. This review will be helpful for better understanding the effects and transport fate of ENPs in aquatic systems.

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Introduction

Nanotechnology has gradually emerged as one of the most promising technologies. A project on emerging nanotechnology has classified nanoparticles into four categories (i.e., engineered, incidental, natural, and generic). Notably, engineered nanoparticles (ENPs), which possess unique physicochemical properties, are extensively utilized for applications to achieve social and economic benefit (Hardman 2006; Mihranyan et al. 2012; Nel 2006). Scientists and economists have predicted a tremendous increase in the development of ENP-based processes and technologies (Hendren et al. 2011; Klaine et al. 2008; Wiesner and Bottero 2007). The production of these nanoparticles is expected to increase by over half a million tons by 2020 to fulfill the surging demand for varied situations in diverse areas (NRC 2012; Stensberg et al. 2011). Therefore, the rapid increase in ENPs has brought about wider consumer applications, such as emerging sensing

applications (He et al. 2010; Robinson et al. 2008), biomedicine (Alivisatos 2003; Bianco and Prato 2003; Elghanian 1997), catalysts (Fathinia et al. 2010; Tang et al. 2009), etc. Additionally, ENPs have also been actively employed for the removal of various contaminants from aqueous systems (Fathinia et al. 2010; Liu and Zhao 2007; Martinson and Reddy 2009; Sheng et al. 2010). For example, cupric oxide nanoparticles reportedly possess a strong elimination ability for both pentavalent and trivalent forms of arsenic (Martinson and Reddy 2009). The good adsorption performance of copper(II) on iron phosphate nanoparticles and multiwalled carbon nanotubes has also been demonstrated by several researchers (Liu and Zhao 2007; Sheng et al. 2010). The degradation of both anionic and cationic dyes can be achieved with TiO_2 nanoparticles through a comparative photocatalytic process under UV light irradiation (Fathinia et al. 2010). However, due to the growing production and exclusive application of these nanoparticles, the unintentional migration and entrance of ENPs into aquatic systems has raised concerns about their toxicological and adverse impacts on ecosystems and human health (Bundschuh et al. 2018). A clear understanding of the interaction mechanisms of these nanoparticles with surrounding organisms and related contaminants is therefore crucial to better assess the impacts that these nanoparticles could have on the environment.

Once released into the aqueous environment, particles interact with the surrounding environments and may undergo deposition and release processes. In particular, the depositional behavior of nanoparticles on naturally occurring interfaces is considered to play a critical role in determining the transport fates of exogenous nanomaterials in the environment and the subsequent potential for their release and impacts on ecological systems (Petosa et al. 2010; Baun et al. 2008). Thus, it is imperative to elucidate the associated environmental exposure risks and have a full understanding of the transport and retention mechanisms of nanomaterials in aquatic environments (Wiesner et al. 2009). The colloidal theories central to describe the potential for nanoparticle attachment to typical interfaces and the stability of colloidal suspensions include Derjaguin-Landau-Verwey-Overbeek (DLVO) forces and non-DLVO interactions. The electrostatic properties of the surface are considered to be the most important surface characteristics governing nanoparticle depositional behavior. The overlap of the electric double layer around charged nanoparticles and the charged interface will lead to electrostatic double layer interactions. Nanoparticles possessing opposite zeta (ζ) potentials than the surfaces are considered to be prone to deposition, whereas nanoparticles exposed to surfaces with the same charges are less likely to be deposited (Li et al. 2014). Under conditions resulting in repulsive interactions, the attachment onto a surface is considered to be dominated by repulsive electrostatic interactions, thus inhibiting deposition, whereas the attractive electrostatic interactions are the major forces in electrostatic conditions corresponding to favorable deposition.

To date, studies have indicated that the entry of ENPs into aqueous environments and the subsequent potential release into and impacts on ecological systems is highly dependent on the surface composition (Lin et al. 2011; Liu et al. 2012), on the physicochemical properties of the particles (i.e., particle size, particle shape, surface charge, and coating) (Nel 2006; Reidy et al. 2013), and on the solution chemistry (i.e., ionic strength (IS), pH, and natural organic matter (NOM) content) (Benn and Westerhoff 2008; Petosa et al. 2010; Wiesner and Bottero 2007). The impacts of specific physicochemical characteristics of particles at the nanoscale may substantially influence their tendency for colloidal interaction. Thus, it is essential to recognize the influencing mechanism between particles and interfaces to predict their transport fate and potential risks in the environment and thus the likelihood of exposure.

This review intends to present a systematic overview of the depositional behavior of ENPs in aquatic systems. The most common ENPs (i.e., carbon-based nanoparticles, metal or metal oxide nanomaterials, and quantum dots) and their related applications and potential risks are summarized. The colloidal forces determining nanoparticle deposition, including traditional DLVO and non-DLVO theory, are outlined. Furthermore, a brief discussion on the most commonly used experimental methods for examining the deposition of ENPs on collector surfaces, such as the packed-bed column method and quartz crystal microbalance (QCM), and the limitations of their applications are given below. Additionally, the factors including collector surface type, particle-specific properties, and solution chemistries impacting the depositional behavior of nanoparticles transported through aquatic systems are also discussed.

Engineered nanoparticles in the aquatic environment

ENPs explicitly refer to engineered materials with a structure size at the nanometer scale (1–100 nm) and serve as building blocks for nanotechnology. The increasing utilization of engineered nanomaterials exploited in domains such as biomedicine (Bianco and Prato 2003), electronics (Compton and Nguyen 2010; King 1999), cosmetics, and energy (Brownson et al. 2011; Peralta-Videa et al. 2011) has significantly promoted the development of nanotech products. Generally, ENPs can be classified into three groups: carbon-based nanoparticles (particularly fullerenes (C_{60}), carbon nanotubes (CNTs), graphene oxides (GOs), and carbon dots (CDs)), metal or metal oxide nanomaterials (e.g., Ag, Cu, Au, zero-valent iron (ZVI), TiO_2, CeO_2, ZnO, Fe_3O_4, Al_2O_3), and some inorganic nanomaterials, such as quantum dots (QDs) and SiO_2. The existence of carbon-based nanomaterials and their functional derivatives in natural or artificial aquatic systems is not

novel today. Nanoparticles, including CNTs and C_{60} nanoparticles, have been employed as nanosorbents of specific contaminants, such as trihalomethanes (Lu et al. 2005), polycyclic aromatic hydrocarbons (Cheng et al. 2004), and naphthalene (Yang et al. 2006). Metal nanoparticles, such as Ag and Cu, which are capable of bactericidal effects, are exploited in some personal care products, textiles, and specific medical use, while certain metal oxide particles are used in sunscreens, cosmetics, paints, and coatings (Lee et al. 2008; OECD 2014; Ravishankar and Jamuna Bai 2011; Ruparelia et al. 2008). The fluorescent properties of QDs make them widely applied in biomedical imaging (Alivisatos 2003), solar cell technology (Nozik 2002), and information sensors (Posani et al. 2006). Silica nanoparticles can be utilized in pharmaceutical products (Motaung and Luyt 2010) and adhesives (Ge et al. 2008), and they are considered to exhibit similar physicochemical behavior as metal oxides such as TiO_2, Al_2O_3, and Fe_2O_3 under specific conditions (Mu et al. 2011; Sahai 2002). Holding great promise in various fields, once nanosized particles are present in aquatic environments, their surface characteristics can be readily influenced by suspended NOM and synthetic contaminants through the surface reactivity properties of the nanoparticles, such as redox and adsorption/desorption, hence impacting particle stability and biotoxicity in natural aquatic environments.

Carbon-based nanoparticles

As one type of the increasing number of engineered nanomaterials, carbonaceous nanoparticles and their functional derivatives hold the most potential for employment in various fields. However, because of extensive manufacturing, carbon-based nanoparticles have emerged as new environmental pollutants that could have a potentially significant impact on aquatic systems and human health once released into the water or soil environment. The exposure routes of nanoparticles include manufacturing, transportation, consumption, and disposal (Lin et al. 2010). Several studies have provided detailed descriptions of fullerene toxicity tested on bacteria and human cells (Fortner et al. 2005; Handy et al. 2008; Indeglia et al. 2018; Klaine et al. 2008; Sayes et al. 2004). The results reveal that the cytotoxicity of fullerene is certainly up to the property of functional groups (Sayes et al. 2004) and may decrease glutathione (Zhang et al. 2009) and cell viability (Ferreira et al. 2014). Oberdörster reported that aquatic organisms (e.g., *Daphnia magna* and *Hyalella azteca*) delay molting and thus significantly reduce spring production due to the toxic effect of fullerene (Oberdörster et al. 2006). CNTs are both chemically and thermally stable (Bianco et al. 2005; Lu et al. 2009) and are hence considered one of the most promising engineered nanoparticles. There are two main forms of CNTs (i.e., single-walled carbon nanotubes (SWNTs) and multiwalled carbon nanotubes (MWNTs)) with

different structures and characteristics (Gao et al. 2004). It has been reported that CNTs exhibit a certain bacterial toxicity (Kang et al. 2008a, 2008b; Vecitis et al. 2010), thus raising concern about their negative effects on aquatic environments. GO, the oxidized state of graphene that presents thin film forms, has been implemented in numerous applications, including electrocatalysis (Tang et al. 2009), energy conversion and storage (Becerril et al. 2008; Brownson et al. 2011), filtration and separation (Joshi et al. 2014; Li et al. 2013), and thermal management (Jang et al. 2010) due to its unique physicochemical properties. However, GO is considered to exhibit the greatest toxicity toward organisms among graphene-based nanomaterials (Akhavan et al. 2013; Liu et al. 2011). Therefore, the release of GO into the environment may cause substantial damage to bacteria (Akhavan and Ghaderi 2010) and human stem cells (Akhavan et al. 2012). Carbon dots are emerging carbon nanomaterials with important properties and have drawn much attention in recent years due to their specific properties, such as a high specific surface area, high biocompatibility and great sorption capacity (Kamrani et al. 2017; Liu et al. 2017; Zhang et al. 2014, 2018). However, the exposure of CDs to the aquatic system may also have negative effects on cell viability (Havrdova et al. 2016) and may be highly toxic to zebrafish (Kang et al. 2015).

Metal and metal oxide nanoparticles

The Organization for Economic Cooperation and Development (OECD) has amply elaborated on some anthropogenic nanomaterials, including metals (Ag, ZVI, Cu, Au, etc.) and metal oxides (TiO_2, CeO_2, ZnO, Fe_3O_4, Al_2O_3, etc.) (OECD 2011). The pathways by which metal and metal oxide nanoparticles enter the aquatic environment include air pollution (Stone et al. 2007), agrochemicals (Khot et al. 2012), and construction (OECD 2007). Nanoparticles suspended in the atmosphere will subsequently be deposited on environmental interfaces and ultimately enter water, soil, or the subsurface. The most prominent pathways for metallic nanoparticles entering the aquatic environment are production, manufacturing, and consumption based on a probabilistic model of engineered nanomaterial emissions (Sun et al. 2014). The complexities between material properties and toxicity are highly dependent on the environmental behavior of metal nanoparticles (Peng et al. 2017). Once these nanoparticles are released into the environment, it is not easy to fully control their transport fate and toxicity due to the complex and unsteady properties under a state of suspension in water. Bour et al. noted that the subsequent release of metal and metal oxide nanoparticles may pose a potential risk to ecosystems by bioaccumulating in crops and some aquatic species (Bour et al. 2015). The degree of toxicity of metal and metal oxide nanoparticles to cells is generally ascribed to the nanoparticle-induced formation of excess reactive

oxygen species (ROS), such as hydroxyl radicals or hydrogen peroxide (Peralta-Videa et al. 2011). The ecotoxicological impacts of nanoscale TiO_2, CeO_2, and ZnO particles on fish were investigated, and notable uptake was found only for cerium in the liver of zebrafish after exposure to contaminated water (Johnston et al. 2010). In addition, CeO_2 nanoparticles have been found to be significantly chronically poisonous to algae (Thill et al. 2006) and may induce an oxidative stress response in human lung cells (Lin et al. 2016). The cytotoxicity of metal oxide nanomaterials was found to be related to the charge of the particle to a certain degree. Hu et al. systematically investigated the in vitro cytotoxicity of various metal oxide nanoparticles (e.g., ZnO, CuO, Al_2O_3, La_2O_3, Fe_2O_3, SnO_2, and TiO_2) to a typical organism, *Escherichia coli* (Hu et al. 2009). Among these nanomaterials, ZnO, with the lowest cation charge, showed the highest cytotoxicity, while TiO_2, with a higher cation charge, exhibited lower toxicity. However, a few studies have also indicated that nanoparticles can be transported into other tissues and organs after being absorbed by the gastrointestinal tract and can induce oxidative stress in various cell types (Wang et al. 2007). The exposure to nanoscale TiO_2 is also associated with enduring toxicity, inducing DNA damage (Wamer et al. 1997) and apoptosis (Rahman et al. 2002). The toxicity of some other widely applied metal or metal oxide-engineered nanoparticles (e.g., silver, iron/iron oxide, copper oxide, gold) has been described in detail in a review (Srivastava et al. 2015). The degradation of metal and metal oxide nanomaterials can be realized through sedimentation, biological treatment, sludge processing, and biofilm reactors (Brar et al. 2010; Walden and Zhang 2016). Nevertheless, the elimination of these toxic nanoparticles through the activated sludge process can only be achieved to a certain degree, and complete removal remains a distant prospect (Westerhoff et al. 2013). The model to evaluate the transport fate and negative toxicity impacts of metal and metal oxide nanomaterials upon aquatic systems requires quantitative improvement.

Other nanoparticles

Apart from carbon-based nanoparticles and metal or metal oxide nanoparticles, other nanomaterials, such as QDs (Nozik 2002; Posani et al. 2006), SiO_2 (Deng et al. 2014; Guleryuz et al. 2014), and ludox silica laponite (Xu et al. 2010a, 2010b), are also widely applied in engineering fields. When present in an engineered aquatic environment, these nanoparticles will inevitably interact with different interfaces (Ryan and Elimelech 1996). QDs are composed of a metalloid crystalline core (e.g., CdTe, CdSe) and a protective shell (CdS, ZnS) that shields the core and increases the bioavailability of QDs. However, the photolysis and oxidation reaction may

impact the characteristics of QD surface coatings by dissolving the core, thereby releasing toxic metals as hydrated ions into the aquatic phase (Aldana et al. 2001; Hardman 2006). It was reported that QDs could lead to significant oxidative stress in the gills of the organism, causing DNA lesions and immune toxicity (Gagne et al. 2008). Studies regarding the uptake and toxicity of silica nanoparticles on aquatic biota have also generally been conducted. Fent et al. reported that fluorescent core-shell silica nanoparticles (FSNP) could pose toxic effects on zebrafish in early life stages (Fent et al. 2010). Exposure to SiO_2 nanoparticles will result in dose-dependent cytotoxicity in cultural human cells closely correlated to increased oxidative stress (Lin et al. 2006).

Forces governing colloidal deposition

ENPs can undergo deposition in aquatic systems (Petosa et al. 2010). The depositional behavior of these nanoparticles highly depends on the particle-surface interactions, which can be described by classical DLVO theory of colloidal forces, including van der Waals (vdW) and electrostatic double layer (EDL) interactions, and non-DLVO interactions, such as steric interactions, hydration forces, magnetic forces, and bridging attraction. A counterbalance of all these interactions is considered to play a critical role in determining the potential for nanoparticle attachment to typical interfaces, as well as in the stability of colloidal nanoparticles and their diffusion properties. The pertinent equations used to calculate the commonly encountered interactions in the deposition of nanoparticles onto corresponding surfaces are summarized in Table 1.

Traditional DLVO forces

van der Waals forces Traditional DLVO theory of colloidal stability can commonly model the attractions and repulsive interactions experienced by nanoparticles when approaching charged surfaces. The fundamental assumption of the theory is that the total interaction energy between two colloidal entities is composed solely of vdW and EDL interactions (Verwey 1955). Hamaker proposed that the valuation of dispersion interactions can be estimated by the potential between two surfaces as the sum of the interactions between particles within surfaces (Hamaker 1937). The vdW forces arise between nanoparticles and charged surfaces due to the presence of intermolecular forces associated with polarization of molecules into dipoles. The existence of vdW interactions resulting from electrical and magnetic polarizations leads to changes in the electromagnetic field within the media and in the separation distance between the two surfaces. The vdW

Table 1 Key equations to evaluate particle-surface interactions

Type of interactions	Expression		Ref.
vdW	$U_{vdW} = \frac{A_{123}a_p}{6D\left(1+\frac{14D}{\lambda}\right)}$	(2)	Gregory (1981)
EDL	$U_{EDL} = 64\pi\varepsilon_0\varepsilon_r a_p \left(\frac{k_B T}{ze}\right)^2 \Gamma_1\Gamma_2 \exp(-\kappa D)$	(3)	Usui (1973)
Hydration forces	$U_{HD} = \frac{c_0}{c}\exp(-cD)$	(4)	Levine et al. (1989)
Steric interactions	$F_{ST} = 2\pi a_p \left(\frac{\kappa_B T}{S^3}\right)\left\{\left[\frac{8l}{5}\left(\frac{2l}{D}\right)^{\frac{5}{4}}-1\right] + \left[\frac{8l}{7}\left(\frac{D}{2l}\right)^{\frac{7}{4}}-1\right]\right\}$	(5a)	Byrd and Walz (2005), de Gennes (1987)
	$U_{ST} = -\int_\infty^D F_{ST}dD$	(5b)	
Bridging attractions	$F_B = -4\pi a_p \varepsilon\Gamma\frac{(L^*-D)}{L_c}$	(6a)	Butt and Graf (2003)
	$U_B = \int_\infty^D F_B dD$	(6b)	

All variables are defined in the Nomenclature

forces can be estimated by the total interactions between molecular pairs on two bodies of differing compositions (1, 2; Eq. (1)) and are described by the following expression:

$$A_{123} = \left(\sqrt{A_{11}}-\sqrt{A_{22}}\right)\left(\sqrt{A_{33}}-\sqrt{A_{22}}\right) \quad (1)$$

where A_{123} represents the overall Hamaker interaction parameter for the deposition of a nanoparticle of composition "1" suspended in a medium "2" and depositing onto a surface of composition "3." The Hamaker constants A_{11}, A_{22}, and A_{33} required in the equation are materials interacting across vacuum (Bergström 1997; Ross and Morrison 1988). Meanwhile, the vdW forces are relatively insensitive to changes in solution chemistry, such as ionization density and pH (Chen et al. 2016b).

Electrostatic double layer forces The surface in an ionic solution can easily become charged due to the high dielectric constant of water. When charged nanoparticles approach a surface in an aqueous system, the overlap of diffuse layers of surface charges and ions around the nanoparticles and the surface results in EDL interactions. The forces may vary from characteristics of the colloidal deposition systems, such as ionization or dissociation of surface functional groups, to crystal lattice defects and isomorphic substitution to specific adsorption, thus exhibiting different attractive or repulsive characteristics (Israelachvili 2011; Israelachvili and Pashley 1982). Equation (3) in Table 1 is based on the linear superposition approximation (LSA) method, which is applicable to low surface potentials and symmetric electrolytes (Usui 1973). The mathematical form of equations can vary in consideration of the various approximations derived from the Poisson-Boltzmann equation, such as the constant potential of the charging surface. It has been demonstrated that changes in the sign and magnitude of surface charges (σ) or surface potential (ψ) may coordinate the EDL forces. Furthermore, the concentrations of given ions

and the types of determining electrolytes can also lead to decay in magnitude of the interactions as well (Hogg et al. 1966; Usui 1973).

Non-DLVO interactions

Apart from classical DLVO forces, some other types of forces extended from precepts of the traditional colloidal model, such as repulsive steric interactions, hydration forces, magnetic forces, and bridging attractions, known as non-DLVO forces (energies), are considered to be influential in the colloidal deposition onto various environmental interfaces in aquatic media as well.

Steric interactions The steric force between particles and layers adsorbed by polymers or NOM might lead to steric hindrance. The equations to represent the extent of steric forces are based on Alexander-de Gennes theory, which was proposed to quantitatively assess the repulsive steric forces and the Derjaguin approximation (de Gennes 1985). The magnitude and range of steric repulsion can be affected by many factors, such as the density of the layer, the adsorbed thickness, and the solvate chemistry (Pincus 1991). It has been demonstrated that steric forces can enhance the stabilization of ENPs in aqueous systems (Franchi and O'Melia 2003; Pelley and Tufenkji 2008). Certain studies have examined the influence of biomolecules, such as bovine serum albumin, alginate, and humic acid (HA), on the transport fate of nanoparticles. The results showed that these types of organic matter exhibit profound effects on the stability of particles due to the repulsive steric forces (Chen et al. 2006; Huangfu et al. 2013; Jekel 1986; Kai and Elimelech 2008; Mylon et al. 2004) and thus decreased the extent of nanoparticle deposition on surfaces.

Hydration forces A depositional system is generally characterized as either hydrophobic or hydrophilic depending on the hydrous nature of the colloidal solution. The effect of hydrophobic moieties is, to a certain extent, believed to account for

the decrease in the entropy of water. Nanoparticles carrying ionic functional groups or hydrophilic biomolecules (e.g., proteins, polysaccharides) on their surfaces are often considered to be capable of relatively high degrees of hydration. The attachment process of particles to surfaces that are highly hydrophilic might become hindered due to the existence of strongly hydrated water molecules, resulting in an additional repulsive interaction. The range of hydration forces is wider compared to EDL repulsion; however, no experiments or theory adequately revealing the essence of hydration forces has yet been reported. Furthermore, the hydration force is thought to have a significant impact on the stability of colloidal particles, especially in high-IS conditions where the EDL energy is at its minima (Healy et al. 1980).

Magnetic forces For some iron-based nanomaterials with a magnetic dipole moment, the magnetic force may be the dominant interaction determining the total particle-particle interaction energy and thus affecting the stability behavior of nanoparticles in aqueous systems (Ghosh et al. 2011; Tang and Lo 2013). Even in the absence of an external magnetic field, nanoscale iron can express magnetic dipole attractions between particles (de Vicente et al. 2000). However, distant magnetic particles are not attracted and remain suspended in solution due to the limit of a gradient magnetic field and the velocity and drag forces associated with flowing water even a few centimeters away (Tang and Lo 2013).

Bridging attraction In addition to steric stabilization, the adsorption of polymers may lead to long-range bridging attraction at lower surface coverage, especially for polymers with high molecular weight and at lower surface coverages (Chen et al. 2015). Factors such as the concentration or conformation of polymers can impact the binding strength and influence the attachment behavior of nanoparticles onto interfaces in aquatic media. The bridging effect can be influenced by the chemical properties of solutions, fluid dynamics, and nanoparticle concentration (Ramachandran and Fogler 2000). An increase in hydrodynamic forces and particle concentration is expected to enhance the bridging process (Ramachandran and Fogler 2000), thus leading to a higher affinity of nanoparticles to interfaces.

Experimental approaches for ENP deposition

The most common laboratory approaches employed to quantitatively evaluate the mechanism for deposition of ENPs on interfaces include packed-bed column experiments and laboratory QCM, as schematically presented in Fig. 1. A recent study also proposed that an approach combining time-lapse magnetic resonance imaging and modeling is a useful pathway to investigate the depositional behaviors of nanoparticles in complex environmentally relevant porous media (Lehoux et al. 2017).

Packed-bed column approach

Classic colloid filtration theory Experiments in columns packed with glass beads, sand, or soil are the most common approaches devised to elucidate the depositional behaviors of nanoparticles, such as SWNTs (Jaisi et al. 2008), fullerene (Brant et al. 2005), carbon dots (Kamrani et al. 2017), cerium dioxide (Li et al. 2011), and silver nanoparticles (Lin et al. 2011), in porous media. The transport and deposition of these nanoparticles in porous media are considered to be mainly controlled by processes of advection, dispersion, and attachment. For nanoscale particles (1–100 nm) transported in a steady-state-packed column system, the influence of hydrodynamic dispersion is relatively insignificant, and detachment is typically negligible. The transport and deposition of nanoparticles/colloids is primarily dominated by advection and could be generally described by a one-dimensional advection-dispersion equation (ADE) with a first-order irreversible deposition term (Babakhani et al. 2017; Huang et al. 2011; Van Genuchten and Wierenga 1976; Wiesner and Bottero 2007):

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} - v\frac{\partial C}{\partial x} - kC \tag{7}$$

where t is time (T), x is the distance traveled in the porous media (L), C is the nanoparticle concentration in the liquid phase (N/L^3, where N is the particle number), D is the dispersion coefficient (L^2/T), v is the interstitial particle velocity (LT^{-1}), and k is the particle deposition rate coefficient (T^{-1}).

The mobility and retention of nanoparticles in an experimental granular medium are commonly interpreted with the colloid filtration theory (CFT) (Yao et al. 1971). The relationship of k with two key parameters of CFT, namely, the attachment efficiency (α) and single-collector contact efficiency (η_0), can be described as follows (Elimelech et al. 1995; Johnson et al. 2007a; Tufenkji and Elimelech 2004a):

$$k = \frac{3(1-\varepsilon)}{2d_c}\alpha\eta_0 v \tag{8}$$

where d_c is the median diameter of the porous media, ε is the packed-bed porosity, α is the fraction of collisions resulting in attachment and sorption of nanoparticles on the collector surface, and η_0 describes the frequency of that type of collision occurring with a grain surface. In column experiments, α can be expressed as

Fig. 1 Schematic illustration of the column experiment and QCM
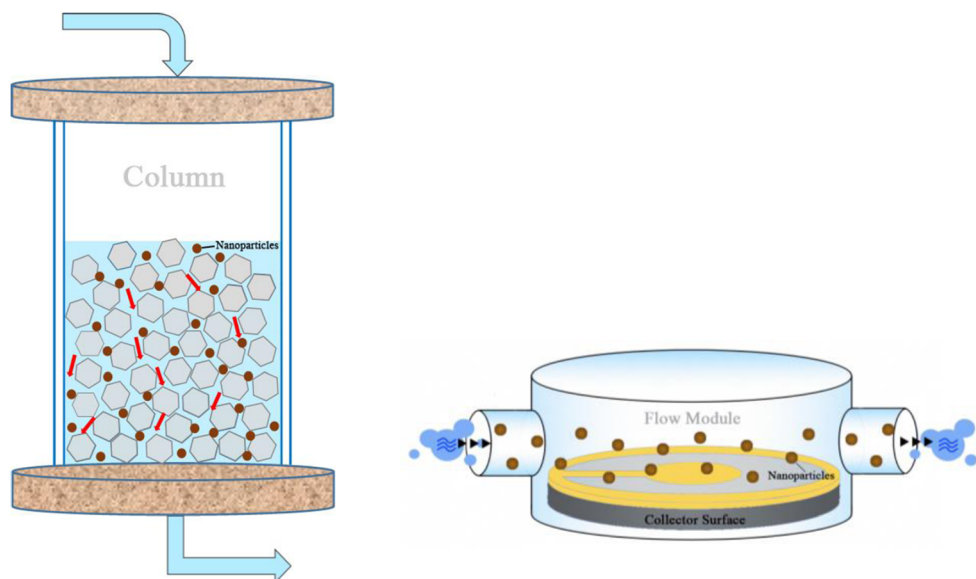
a function of the ratio of the effluent concentration (C) at time t to the influent concentration (C_0) of the nanoparticle suspension, as shown in Eq. (9):

$$\alpha = \frac{-2d_c}{3(1-\varepsilon)\eta_0 L} \ln(C/C_0) \qquad (9)$$

where L is the length of the packed bed. The determination of α requires experimental data from column studies. η_0 can be obtained from theoretical correlation equations or experimental approaches conducted under favorable depositional conditions (i.e., in the absence of repulsive energy barriers between nanoparticles and surfaces).

Deviation from CFT in column experiments It should also be mentioned, however, that the correlations (Eqs. (7), (8), and (9)) based on CFT for nanoparticle deposition in porous media are only explicitly valid for saturated transport systems, which are dominated by advection with irreversible deposition, and cannot fully or adequately describe conditions in which dispersion is the dominant mechanism for nanoparticle transport (Logan 1999). In this case, the depositional behavior of nanoparticles in columns is largely affected by the properties of the granular material surface (e.g., surface charge, hydrophobicity, roughness), which can be easily varied by modification or coatings, such as biofilm or polymers (Lin et al. 2011; Shen et al. 2012; Song et al. 2011; Xiao and Wiesner 2013), and the physicochemical forces between nanoparticles and the collector surface, which determine the fraction of nanoparticles retained following deposition on the collector surface (Ryan and Elimelech 1996).

Although the measured deposition profiles of nanoparticles can be well interpreted by CFT under favorable attachment conditions (in the absence of an energy barrier), deviations in the experimental profiles from theoretical CFT predictions are frequently observed for the deposition of nanoparticles in porous media under unfavorable depositional conditions in which repulsive forces exist (Adrian et al. 2018; Li et al. 2017; Raychoudhury et al. 2014; Ryan and Elimelech 1996; Tufenkji and Elimelech 2004b, 2005; Wang et al. 2012, 2016). Several factors, including heterogeneity in the surface characteristics of particles (Li et al. 2004; Simoni et al. 2000), the interaction energy distributions of deposition (Jaiswal et al. 2009; Tufenkji et al. 2003), and particle detachment (Tufenkji et al. 2003), have been proposed to account for the discrepancies between experimental results and predictions based on CFT. Moreover, the transport and deposition of nanoparticles in porous media are not driven solely by advection, other mechanisms (i.e., detachment, straining, blocking and ripening, as shown in Fig. 2) and simultaneous combinations of these mechanisms also contribute to the deposition of nanoparticle transport in porous media. Overall, the observation of depositional behavior that diverges significantly from the depositional behavior in classic "clean-bed" filtration systems has important implications for the transport of nanoparticles in real environmental conditions with intrinsic complexities and prominent heterogeneities.

Detachment Deposition of nanoparticles onto the solid matrix in porous media is generally referred to as attachment and the opposite mechanism as detachment. Based on DLVO theory, nanoparticle attachment in primary minima has been generally considered to be irreversible based on the reduction in solution electrolyte strength due to the increase in energy well depth, which means that a particle needs to overcome greater attractive energies to detach from primary minima. Increasing
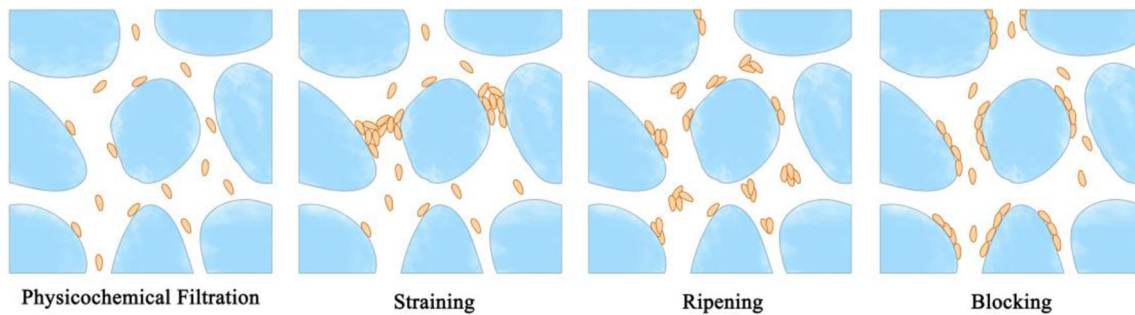
Fig. 2 Schematic of physicochemical filtration, straining, ripening, and blocking in nanoparticle transport in porous media

experimental observations, however, suggest that colloids attached at the primary minima can also be detached by decreasing electrolyte concentration (Molnar et al. 2015; Shen et al. 2014; Tosco et al. 2009; Yi and Chen 2013). The attachment of nanoparticles from collector surfaces might occur due to a change in solution chemistry and/or a disturbance in the hydrodynamics in the system (Bergendahl and Grasso 1999; Li et al. 2017; Wang et al. 2016). Although the importance of solution chemistry for nanoparticle deposition has been widely recognized, several theoretical studies have proposed that surface heterogeneity also plays a critical role in the detachment of nanoparticles from primary minima by reducing the electrolyte strength (Pazmino et al. 2014; Shen et al. 2012; Wang et al. 2016). In particular, a recent study reports that nanoparticles attached on the heterogeneous surface in a primary minimum could not be detached by IS reduction under constant surface potential (CSP) conditions due to the increases in adhesive forces and detachment energy barriers with decreasing solution IS, while the detachment from chemically heterogeneous surfaces by IS reduction could only occur under conditions with a high flow velocity under the linear superposition approximation condition (Shen et al. 2018). Therefore, the heterogeneous properties of the collector surface should be considered in a detachment model for accurate predictions of nanoparticle release behaviors in the subsurface environment, and the fabricated surfaces can be modified via physiochemical approaches to inhibit reversible attachment on collector surfaces.

Straining Straining is a particle-trapping process deemed to occur when the pore spaces between collectors are too small to allow the particles to pass (Fig. 2) (Bradford et al. 2006, 2013; Jaisi et al. 2008; Johnson et al. 2007b; Porubcan and Xu 2011). The contribution of straining to particle removal is not considered in the CFT because the straining process is independent of advection and dispersion and less dependent on colloid-surface interaction (Molnar et al. 2015). Extensive studies, however, have demonstrated that the straining effect is an important mechanism for nanoparticle deposition in porous media under unfavorable attachment conditions (El Badawy et al. 2013; Hong et al. 2009; Raychoudhury et al.

2014; Sun et al. 2015; Wang et al. 2015). Additionally, straining is not purely a physical process as a function of pore throat and particle diameter; the factors of influent concentration (Bradford et al. 2006), hydrodynamics (Du et al. 2013), and ionic chemistry (Shen et al. 2008) are also observed to influence the straining strength in porous media. Straining is usually related to two mechanisms: wedging and bridging. Wedging is the retention of larger particles at two bounding surfaces without particle interference, while bridging refers to the process of multiple nanoparticles arriving simultaneously and accumulating in a pore constriction (Babakhani et al. 2017; Bradford and Torkzaban 2008; Zhang et al. 2012). While the transport models modified by an incorporation of the straining effect can provide more accurate numerical simulations for nanoparticle retention behavior when fit to experimental observations (Flury and Qiu 2008; Shen et al. 2008), they are also subject to the limitations of inferred mechanisms from kinetic coefficients, which has been elaborately reviewed by Molnar et al. (2015). Furthermore, distinctions among the predominance of straining in narrow pore throats versus wedging in grain-to-grain contacts versus retention on the open surfaces of the collector should be made when considering the role of straining relative to other mechanisms (i.e., surface charge heterogeneity and roughness) (Johnson et al. 2011).

Blocking and ripening Blocking and ripening have been demonstrated as important processes that can prevent or accelerate subsequent nanoparticle deposition on collector surfaces in porous media, respectively. Because the surface of porous media may have a limited capacity for the attachment of particles, with increasing accumulation of nanoparticles on the collector surfaces, the interactions between deposited and incoming nanoparticles may reduce the available surface sites for the attachment of subsequent nanoparticles, thus resulting in the so-called blocking (Camesano et al. 1999; Nascimento et al. 2006). During the initial stage of colloid attachment, the attached nanoparticles on collector surfaces are sparsely distributed and essentially do not influence further attachment of incoming nanoparticles from the bulk solution. However, in the later stage of deposition, the deposited particles can act as

additional collectors by forming multiple layers for the attachment of incoming particles, known as ripening (Camesano et al. 1999; Nascimento et al. 2006). Ripening is the opposite of the blocking mechanism, i.e., a gradual decrease in nanoparticle concentration in the effluent and a gradual increase in the attachment rate of attachment with time are observed in the blocking process, while a progressive increase in nanoparticle concentration in the effluent and a decrease in the attachment rate over time are observed in the ripening process (Camesano et al. 1999; Liang et al. 2013; Lu et al. 2013; Nascimento et al. 2006). Although blocking and ripening are two mutually exclusive processes, the occurrence of both phenomena can occur simultaneously in a system due to heterogeneity in porous media (Babakhani et al. 2017; Camesano et al. 1999; Nascimento et al. 2006). Furthermore, it has also been reported that blocking can transition to ripening under certain conditions (i.e., with increasing electrolyte strength) (Afrooz et al. 2016; Chen et al. 2011). Correspondingly, the transition from ripening to blocking was also deemed to occur with decreasing electrolyte concentration in porous media (Chen et al. 2012; Liu et al. 1995; Wang et al. 2015). The shift of the transport behavior from blocking to ripening indicates that temporal variations exist in the nanoparticle deposition rate. The transition between blocking and ripening is crucial because it might alter the predominance of other underlying transport phenomena and thus provide insights into the role of each individual phenomenon when these two behaviors occur along with other transport mechanisms (i.e., straining).

Quartz crystal microbalance approach

The QCM method is also an effective way to explore the mechanisms of nanoparticle deposition onto collector surfaces due to its simplicity and ultrasensitive capability to capture small mass changes over time (Chowdhury and Walker 2012). In experiments using QCM, the increase in the deposited mass of nanoparticles on the crystal sensor leads to a negative shift in resonance frequencies (Δf). The initial deposition rate of nanoparticle, corresponding deposition efficiency in the QCM experiment, is obtained by calculating the rates of frequency changes over a period of time (Chang and Bouchard 2013; Fatisson et al. 2009; Qu et al. 2012):

$$r_f = \left| \left(\frac{\mathrm{d}\Delta f_{(3)}}{\mathrm{d}t} \right)_{t \to 0} \right| \tag{10}$$

$$\alpha = \frac{r_f}{(r_f)_{fav}} = \frac{\dfrac{\mathrm{d}\Delta f}{\mathrm{d}t}}{\left(\dfrac{\mathrm{d}\Delta f}{\mathrm{d}t} \right)_{fav}} \tag{11}$$

$(r_f)_{fav} = \left(\frac{\mathrm{d}\Delta f}{\mathrm{d}t} \right)_{fav}$ in Eq. (10) represents the most favorable deposition rate of nanoparticles and is equivalent to the initial slope of the frequency shift. The collector surfaces are commonly oppositely charged with respect to the nanoparticles or pretreated with a coating of a cationic polymer, such as poly-L-lysine (PLL), to create favorable conditions for nanoparticle deposition. Chen and Elimelech initially adopted this technique to examine the aggregation and depositional kinetics of fullerene C_{60} nanoparticles onto silica surfaces (Chen et al. 2006). Additionally, numerous studies have been performed to derive the depositional behavior of various nanoparticles (e.g., zero-valent iron (Fatisson et al. 2010), cerium dioxide nanoparticles (Liu et al. 2012), carbon nanotubes (Yi and Chen 2011), titanium dioxide nanoparticles (Fatisson et al. 2009), and CdTe quantum dots (Quevedo and Tufenkji 2009)) on various surfaces.

In comparison with the column experiment, the deposition onto collectors employing QCM technology is less dependent on the hydrodynamic parameters (i.e., associated with a homogeneous flow state in an optimized experimental model vs. associated with the complex flow geometry in a column), as schematically shown in Fig. 1. Due to the different governing mechanisms in each experimental system (i.e., physiochemical filtration, straining, blocking, and ripening in the column vs. moderate convective-diffusive transport to the flat surface in QCM), deviations in the trends for deposition kinetics between QCM and column experiments have been observed (Liu et al. 2012; Quevedo et al. 2014). The deposition rates of nanoparticles were reported much lower in the QCM relative to column experiment when suspended in the same electrolyte concentrations (Quevedo et al. 2014). The factors of nanoparticle aggregation and the physiochemical heterogeneity of the sand surface in a column may also contribute to the differences in the observed depositional behavior in the two experimental systems (Liu et al. 2012).

Limitations of employing the QCM method

The QCM technique has been demonstrated to be a powerful tool for characterizing and quantifying the deposition of ENPs on various surfaces in a natural aqueous environment (Chen and Elimelech 2006; Daskal et al. 2017; Kai and Elimelech 2008). QCM with dissipation (QCM-D) can also be used to monitor the dissipation of the crystal surface and provide insights into the dissipative properties and the layer structure of deposited nanoparticles via a combination of resonance frequency shifts (Che et al. 2018; Chowdhury et al. 2014a; Fatisson et al. 2009; Quevedo and Tufenkji 2009; Yi and Chen 2014). Although the QCM method holds great potential to provide rapid, highly sensitive, and real-time monitoring of the deposition of nanoparticles on collector surfaces, there exist great challenges and limitations for examining nanoparticle deposition by employing QCM-D measurements.

First, QCM is a mass-sensitive biosensor, and its sensitivity increases with the increase in the fluctuating mass of deposited nanoparticles. The resonance frequency shifts interpreted in

terms of rigid mass change based on the Sauerbery equation, which relies on the assumption that the rigidly homogeneous layers (Sauerbery 1959) are only valid when the deposited mass causes low dissipation in frequency shifts. However, for deposition systems in which deposited layers of heterogeneous nanoparticles form on the crystals, the frequency changes are not consistent with the predictions of the Sauerbrey relation (Reviakine et al. 2011; Sauerbery 1959). The interpretation of ENP deposition kinetics by the QCM method is limited by the formation of large aggregates (> 700 nm) (Quevedo et al. 2014), which might also lead to non-Sauerbrey behavior. Thus, studies of nanoparticle deposition via QCM technology need to take the viscoelastic properties of the deposited layer into account to perform a more detailed theoretical analysis (Johannsmann et al. 2009).

Furthermore, when studying the depositional behavior of nanoparticles on fluid surfaces, the QCM response is highly dependent on the properties of the interfacial fluid in this system. Difficulties might be encountered when the solution or surfaces are not appropriately prepared for the sensing process (i.e., a highly alkaline or acidic solution), thus making the QCM-D technique inapplicable to the determination of subsequent nanoparticle deposition (Chen et al. 2016b). It should also be noted that external sunlight and additional oxidants or reductants can also change the sensitivities of QCM crystal sensors and the properties of nanoparticles, thus leading to instability and variability in the nanoparticle deposition kinetics. Meanwhile, in deposition studies, the QCM method is sensitive to ambient or slightly elevated temperatures due to the limitations of current QCM piezoelectric sensing materials.

Factors affecting ENP deposition on collector surfaces

Most studies on the environmental fate of ENPs have mostly focused on two interactions: those between nanoparticles and colloidal particles and those between nanoparticles and aquatic interfaces. The internal interaction between colloids, expressed as aggregation behavior, has been widely investigated for fullerene (Chen and Elimelech 2006), cerium oxide (Buettner et al. 2010), manganese dioxide (Huangfu et al. 2013), MWNTs (Yi and Chen 2011), titanium dioxide (Thio et al. 2011), etc. However, the depositional behavior of nanoparticles on naturally occurring surfaces is also considered to play a crucial role in predicting the transport potential of colloidal nanoparticles (Elimelech et al. 1995). The deposition process of ENPs is highly subject to the factors of collector surface type, particle-specific properties (i.e., particle size, particle shape, particle coating, and aggregation), and solution chemistries (i.e., IS, pH, and NOM content) (Liu et al. 2012; Petosa et al. 2010; Wiesner and Bottero 2007).

Collector surface type

The different types of interfaces are considered to be critical factors in determining the particle-collector interactions and thus the deposition and release kinetics of ENPs on environmental surfaces (Chang and Bouchard 2013; Kim et al. 2013, 2014). Collector surfaces with different compositions or ζ potentials may result in different deposition conditions (i.e., favorable conditions for oppositely charged nanoparticles and surfaces and unfavorable conditions for nanoparticles and surfaces with the same charge). It has been reported that the affinities of silver nanoparticles for kaolinite were observed to be significantly higher than for glass beds (GBs) because GBs were more uniformly negatively charged than kaolinite, which presented multiple surfaces of different energies (Zhou and Gunter 1992). It was also reported that surfaces coated with iron oxide and alumina played a more critical role in the deposition kinetics of nanoparticles under unfavorable conditions than did silica (Liu et al. 2012). Because silicon oxide and metal oxide (i.e., alumina and iron oxide) surfaces are some of the most common naturally occurring surfaces associated with transported and deposited ENPs (Quevedo and Tufenkji 2009; Tamura et al. 1996), the depositional behavior of various nanomaterials on three different types of environmental surfaces (i.e., SiO_2, Al_2O_3, and Fe_3O_4) is discussed below. Representative studies devoted to examining the deposition of ENPs on these surfaces are summarized in SI Table S1. The experimental approach performed in each study and the water chemistry are listed. The main findings of these studies are also included.

Deposition on silica surfaces

The silica surface is considered a model collector that represents the sand grain media that nanoparticles are likely to encounter during migration in aquatic systems (Quevedo and Tufenkji 2009). Numerous studies have investigated the depositional behavior of ENPs on bare silica surfaces (Chang and Bouchard 2013; Fatisson et al. 2010; Feriancikova and Xu 2012; Furman et al. 2013; Li et al. 2014; Martin et al. 1991; Quevedo and Tufenkji 2009; Sotirelis and Chrysikopoulos 2015; Yi and Chen 2011). The silica surface is negatively charged (Bergna and Roberts 2005; Yuan et al. 2008) under a normal range of environmental pH values (pH = 5–9) (Crittenden and Montgomery Watson 2005) because its pH_{zpc} is 2.9 (Sverjensky 1994), thus providing favorable conditions for the deposition of positively charged nanoparticles (Furman et al. 2013) but unfavorable conditions for negatively charged ones, as shown in Fig. 3.

For carbonaceous nanoparticles with the same negative charge as the silica surface (i.e., GO, C_{60}, MWNTs, and QDs), attachment onto a silica surface is likely to be largely inhibited by repulsive electrostatic interactions. In cases with
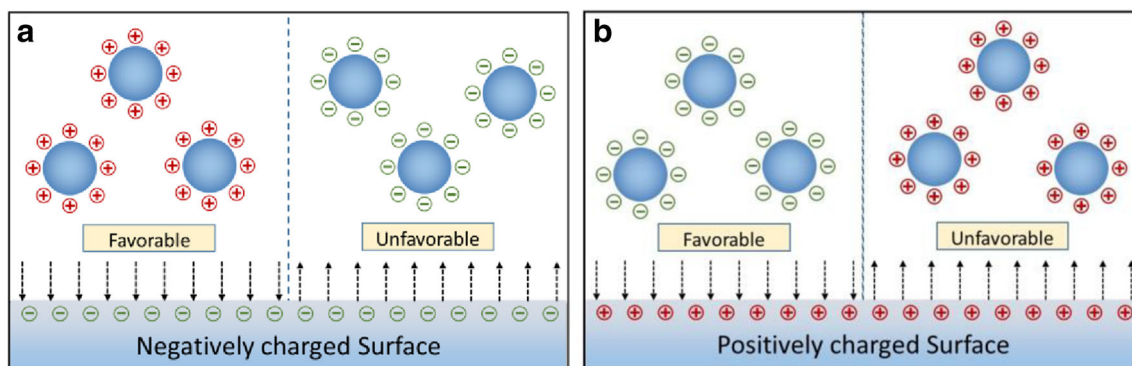
Fig. 3 Different conditions for charged nanoparticle deposition on charged surfaces: **a** favorable condition for positively charged nanoparticles and unfavorable condition for negatively charged nanoparticle deposition on negative surface; **b** favorable condition for negatively charged nanoparticles and unfavorable deposition condition for positively charged nanoparticle deposition on positive surface

pH values between 2 and 10, both quartz sand and GO are negatively charged; thus, it is very unlikely for GO attachment onto the surface due to the existence of a repulsive energy barrier (Sotirelis and Chrysikopoulos 2015). However, the stability of suspended particles is expected to decrease with the decrease in absolute ζ potential values (Sygouni and Chrysikopoulos 2015). In addition, the absolute ζ potential values of both GO and silica substrate suspensions are readily altered by changes in solution chemistry, especially the ion conditions. It has been demonstrated that an increase in IS decreases the ζ potential values of nanoparticles due to compression of the EDL (Feriancikova and Xu 2012; Sygouni and Chrysikopoulos 2015), thus resulting in a higher deposition rate. A study conducted on MWNT nanoparticles showed that there is almost no deposition on silica surfaces in the absence of NaCl as a result of the strong repulsive electrostatic forces between the negatively charged MWNTs and silica surfaces (Martin et al. 1991), while higher deposition rates were observed under high IS because of the reduced repulsive electrostatic forces (Chang and Bouchard 2013). This phenomenon was consistent with previous observations of deposition of CNTs (Yi and Chen 2011) and other nanoparticles such as ZVI (Fatisson et al. 2010), QDs (Quevedo and Tufenkji 2009), and iron oxide (Li et al. 2014) onto unfavorable surfaces. Furthermore, the presence of repulsive interactions under conditions deemed unfavorable was sufficient to weaken the attachment of particles on the surface, thus leading to the release of deposited nanoparticles. Similar results have also been stated in previous studies of particle deposition under unfavorable conditions (Hahn et al. 2004; Tufenkji and Elimelech 2004b). It has been indicated that the drop of divalent cation (i.e., Ca^{2+}) concentration will give rise in the surface potential of both nanoparticles and silica surfaces, resulting in a reduction in energy barrier, thus facilitating the detachment of nanotubes (Chang and Bouchard 2013).

Nevertheless, for some metal-based nanoparticles capable of isoelectric point (IEP) in the ambient pH range, the electrostatic condition corresponds to favorable deposition where the attractive electrostatic forces dominate when nanoparticles are positively charged or the solution chemistry has been changed. The deposition under favorable conditions is considered to be trapped in the primary energy well of the DLVO interaction energy profile. The deposition of positively charged CeO_2 nanoparticles onto negatively charged SiO_2 surfaces was dominated by attractive interactions and in good accordance with DLVO calculation (Li et al. 2011). The results also indicated that the positive potential value will decrease with a further increase in pH; thus, the deposition onto the silica surface may be inhibited. A similar trend was also reported by Pomorska in a study on the depositional behavior of TiO_2 onto self-assembled monolayer (SAM)-modified Au surfaces with opposite charges by employing QCM technology (Pomorska et al. 2011). Under these favorable conditions, no energy barrier is present to date so that DLVO calculations can fully predict favorable conditions for deposition of the positively charged particles onto the negatively charged silica surface (Fatisson et al. 2009). Both $nTiO_2$ and quartz sand are negatively charged at neutral pH conditions where repulsive electrostatic interactions are expected. The ζ potential of nanoscale TiO_2, however, has been found to be more positively charged at very low pH conditions (Thio et al. 2011), leading to increased deposition on the silica surface. In addition, the magnitude of the deposition of TiO_2 onto silica was significantly higher in the presence of Ca^{2+} compared to Na^+ at the same IS. This can be attributed to the higher efficiency of Ca^{2+} in screening the surface charge of nanoparticle reduction of electrostatic repulsive forces (Bizmark and Ioannidis 2015; Domingos et al. 2010), which resulted in stronger electrostatic attractions between positively charged NPs and a negative silica surface. The increase of IS was also reported to create conditions favorable for deposition of some negative nanoparticles. Jiang et al. have indicated that there was no electrostatic repulsion force between ZnO nanoparticle and silica surfaces when IS increased up to 150 mM NaCl and 5 mM CaCl$_2$; thus, favorable deposition was obtained (Jiang et al. 2010). This observation was consistent with the depositional behavior of

TiO$_2$ observed at higher IS (Thio et al. 2011). For those positively charged nanoparticles, the potentials will become less positive or more negative with the increase of IS as a result of the suppression of the EDL and the increase in screening of the silica surface charge (Thio et al. 2011). Therefore, the electrostatic attraction between particles and the surface is weakened and leads to lower deposition rates at high IS. The depositional behaviors on a silica surface can be qualitatively predicted by the conventional DLVO theory of colloidal stability (Derjaguin and Landau 1941; Elimelech et al. 1995).

Deposition on alumina surfaces

Metal oxide surfaces, such as alumina and iron oxide, that exist in aquatic systems are commonly encountered by migrating ENPs. Generally, metal-based surfaces are positively charged at natural ambient pH range (5.0–9.0) (Chowdhury et al. 2013; Crittenden and Montgomery Watson 2005) and thus provide favorable attachment conditions for negatively charged nanoparticles, such as MWNTs, GO, and QDs, but unfavorable deposition conditions for positively charged nanomaterials (Fig. 3). It is generally believed that iron oxide and Al$_2$O$_3$ play significant roles as collector surfaces in the depositional behavior of nanoparticles under environmentally relevant conditions (Liu et al. 2012).

The aluminum oxide surface is representative of oxide patches on the surface of aquifer or filter grains. Because the point of zero charge (pzc) of Al$_2$O$_3$ is reported to range from pH 6 to 10 (Kosmulski 2009; Yopps and Fuerstenau 1964), the ζ potential of bare Al$_2$O$_3$ is usually positive (2–13 mV) under the environmental conditions investigated (Quevedo et al. 2013). A previous study on the deposition kinetics of graphene investigated by Chowdhury et al. selected aluminum oxide surface as a model metal oxide surface (Chowdhury et al. 2014b). The results showed that the deposition of GO on the aluminum oxide surface was favorable and primarily governed by electrostatic properties. Nevertheless, the deposition mechanism of GO on Al$_2$O$_3$ was quite different from that on the SiO$_2$ surface, as stated before, because GO deposition was less efficient on the SiO$_2$ surface than on the Al$_2$O$_3$ surface due to the strong electrostatic repulsions between both negatively charged SiO$_2$ and GO (Ren et al. 2014). For quantum dots, the deposition rate on the bare Al$_2$O$_3$ surface generally decreased with increasing IS, which was in qualitative agreement with DLVO theory (Quevedo et al. 2013). A similar trend was also observed when examining the depositional behavior of SiO$_2$ nanoparticles on alumina by employing QCM-D and an optical reflectometry approach (Guleryuz et al. 2014). However, for extremely high pH solutions, increasing the concentration of NaCl could enhance the deposition of negatively SiO$_2$ nanoparticles due to the compressibility effect of the electric diffuse double layer. In general, the deposition of nanoparticles on Al$_2$O$_3$ highly depends on the solution chemistry, especially pH values, as well as the concentration and valence of background electrolytes. It has been demonstrated that divalent cations (i.e., Mg^{2+} and Ca^{2+}) can destabilize nanoparticle deposition due to their effective charge screening and neutralization (Ren et al. 2014). Furthermore, the deposition of nanoparticles on favorable surfaces is generally considered more irreversible compared to unfavorable surfaces (Chang and Bouchard 2013; Wang et al. 2017). Previous studies have shown that the extent of humic acid deposition on alumina surfaces is considerably high and irreversible (Eita 2011). Research on the deposition of TiO$_2$ nanoparticles onto SAM indicated no obvious release for all tested electrolyte concentrations, confirming that favorable conditions can effectively avoid the release of deposited nanoparticles (Wang et al. 2017).

Deposition on iron oxide surfaces

The mechanism for nanoparticle deposition onto iron oxide surfaces is similar to that for aluminum oxide surfaces due to the same positive ζ potentials of iron oxide in aquatic environments (Liu et al. 2012). The zero charge potentials of iron oxides were reported to be close to the pH values of many natural aquatic systems (Cornell and Schwertmann 1996), typically in the range of 6.6–6.9 for iron oxides in the form of hematite (Parks 1965). Research on the transport of CeO$_2$ showed that the deposition rate on iron oxides was higher than that on Al$_2$O$_3$ surfaces; however, both were lower than that on SiO$_2$ surfaces due to the higher negative charge of the SiO$_2$ surface (Liu et al. 2012). Findings in a column experiment regarding deposition of silver nanoparticles indicated that the attachment of negatively charged silver nanoparticles on hematite-coated glass beads (FeO-GB) was significantly higher than that on bare silica collectors under relatively low pH conditions (Lin et al. 2011), because the iron oxide fraction featured a positive charge and was thus more favorable for deposition of the silver nanoparticles (Lin et al. 2011). The surfaces coated by PLL polyelectrolyte were commonly selected as comparison surfaces for favorable deposition of oppositely charged nanoparticles (Chang and Bouchard 2013; Chen and Elimelech 2006; Chowdhury et al. 2014b; de Kerchove and Elimelech 2007; Kai and Elimelech 2008). On a PLL-coated surface, the primary energy minima dominated nanoparticle depositional behavior, while the secondary energy minima were the main mechanisms governing depositional behavior on the unfavorable surface (Chowdhury et al. 2014b).

Particle-specific properties

Particle size Particle size is a factor that attracts great attention due to its significant effect on nanoparticle retention and transport in aquatic environments. The properties and stabilities of

nanoparticles may vary with variation in particle size (Auffan et al. 2009). During nanoparticle transport in porous media, the dominant depositional mechanism responsible for nanoparticle-collector contact might also vary with varying particle size. The single-collector contact efficiency mentioned earlier accounts for nanoparticle transport through three mechanisms, namely interception, gravitational settling, and Brownian diffusion. Particles at the nanoscale (1–100 nm) tend to collide with surfaces via diffusion; however, when the particle size grows larger, the effects of interception and gravitational settling become dominant (Petosa et al. 2010; Wiesner and Bottero 2007). Thus, nanoparticles with larger sizes are generally found to be less mobile and are more likely to be deposited on surfaces (Darlington et al. 2009; Guzman et al. 2006; Lecoanet et al. 2004; Wang et al. 2015). However, for some smaller nanoparticles, higher retention on surfaces is possible under favorable deposition conditions in which diffusion is enhanced, resulting in more frequent particle collisions and attachment (Wang et al. 2012).

Moreover, the sizes of nanoparticles can be easily altered via aggregation during the transport process (Phenrat et al. 2007; Solovitch et al. 2010), making it difficult to determine the exact role of particle size in particle migration. The conventional DLVO theory discussed previously has proven to be an effective tool for exploring the influence of colloid size on their attachment to different interfaces. It predicts a remarkable increase in the total interaction energy with increasing particle size, resulting in a sharp decrease in the rates of aggregation of colloidal particles (Reerink and Overbeek 1954). Furthermore, based on this classical colloidal theory, the increase in particle size may also increase the height of the secondary energy minimum, enhancing attractive vdW interactions and suppressing the double layer thickness, thus impacting the depositional behavior of these colloids on aquatic surfaces.

Particle shape Changes in nanoparticle shape may substantially alter their interfacial properties and interaction propensities, resulting in unique aggregation and depositional behavior under specific aquatic conditions (Afrooz 2015; Bhattacharjee et al. 2000; Hunter and Chan 1987; Mani et al. 2003). It is generally accepted that the shape of colloids influences the effective drag force acting on the particle in an aqueous suspension (Youngren and Acrivos 1975). Both vdW and EDL forces can be affected by a change in particle shape according to DLVO modeling (Elimelech et al. 1995). In addition, research examining the role of particle shape on the stability of CdSe nanoparticles in aqueous suspensions indicated that the mobility of these rod-shaped particles increased with increasing hydrodynamic radius (Mulvihill et al. 2010). The influence of shape-dependent characteristics on the stability of nanoparticles has also been reported in a range of environmentally relevant solution chemistries; anisotropic nanorods tended to be more

stable than nanospheres in aqueous environments and thus have unique implications for interaction with biological entities (Keller et al. 2010).

Particle coating The extent and type of particle surface modification should also be taken into account when investigating the depositional potential of nanoparticles in aquatic environments. Some ENPs may be subjected to surface modification with polymeric coatings for certain purposes; however, the properties of these particles may in turn be altered by these coatings to some extent. Lin et al. found that steric interactions could hinder the deposition of polymer-coated nanoparticles on the collector surface, and the osmotic contribution to the steric interaction energy was greater than the elastic contribution (Lin et al. 2012; Lin and Wiesner 2012a, 2012b). For polyelectrolyte-coated nanoparticles, electrosteric repulsive interactions might reduce the affinity of particles for surfaces, thus increasing their mobility (Saleh et al. 2008). Furthermore, it has been reported that the attachment of nanoparticles with polymeric coatings onto uncoated glass beads was greater than that on a surface coated with the same polymer (Lin et al. 2012; Lin and Wiesner 2012b), indicating that the repulsive EDL interaction was weaker at the shifted contact frontier of polymeric coatings for particles approaching the uncoated surface (Fig. 4).

Poly(acrylic acid) (PAA) is a common coating polymer, and many studies in the literature have focused on its effects on nanoparticle transport and retention behavior (Dzumedzey et al. 2017; Fatisson et al. 2010; Luccardini et al. 2006; Phenrat et al. 2010; Quevedo et al. 2013; Sirk et al. 2009). This polymer likely stabilizes migrating nanoparticles (Quevedo et al. 2013; Sirk et al. 2009). The reduced deposition of PAA-coated nanoparticles could be attributed to the mechanism of electrosteric repulsion exerted by the polymer coating. When under ion solutions, the polymer coating can lead to electrostatic stabilization and thus inhibit the affinity of nanoparticles for surfaces (Elimelech et al. 1995; Franchi and O'Melia 2003). Nevertheless, the PAA present on particles can be strongly influenced by changes in the IS of the solution (Claesson et al. 2005; Quevedo et al. 2013). Under low-IS conditions, a considerable steric hindrance may be obtained due to the strong electrostatic repulsive force between the anionic groups of the PAA; however, as the salt concentration increases, the extent of steric stabilization decreases, which is attributed to the collapse of the polyelectrolyte layer caused by the reduction in electrostatic repulsion between PAA molecules (Celebi et al. 2007). Consequently, PAA-coated nanoparticles exhibit significant transport potential and reduce the extent of nanoparticle deposition at high salt concentrations.
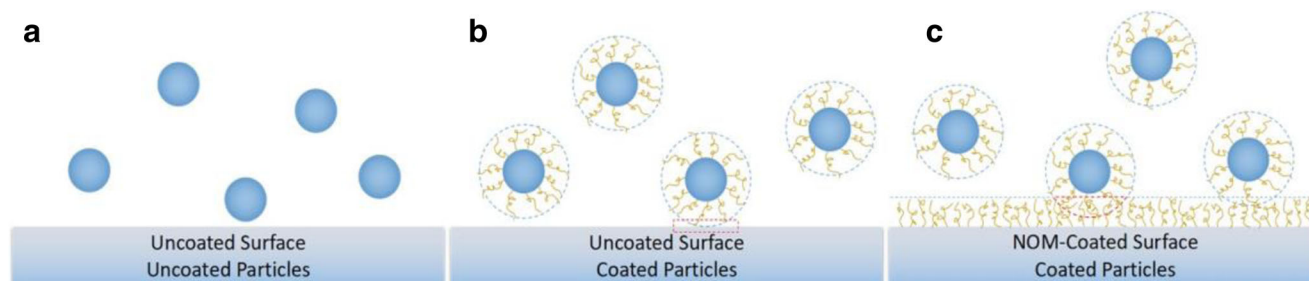
Fig. 4 Illustration of the interactions between particles and collector surfaces with and without surface coatings: **a** no steric interactions between uncoated nanoparticles and uncoated surface, **b** interaction between coated nanoparticles with an uncoated surface (bridging is possible if the polymer can adsorb onto the collector surface (Lin et al. 2012)), and **c** steric stabilization between nanoparticles and surface both coated with polymers

Particle aggregation The aggregation behavior of nanoparticles is also a nonnegligible factor affecting their deposition in aquatic environments. Homoaggregation and heteroaggregation were the two main types of aggregation exhibited by ENPs in an aqueous environment (Hotze et al. 2010). Homoaggregation of nanoparticles leads to higher deposition rates due to the increasing collisions between aggregates and the medium in the groundwater environment (Kanel et al. 2008). In surface waters, homoaggregation can also enhance nanoparticle sedimentation when the formed aggregates reach a sufficient size for interception because of the dominant role of gravity forces other than Brownian diffusion (O'Melia 1980). However, heteroaggregation between colloidal particles with high mobility and low density could facilitate the transport of nanoparticles in porous media by decreasing the particle collision efficiency (Hotze et al. 2010) and thereby increasing their residence time in the aquatic environment. Previous studies on the transport of nanoparticles, such as nanoscale ZVI and TiO₂, have reported that the copresence of suspended clay particles could be used as support for the formation of heteroaggregation to enhance the nanoparticle mobility (Cai et al. 2014; Hydutsky et al. 2007; Tang et al. 2018).

The formation of aggregates subsequently decreases the deposition rate of nanoparticles, such as fullerene (Kai and Elimelech 2008), graphene oxide (Chowdhury et al. 2014a), and quantum dots (Quevedo et al. 2013), due to the reduced diffusion coefficient of the nanoparticles, thus decreasing the convective-diffusive transport of aggregates toward the collector surface. The growth of aggregates over time might further decrease the deposition rate of nanoparticles (Chen and Elimelech 2006; Wei et al. 2016). For the packed column experimental system, the retention of particle aggregates may increase due to deposition into the secondary energy minimum (Pelley and Tufenkji 2008; Saleh et al. 2008) or the effect of physical straining (Bradford et al. 2013; Petosa et al. 2012). Similarly, the deposition of aggregated ENPs in QCM experiments might not always be straightforward to interpret using the traditional approach of mass loading, and nonnormal frequency shifts might be obtained (Lin and Wiesner 2012a; Pomorska et al. 2011).

Solution chemistry

Ionic strength IS is one of the principal parameters that have been extensively investigated with regard to colloidal stability in aqueous solutions (Chowdhury et al. 2014a; Fatisson et al. 2010; Kamrani et al. 2017; Maciejewska-Prończuk et al. 2017; Quevedo et al. 2013). From numerous studies conducted on the effect of iron on the fate and transport of nanoparticles, a general conclusion can be obtained: the extent and magnitude of surface interaction forces, including EDL forces, are significantly affected by IS. In particular, the deposition rate of nanoparticles increases with increasing IS in a specific range, mostly under the critical coagulation concentration (CCC) of colloids. In the narrow range of electrolyte concentrations around the CCC, especially when approaching the critical deposition concentration (CDC), the energy barrier to deposition is almost eliminated as most of the electrostatic repulsive forces are suppressed (Chen and Elimelech 2006). Therefore, particles may undergo the transition from kinetic stability to rapid aggregation, thus resulting in diminished stability (Lin et al. 2010). Moreover, the observed mechanism responsible for the increase in deposition is qualitatively consistent with the DLVO theory (Chowdhury et al. 2014a; Fatisson et al. 2009; Li et al. 2014; Lin and Wiesner 2012a; Wang et al. 2017). Nevertheless, the rate of deposition may tend to be unstable, even potentially decreasing sharp, when the CCC of the particles is exceeded due to the concurrent formation of aggregates. The CDCs of some common ENPs are summarized in SI Table S1.

The depositional behaviors of those particles are highly dependent on the particle and substrate charges, as discussed above. According to classic DLVO theory, an increase in IS will reduce the magnitude of the EDL and repulsive forces (Fig. 5), leading to higher retention in the media. When the particles and the surface are both negatively charged, an intense electrostatic repulsion arises between the particles and the surface due to the ineffective screening effect at low-IS

Fig. 5 Illustration showing adsorbed NOM molecules extruding beyond the electric double layer under high IS
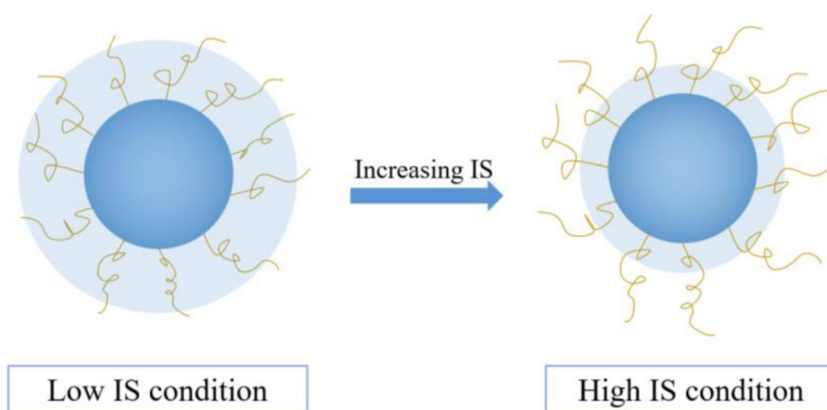
conditions (Chowdhury et al. 2013). An increase in electrolyte concentrations may decrease the thickness of the diffuse double layer and electronegativity of the particles, thus leading to a decline in repulsive electrostatic interactions and therefore enhanced depositional behavior (And and Johnson 2005; Johnson et al. 2007a; Tufenkji 2006; Tufenkji and Elimelech 2005). Furthermore, the strong effect of IS on particle stability may also result in the release of deposited particles from the surface, which was considered associated with the type and extent of particles and the kinetics of varying IS values (Nocito-Gobel and Tobiason 1996; Mcdowellboyer 1992). The primary and secondary minima and the height of the energy barrier can be reduced by changes in solution chemistry, particularly by decreases in the IS (such as by introducing deionized water) and thus facilitating the release of particles (Yi and Chen 2014). Numerous studies examining nanoparticle depositional behavior have emphasized the significant role of cation valence combined with IS on suspension stability, and a general result is that divalent cations (i.e., Mg^{2+}, Ca^{2+}) are more efficient than monovalent cations in accelerating the retention of nanoparticles on interfaces (Chowdhury et al. 2014a; Liu et al. 2012; Quevedo and Tufenkji 2009; Tang and Cheng 2018). Several studies also indicated that the nanoparticles in Ca^{2+} systems exhibiting a higher trend of deposition due to bridging effects were more effective in reducing the energy barrier, while Na^+ or divalent Mg^{2+} do not usually exert bridging ability (Chang and Bouchard 2013; Gutierrez et al. 2010; Nguyen and Chen 2007). The effects of electrolyte valence on nanoparticle stability can also be well interpreted by classic and extended DLVO theory (Zhang et al. 2011). Overall, IS and cation valence provide several strong variables with respect to predicting the environmental mobility and stability of nanoparticles.

Water pH The pH value of the solution is closely interrelated with the ζ potential and electrokinetic properties of nanoparticles. The influence of pH on electrical properties of carbonaceous materials, such as fullerenes (Chen and Elimelech 2009) and carbon nanotubes (Liu and Gao 2005), has been

investigated previously, and the results showed an inappreciable pH effect in an environmentally relevant range of pH values. Nevertheless, in contrast to the above carbon-based nanoparticles, the mechanism of the pH effect on metal oxide nanoparticles (e.g., TiO_2, ZnO, CeO_2) (Chowdhury et al. 2011; Keller et al. 2010; Zhou et al. 2012) was more complicated. Metal-based nanoparticles with an IEP within the natural aquatic pH range can experience electrostatically favorable and unfavorable depositional conditions during transport (Lanphere et al. 2013). Allowing for the considerable impact of pH on the stability of suspended nanoparticles, depositional behavior may be observed at some specific pH values. The deposition of QDs can only occur at relatively low pH conditions (Quevedo and Tufenkji 2009), and GO nanoparticles have also been found to present stronger binding strength to positively charged surfaces at low pH values, resulting in enhanced deposition (Chen et al. 2016a). However, extremely high pH conditions may lead to the destabilization of deposited nanoparticles, thus releasing them from the surface (Chen and Elimelech 2006). Nanoparticles exhibited the lowest degree of stability under conditions in which the pH value was around the pzc or the IEP; hence, an intense transport of particles in solutions with such conditions may occur. It is generally believed that when the pH of solution is below the particle pzc, the nanoparticle surface becomes positively charged, and a further decrease in pH may increase the ζ potential of the surface. In contrast, at pH values above the pzc, the surface is negatively charged, and the higher pH value renders the ζ potential more negative. As a result, the stability of nanoparticles with high ζ potentials (negative or positive) is relatively higher than those with low ζ potentials. Under lower pH conditions, almost all acidic functional groups remain protonated. However, hydroxide ions were still preferentially adsorbed over hydronium ions (Zimmermann et al. 2001) and were a contributor to the negative surface charge of nanoparticles such as fullerene (Ma and Bouchard 2009), even in a neutral solution. Under alkaline conditions with a high pH, particles with surface hydroxyl groups develop a noticeable negative charge density due to deprotonation (Svecova et al. 2008).

Natural organic matter NOM is a heterogeneous mixture of natural macromolecules that are known to be redox-reactive (Chen et al. 2003). NOM is composed of a great variety of organic compounds, such as short-chain acids (humic substances) and large molecules (polysaccharides and proteins) (Allan and Castillo 2007; Walker and Bob 2001; Wilkinson et al. 1999). As one of the naturally occurring surface coatings, NOM widely existed in the aquatic environment and the interactions between NOM and ENPs are considered to play a key role on the physical behaviors including aggregation, deposition, and thus transport of ENPs (Yu et al. 2018). NOM primarily interacts with nanoparticles by adsorption onto particle surfaces, and such behavior can readily alter the magnitude or even the sign of the surface charge, thus causing electrostatic repulsion or steric repulsion between the macromolecular coating and the nanoparticles (Nebbioso and Piccolo 2013; Ramos-Tejada et al. 2003; Tombácz et al. 2004).

HA, alginate, and bovine serum albumin (BSA) are typical forms of NOM that have attracted extensive attention in recent years, and the enhancement in nanoparticle stability and retardation of nanoparticle deposition rate by the adsorption of these organic macromolecules onto nanoparticle surfaces have been widely investigated (Aiken et al. 2011; Chowdhury et al. 2012; Furman et al. 2013; Kai and Elimelech 2008; Liao et al. 2017; Phenrat et al. 2010; Thio et al. 2011; Wu et al. 2018) and reviewed (Philippe and Schaumann 2014). Increased stability and altered depositional behavior of various nanoparticles, such as nC_{60} (Qu et al. 2010), GO ENPs (Chowdhury et al. 2013), TiO_2 (Chen et al. 2012), CeO_2 (Liu et al. 2012), ZnO (Jiang et al. 2010), and Ag ENPs (Furman et al. 2013), were noted for HA by introduced steric hindrance. The trend of the interaction between nanoparticles and an alginate-coated surface, however, exerted some differences from that observed for HA-coated surfaces (Kai and Elimelech 2008). Because alginate has a polysaccharide structure and larger macromolecules than humic acid, the alginate surface was likely rougher than the HA surface. In addition, alginate can undergo esterification in the presence of divalent cations, which may result in the modification of the physical properties and surface morphology of the alginate-coated surface (de Kerchove and Elimelech 2007). BSA is a model protein in aquatic systems that might influence nanoparticle deposition due to its large molecular weight, molecular shape, or abundant functional groups (Huangfu et al. 2013; Kubiak-Ossowska et al. 2017; Park et al. 2017). Previous publications have proposed that the specific conformation of BSA with respect to particles may alter the nature and magnitude of surface interaction forces exerted by BSA molecules (Xu and Logan 2005; Yang et al. 2012), and the attractive regions of BSA may even increase particle deposition (Flynn et al. 2012). A recent study has also demonstrated that different charged proteins induce different depositional behaviors (i.e., enhancing or hindering) of bacteria in porous media (Wu et al. 2018).

It is commonly believed that NOM has a negative potential under environmentally relevant conditions (Buffle et al. 1998), and the addition of NOM, such as HA, to salt solutions can make the charged metal oxide nanoparticles more negative and thus influence their physicochemical properties (Hu et al. 2010; Jiang et al. 2012; Yang et al. 2009). Other researchers also stated that particles coated with organic matter possessed a more negative surface charge than bare nanoparticles (Loux and Savage 2008). In ionic solutions, the presence of NOM can also enhance the stability of ENPs, even at high concentrations of monovalent ions such as NaCl and KCl (Chang and Bouchard 2013; Gallegourrea et al. 2014; Ghosh et al. 2011; Quevedo et al. 2013). Under relatively high-IS conditions, the thickness of the EDL is compressed so that the adsorbed layer of NOM may extend beyond this diffuse double layer (Fig. 5). The mobility of nanoparticles tends to be higher under low-IS conditions due to the dominant role of the interaction between NOM and the particle surface rather than the EDL interaction.

However, this trend is not always applicable to divalent ions (i.e., Ca^{2+} and Mg^{2+}) because divalent species can promote complex interactions between HA and nanoparticles, resulting in the formation of aggregates (Kai and Elimelech 2007; Mashayekhi et al. 2012). The deposition rates of fullerene on surfaces coated with alginate and Suwannee River humic acid (SRHA) or Suwannee River fulvic acid (SRFA) in the presence of calcium ions were observed to be relatively higher than the suppressed deposition rates in a sodium ion solution (Kai and Elimelech 2008). The introduction of divalent ions may also give rise in conformational changes inside NOM molecules (Mylon et al. 2004) or changes in hydrophobicity (Oliveira 1997). In addition, the electron transfer reactions and the adsorption of other ions and molecules in solution can be greatly impacted by the presence of NOM. The elucidation of the mechanism by which NOM interacts with nanoparticles is of vital importance to predicting the fate and transport of nanoparticles in environmental systems. However, the composition and structure of NOM is heterogeneous and shows substantial geographical and seasonal variability due to the many different origins of NOM, resulting in varying effects on the transport behavior of nanoparticles in aqueous suspension (Chefetz and Xing 2009; Kang and Xing 2005; Ritson et al. 2014).

Challenges in predicting nanoparticle deposition in natural environment

The applications of ENPs have radically increased due to their remarkable properties (Baun et al. 2008), thus leading to their emergence in aquatic environments. Increasing attention has been focused on the toxic impacts of ENPs on humans and ecosystems during their environmental transport, with

numerous studies performed to investigate their environmental behaviors, such as deposition. However, we still face many challenges to scientifically assess the environmental impacts of nanoparticles.

First, most studies on nanoparticle deposition have been conducted under controlled laboratory-scale conditions (Batley et al. 2013; Li et al. 2012), and only a few studies have focused on the transport and fate in a realistic aquatic environment (Espinasse et al. 2018; Ong et al. 2017). Findings in experimental conditions can provide some guidelines to better predict the transport fate of nanoparticles; however, some inherent limitations still exist when extrapolating to far more complex natural environmental settings. In the actual natural environment, in addition to factors such as particle-specific properties and solution chemistry, the effects of naturally occurring colloidal particles, bacteria, sunlight, and oxidants may also alter the transport behavior of ENPs suspended in aquatic systems, and the potential effect of biota on ENP behavior in natural environments must also be considered (Klaine et al. 2008). Therefore, the results in actual conditions may exhibit certain differences from experimental observations in designed systems.

Moreover, the detection of ENPs within environmental components will be crucial to establishing better experiments or models for predicting the deposition and release of nanomaterials into the environment. Nevertheless, the environmental concentration of nanomaterials is expected to be at a very low level. Thus, the accurate detection and quantification of nanomaterials in the environmental background is also quite challenging. Some analytical techniques, such as inductively coupled plasma mass spectrometry (ICP-MS) and X-ray absorption and fluorescence spectroscopies (Unrine et al. 2010), have also proved to be able to detect and characterize ENPs at concentrations relevant to the environment. Single-particle ICP-MS (spICP-MS) is a promising technique that has emerged in recent years and can both size and count metal-containing nanoparticles (Pace et al. 2011). Furthermore, spICP-MS could possibly detect ENPs among a high background concentration of naturally occurring particles (Montaño et al. 2014) due to its high specificity and sensitivity (Jiménez et al. 2011; Laborda and Jiménezlamana 2011). However, spICP-MS also faces several analytical obstacles, such as shape and size detection limit, that restrict its applicability (Pace et al. 2011), despite its utility for environmentally relevant samples.

Finally, once released into the natural aquatic environment, the physical and chemical properties of some specific nanoparticles will be readily altered by their environmental transformation. Some studies have adapted in situ transmission electron microscopy using graphene liquid cells (GLC-TEM) to examine the properties of nanomaterials (Park et al. 2015; Wang et al. 2014; Yuk et al. 2012). Field-flow fractionation (FFF) is also a feasible technique for the analytical separation and characterization of nanoparticle suspensions (Messaud et al. 2009; Vastamaki et al. 2005). This technique provides effective separation of nanoparticles with high resolution and information about the mass distribution and chemical composition of nanomaterials. The nanoparticle tracking analysis (NTA) method based on a laser-scattering technique could be used as the online detector of FFF to provide information about particle number concentration (PNC) and particle size distribution (PSD) of ENPs (Bartczak et al. 2015; Luo et al. 2018). However, there is insufficient characterization of particle properties and the relationships between deposition and release behavior. Hence, a quantitative characterization of surface composition and surface chemistry is also essential (Yuk et al. 2012).

In conclusion, while the work devoted to investigating the environmental depositional behavior of ENPs is growing, knowledge gaps associated with their fate and exposure still exist. Therefore, it is important to have a better understanding of the environmental and health impacts of nanoparticles for the development of sustainable nanotechnology.

Compliance with ethical standards

Conflict of interest The authors declare no competing financial interest.

Abbreviation

ENPs	engineered nanoparticles
DLVO	Derjaguin-Landau-Verwey-Overbeek
NOM	natural organic matter
CNTs	carbon nanotubes
GOs	graphene oxide
CDs	carbon dots
ZVI	zero-valent iron
QDs	quantum dots
SWNTs	single-walled carbon nanotubes
MWNTs	multiwalled carbon nanotubes
OECD	Organization for Economic Cooperation and Development
ROS	reactive oxygen species
FSNP	fluorescent core-shell silica nanoparticles
vdW	van der Waals
EDL	electrostatic double layer
LSA	linear superposition approximation

QCM	quartz crystal microbalance
CFT	colloid filtration theory
ADE	advection-dispersion equation
CSP	constant surface potential, IS ionic strength
PLL	poly-L-lysine
QCM-D	quartz crystal microbalance with monitoring
IEP	isoelectric point
SAM	self-assembled monolayer
FeO-GB	hematite-coated glass bead
pzc	point of zero charge
PAA	polyacrylic acid
CCC	critical coagulation concentration
CDC	critical deposition concentration
HA	humic acid
BSA	bovine serum albumin
$SRHA$	Suwannee River humic acid
$SRFA$	Suwannee River fulvic acid
ICP-MS	inductively coupled plasma mass spectrometry
$spICP$-MS	single-particle inductively coupled plasma mass spectrometry
GLC-TEM	transmission electron microscopy using graphene liquid cells
FFF	field-flow fractionation
NTA	nanoparticle tracking analysis
PNC	particle number concentration
PSD	particle size distribution

Nomenclature

A_{123}	Hamaker constant of nanoparticle-medium-substrate system
U_{vdW}	Van der Waals interaction energy
a_p	particle radius
D	particle to surface separation distance
λ	characteristic wavelength
U_{EDL}	electrical double-layer interaction energy
ε_0	dielectric permittivity in vacuum, 8.85×10^{-12} F/m
ε_r	relative dielectric permittivity of solution
k_B	Boltzmann constant, 1.3805×10^{-23} J/K
T	absolute temperature
e	electron charge, 1.602×10^{-19} C
$\ddagger$	counterion valence
Γ_i	dimensionless surface potential for particle or collector, $\Gamma_i = \tanh[(\ddagger e \,\psi_i)/(4k_B T)]$
κ	inverse Debye length
U_{HD}	the hdration interaction energy
c_0, c	empirical constants
F_{ST}	the steric force
S	distance between polymer chains on a surface
l	the film thickness
U_{ST}	the steric interaction energy
F_B	the bridging force
U_B	the bridging interaction energy
L_C	units segment length in polymer chain
L^*	critical hydrocarbon chain length
C	nanoparticle concentration in the liquid phase
x	the distance traveled in the porous media
v	the interstitial particle velocity
k	the particle deposition rate coefficient
α	the attachment efficiency
η_0	single-collector contact efficiency
d_c	the median diameter of the porous media
ε	the packed-bed porosity
L	the length of the packed bed
C_0	the influent concentration

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