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Co-transport of polystyrene microplastics and kaolinite colloids in goethite-coated quartz sand: Joint effects of heteropolymerization and surface charge modification

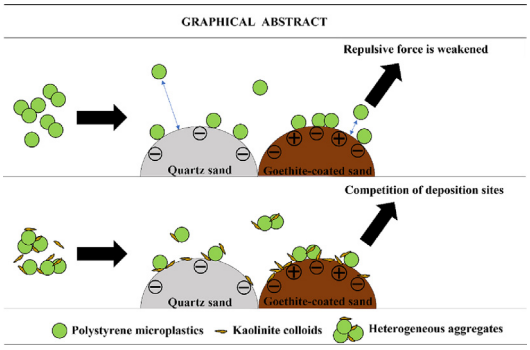
Bokun Chang [a], Bing He [a], Gang Cao [a,b], Zhiying Zhou [a], Xiaoqi Liu [a], Yajun Yang [a], Chenyang Xu [a], Feinan Hu [a,c], Jialong Lv [a,*], Wei Du [a,*]

[a] College of Natural Resources and Environment, Northwest A&F University, Yangling 712100, China
[b] Soil Physics and Land Management Group, Wageningen University & Research, 6700 AA Wageningen, The Netherlands
[c] Institute of Soil and Water Conservation, Chinese Academy of Sciences and Ministry of Water Resources, Yangling 712100, China

HIGHLIGHTS

- The electrostatic repulsive force is the key influential factor for MPs transport.
- Surface modification-induced charge heterogeneity by goethite inhibits MPs transport.
- Coexisting kaolinite colloids facilitate the transport of MPs in saturated porous media.
- Kaolinite colloids compete with MPs for limited deposition sites on porous media.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigated the transport behavior of polystyrene microplastics (MPs) in saturated quartz sand and goethite-coated sand in the presence of coexisting kaolinite colloids. Column experiments were conducted under a wide range of solution chemistry conditions, including pH levels of 6.0, 7.0, and 9.0, as well as background Na^+ concentrations of 5 mM and 25 mM. We found that: (1) The individual transport of MPs in porous media diminished both with increasing background ion strength and decreasing pH, and its transport ability was significantly dominated by the interactions between MPs and porous media rather than the interplay between MPs, which has been further corroborated by the aggregation stability experiments of MPs particles. (2) MPs had a much lower ability to move through goethite-coated sand columns than quartz sand columns. This is because goethite coating reduces the repulsion energy barriers between porous media and MPs. The increased specific surface area and surface complexity of sand columns after goethite coating should also account for this difference. (3) MPs transport would be subjected to the differentiated impact of co-transported kaolinite colloids in the two types of porous media. The promotion effect of kaolinite colloid on MPs' transport capacity is not significantly affected by background ionic strength changes when quartz sand is served as the porous medium; however, the promotion effect is highly correlated with the background ionic strength when goethite-coated sand is served as the porous medium. In comparison with low background ionic strength conditions, kaolinite colloids under high background ionic strength conditions significantly facilitated MPs transport. This is mainly because under high background ionic conditions, kaolinite colloids are more likely to be deposited on the

* Corresponding authors.
E-mail addresses: ljlll@nwsuaf.edu.cn (J. Lv), weidu@nwafu.edu.cn (W. Du).

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surface of goethite-covered sand, competing with MPs for the limited deposition sites. The extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory is applicable to describe the transport behavior of MPs.

1. Introduction

Plastics are widely employed in a variety of fields, including agriculture, electronics, automobile, construction, and packaging (Akdogan and Guven, 2019; Zhang et al., 2021a). The global annual production of plastics is estimated at >320 million tons according to recent reports (Guo et al., 2020). Plastic pollution is regarded as a threat to natural ecosystems, and its negative impacts on microorganisms, plants, marine and terrestrial creatures, as well as human health, have received widespread attention (de Souza Machado et al., 2018; Ng et al., 2018). The potential risks of plastics are highly related to their distribution in the natural environment (Peng et al., 2017; Tan et al., 2022). Mulching with plastic film, dumping sewage sludge, irrigating and flooding waste waters, and atmospheric deposition are considered to be major sources of plastics in soil (Elgarahy et al., 2021; Jung et al., 2021). Researchers have found that soil is a major source-sink of plastic waste, and the amount of plastic released in terrestrial ecosystems is 4 to 23 times greater than in marine ecosystems. However, the majority of studies on plastics have focused on aquatic environments, and far less has been done on plastics in soil. Through physical fragmentation, mechanical abrasion, chemical weathering, UV radiation, and (micro)biodegradation, large-sized plastic debris left in the soil gradually transforms into microplastics (MPs, <5 mm in diameter) that are more difficult to be removed from the environment (Koelmans et al., 2016). The global abundance of MPs has increased exponentially over the past few years, resulting in the environmental problems associated with MPs pollution becoming increasingly prominent (Luo et al., 2021). For example, MPs can carry harmful pollutants over long distances, threatening ecosystems and human health (Alimi et al., 2018). As a result, efforts to understand the distribution and transport mechanisms of MPs in the environment, as well as their harmful environmental effects, are critical (Castro-Castellon et al., 2022).

MPs transport is influenced not only by their own properties (e.g., size, shape, type), but also by their ambient transport environments (e.g., porous media types, temperature, and water chemistry conditions) (Lu et al., 2021a; Oladoja and Unuabonah, 2021; Xu et al., 2021; Zhao et al., 2021). Previous studies have shown that, the surface properties of MPs and their interactions with other colloids, such as clay colloids, in a suspension are influenced by hydrochemical conditions such as pH and ionic strength (Li et al., 2020; Li et al., 2021b). Furthermore, the hydrochemical environment also impacts the surface charge of the soil matrix, which affects the deposition of MPs on the surface of the porous medium (Wang et al., 2022). These internal and external factors are coupled together during the transport of MPs and, along with other solutes in the soil solution, have an impact on and control the environmental transport characteristics of MPs. Kaolinite, for example, as the most prevalent clay mineral and active component in soil, its colloidal particles can adsorb onto the surface of MPs via electrostatic attraction, ligand exchange and cation bridging, resulting in the formation of the kaolinite-plastic heterogeneous aggregates (Li et al., 2020). The formation of heteroaggregates can result in changes in particle size and surface properties(surface charge heterogeneity) (Katzourakis and Chrysikopoulos, 2014; Li et al., 2021b), which significantly impact the transport of MPs in water. The presence of kaolinite colloids or biocolloids has been found to significantly impact the transport of nanoparticles in porous media, highlighting the potential role of these colloidal particles in the spread of environmental contaminants (Chrysikopoulos et al., 2017; Georgopoulou et al., 2020). In addition, recently developed mathematical models can well simulate the coagulation and migration behavior of colloids in porous media (Katzourakis and Chrysikopoulos, 2014; Katzourakis and Chrysikopoulos, 2015; Katzourakis and Chrysikopoulos, 2021).

Goethite, the most widely distributed and active iron oxide in soils and sediments, is of interest because of its unique electrostatic properties (Chen et al., 2021; Duster et al., 2017). Goethite has a positive charge in acidic or near-neutral pH waters and has a high affinity for negatively charged quartz sand (Qi et al., 2019; Wang et al., 2013). It has been demonstrated that positively charged hydroxy iron oxide can change the complexity and electronegativity of the quartz sand surface, thereby controlling the deposition of negatively charged quartz sand (Rastghalam et al., 2019). There have been some developments in research on the effects of coexisting clay minerals on the transport of MPs when purely quartz sand acts as a porous medium (Rastghalam et al., 2020). However, it is not clear whether the findings of these studies can be applied to disclose the MPs' transport in goethite-coated heterogeneous porous media appropriately. As the transport of MPs shifts from individual to co-transport with clay minerals and the simulated environment shifts from a simple homogeneous purification system to a heterogeneous, relatively complex multi-component system, more research is needed to depict the aforementioned changes in MPs transport, and the underlying mechanism should be elucidated.

Here, polystyrene microspheres were used as a model substance to systematically study their transport in different hydrochemical environments (i.e. pH 6.0, 7.0, and 9.0 and background electrolyte Na^+ concentrations of 5 mM and 25 mM). In this study, we aim to (1) investigate the effect of goethite on MPs transport in saturated porous media; (2) investigate the effect of kaolinite on MPs transport in saturated porous media; and (3) clarify the differences in MPs transport mechanisms between homogeneous and heterogeneous environments using the extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) theory.

2. Materials and methods

2.1. Preparation of micro-nano plastic and kaolinite colloid particles

2.1.1. Micro-nano plastic particles

In this study, the monodisperse fluorescent polystyrene (Tianjin Bessler Chromatography Technology Development Center, China) with particle size of about 1 μm (Fig. S1), density of 1.05 g/cm^3, maximum excitation wavelength of 470 nm, maximum emission wavelength of 526 nm, excitation and emission slit width of 5 nm, has been employed. The concentration of MPs in surface water can vary significantly, ranging from 0.1 to 10,000 particles per liter (Guo et al., 2020; Sharma et al., 2021). To investigate the behavior of MPs in porous media such as soil and sediment, previous studies have used concentrations of MPs ranging from 1 to 30 mg per liter (mg/L) (Li et al., 2020; Li et al., 2021a). In this study, the concentration of the MPs suspension was set at 10 mg/L to observe the transport and deposition behavior of MPs under the specific conditions of the experiment. MPs surface functional groups were analyzed using Fourier transform infrared spectroscopy (FTIR) (Nicolet iS 50, Thermo Scientific, America) (Fig. S2).

2.1.2. Kaolinite colloid particles

10 g of kaolinite powder was dissolved in 1 L of pure water and ultrasonically dispersed for 1 h at 50 % power (450 W) using a cell crusher (XO-900D, Nanjing Xian-Ou Instrument Manufacturing Co, Ltd., China). The centrifugal speed required to extract kaolinite colloids (<1 μm diameter) was calculated according to Stokes' law to be 9500 r/min for 5 min and 54 s(Fig. S3) (Li et al., 2012). The concentration of the extracted kaolinite colloid was approximately 400 mg/L as measured by the weight method. The concentration of inorganic clay mineral colloids in the natural environment ranges from a few hundred μg/L to several hundred mg/L (Li et al., 2020). It was decided to set the concentration of kaolinite colloid at

50 mg/L in the experiment in order to ensure that a certain amount of kaolinite colloid can be adsorbed on the surface of MPs. Model kaolinite colloids were characterized using FTIR (Fig. S4), and X-ray diffraction (XRD) (Ultima IV, Hitachi, Japan) (Fig. S5).

The suspension stock solutions of MPs and kaolinite colloids were respectively diluted to 10 mg/L and 50 mg/L by background electrolyte solution for MPs transport separately and co-transport with kaolinite colloids (Tong et al., 2020). The background electrolyte (NaCl) concentrations of 5 mM and 25 mM were set as low and high ionic strengths in the experiments, which conformed to the Na^+ concentration range in natural aquatic systems. Considering that the kaolinite colloid is dispersed in the pH range of 6–9 (Tong et al., 2020), and the initial pH of all suspensions was adjusted to 6, 7, and 9 using 0.1 mol/L NaOH or 0.1 mol/L HCl, respectively. Prior to carrying out the transport experiments, the MPs suspension was sonicated for 20 min in order to ensure its stability. A high sensitivity zeta potential analyzer (ZETA PALS, Brookhaven, USA) was used to determine the zeta potential of MPs and MPs-kaolinite under different hydrochemical conditions. The hydrodynamic diameter (d_{DLS}), surface morphology, and primary dimensions of MPs and MPs-kaolinite heterodimers were measured by dynamic light scattering (Brookhaven, Omni, USA) and transmission electron microscopy (TEM), respectively. The scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS) (S-4800, Hitachi, Japan) was applied to observe the deposition of kaolinite colloids and MPs at the entrance of the sand column (Fig. S6).

2.2. Porous media

The quartz sand (SiO_2, Shaanxi Zhouzhi County Quartz Sand Co, Ltd. China) with a particle size of 0.74 to 1.18 mm was washed with HNO_3 (1.5 mol/L), NaOH solution (0.25 mol/L), and deionized water (DI) sequentially to remove impurities (Ling et al., 2021). Then it was dried at 105 °C and stored in sealed jars under dry conditions at room temperature (Dong et al., 2021). The bulk density of quartz sand is about 1.7 g/cm^3. Mixing together 87.5 mL of 0.17 mol/L $Fe(NO_3)_3$ solution, 90 mL of 0.52 mol/L NaOH solution, and 500 g of quartz sand. After the hydroxyiron oxide precipitated onto pure quartz sand, the mixture was dried in an oven at 105 °C for 12 h with regular stirring to prevent the salt from crystallizing on the surface. Subsequently, the mixed solids were washed with HCl (1 mol/L) and deionized water and then dried at 60 °C for 24 h (Qi et al., 2019; Wang et al., 2017). Fe signals were detected in the coating phase, according to SEM-EDS analysis, demonstrating the presence of the hydroxy iron oxide coating on the quartz sand surface. Using a BET surface area analyzer (ASAP 2460, Micromeritics, America), the specific surface areas of quartz sand and goethite-coated sand were measured, which were 0.0112 g/m^2 and 0.0591 g/m^2, respectively. 10 g of quartz sand/goethite-coated sand and 40 mL of background electrolyte solution were placed in a 100 mL triangular flask and sonicated for 10 min at 50 % power (450 W) using a cell crusher (XO-900D, Nanjing Xian-Ou Instrument Manufacturing Co., Ltd., China). The zeta potentials of goethite-coated sand and quartz sand under different water chemistry conditions were determined using a high sensitivity zeta potential analyzer, which are listed in Table 1. The method for determining the zeta potential of quartz sand and goethite-coated sand in detail can be found in the supplementary material, specifically in Text S4.

2.3. Column experiments

The column transport experiment was used to investigate the vertical transport behavior of MPs in saturated porous media. Clean quartz sand was wet-packed into a cylindrical Plexiglas column (10 cm in length and 2 cm in diameter). The average porosity of the filled column was 0.42 and the average pore volume (PV) was 45 mL. The porous medium is supported at the top and bottom of each column by a nylon mesh with a 50 μm pore size. A series of column transport experiments (MPs transport individually and co-transport with kaolinite colloids) were conducted under different hydrochemical conditions (background Na^+ ions, ion concentrations of 5 mM and 25 mM, pH 6.0, 7.0, and 9.0). The direction of water flow in the column experiments was always bottom-up, considering that gravity is an important driving force for colloidal deposition (Chrysikopoulos and Syngouna, 2014). Prior to performing the column transport experiment, a peristaltic pump (HL-2B, Shanghai Luxi Analytical Instrument Co., Ltd., China) was used to inject at least 10 PV of deionized water from bottom to top in order to remove impurities from the quartz sand column, then 3 PV of background electrolyte was injected to stabilize the chemical conditions of the solution. For this experiment, 3 PV of MPs/MPs-kaolinite mixture suspension with the same background ionic strength was first flowed through the sand column with a constant flow rate of 1.0 mL/min, followed by 3 PV of background electrolyte. Effluent was continuously collected at 10-min intervals using a four-channel partial automatic collector (EBS-20, Shanghai Luxi Analytical Instrument Co., Ltd., China). The concentration of MPs in the effluent was measured by fluorescence spectrophotometer (LS55, Perkin Elmer, USA) and the breakthrough curve (BTC) was plotted according to the changes in MPs concentration. Each treatment was repeated twice.

The deposition sites competition experiments were carried out with background ionic strengths of 5 mM and 25 mM and a pH of 7 to determine whether kaolinite and MPs compete for deposition sites in porous media. Previous studies have demonstrated that kaolinite colloids can pass through a sand column almost completely after 2 PV and completely after 3 PV (Ji et al., 2023). First, the kaolinite colloid of 3 PV was injected into the sand column, ensuring that part of the kaolinite colloid was deposited on the surface of the porous medium. The background electrolyte solution of 3 PV (to exclude the undeposited kaolinite colloid in the pore water to prevent it from affecting the transport of MPs), the MPs suspension of 3 PV, and the background electrolyte solution of 3 PV were then passed through the porous sand column orderly. Previous studies have shown that the surface covered by kaolinite does not affect the overall porosity of porous media (Li et al., 2020).

Table 1
Summary of experimental conditions and physical and chemical information.

MPs (mg/L)	Kaolinite (mg/L)	pH	Cations	Ionic strength (mmol/L)	DLS diameter (nm)	PS-Zeta (mV)	Quartz sand-zeta (mV)	G-C sand-zeta (mV)
10	0	6	Na^+	5	988(±13)	−20.3(±2.7)	−39.0(±4.0)	−20.0(±1.1)
10	0	7	Na^+	5	1150(±175)	−23.3(±5.7)	−41.3(±2.7)	−22.3(±2.0)
10	0	9	Na^+	5	1204(±252)	−28.2(±4.6)	−43.4(±2.6)	−22.9(±2.7)
10	0	6	Na^+	25	1052(±160)	−12.7(±0.8)	−34.7(±3.1)	−16.5(±2.5)
10	0	7	Na^+	25	1091(±53)	−14.0(±1.0)	−36.0(±3.0)	−17.6(±3.0)
10	0	9	Na^+	25	1160(±140)	−19.0(±2.6)	−37.6(±3.2)	−17.8(±3.2)
10	50	6	Na^+	5	2350(±450)	−33.3(±9.7)	−36.3(±2.9)	−18.1(±7.1)
10	50	7	Na^+	5	3170(±220)	−35.8(±7.2)	−37.2(±9.8)	−23.2(±4.7)
10	50	9	Na^+	5	3640(±480)	−40.3(±6.3)	−38.3(±1.3)	−23.3(±4.4)
10	50	6	Na^+	25	2410(±350)	−22.7(±3.0)	−36.5(±1.3)	−16.9(±5.0)
10	50	7	Na^+	25	1890(±470)	−25.6(±1.6)	−37.9(±4.1)	−17.2(±0.9)
10	50	9	Na^+	25	3000(±260)	−30.0(±2.6)	−38.3(±4.0)	−21.5(±5.1)

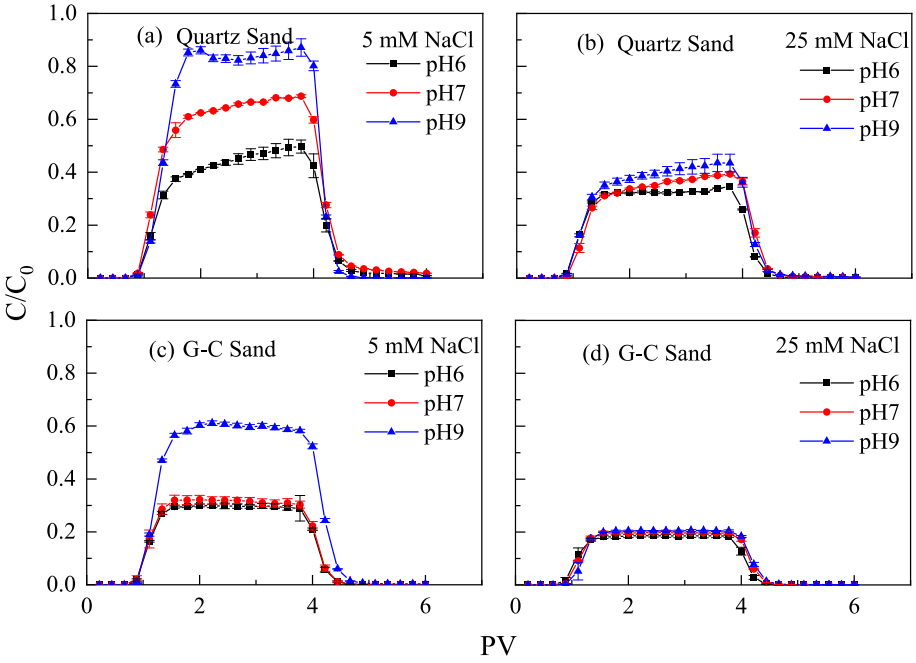

Fig. 1. BTCs of MPs transport alone in porous (a, b) sand and (c, d) G-C sand column under various pH and ionic strength conditions.

2.4. Deposition experiments

MPs deposition experiments on porous media were performed to determine MPs binding affinity to the two porous media during transport. Add 5 g of quartz sand (or gothite-coated sand) and 20 mL of MPs suspension (or MPs-kaolinite mixture) to a 50 mL volumetric flask, and the hydrochemical conditions of the suspension were kept consistent with the transport experiment. The mixed suspension were shaken at 100 rpm/min for 135 min, the same duration as the MPs transport experiment. The triangular vials were covered by sealing film to avoid sample loss during shaking. Blank control experiments (MPs only, no porous media) were also carried out, which verified that MPs deposited on the glassware could be ignored. All deposition experiments were repeated three times, and a spectrofluorometer was used to determine the mass loss of MPs in suspension.

2.5. Aggregation experiments

The aggregation kinetic curves of MPs were measured by a dynamic light scattering instrument with an incident wavelength of 635 nm and a scattering angle of 90°. A suspension of MPs at a concentration of 20 mg/L was prepared, and it was ensured that the suspension could be stably dispersed after sonication. The pH of the suspension was adjusted to 6, 7, and 9 with HCl and NaOH solutions. 1.8 mL of MPs suspension was mixed with equal volumes of electrolyte solutions of different concentrations, i.e., the measured concentration was 10 mg/L. The d_{DLS} values were recorded every 9 min, and an aggregation kinetic curve was obtained after monitoring for 135 min (consistent with the transport experiment time).

2.6. Mathematical model

The interactions between MPs and porous medium were quantitatively characterized by the XDLVO theory, in which the XDLVO energy (ΔG^{XDLVO}) is made up of van der Waals attraction (ΔG^{LW}), electric double layer repulsion (ΔG^{EDL}), and Lewis acid-base interactions (ΔG^{AB}) (Gregory, 1981; Hogg et al., 1966; Van Oss et al., 1988). Other details are provided in Supplementary Data Text S1.

3. Results and discussion

3.1. Individual transport of MPs in saturated quartz sand

The effects of pH and ionic strength on the transport behavior of MPs are illustrated in Fig. 1, from which the following observations could be obtained: First, the transport capacity of MPs in saturated porous media decreases with increasing ionic strength at a given pH. For example, the mobility of MPs decreased from 45.9 % to 32.8 % at pH 6, from 66.8 % to 36.5 % at pH 7, and from 80.3 % to 59.7 % at pH 9 in quartz sand filled column system as Na^+ concentration increased from 5 to 25 mM, respectively (Fig. 1a and b). Second, the MPs' transport ability increases with increasing pH. For instance, the MPs transport rates increased from 45.9 % to 80.3 % as the pH ranged from 6 to 9 at an ionic strength of 5 mM, and from 32.8 % to 40.4 % as the pH ranged from 6 to 9 at an ionic strength of 25 mM in quartz sand filled column system, respectively (Table 2). Classically, ionic intensity and pH could influence the transport behavior of MPs primarily by changing the interparticles stability of MPs and their interaction with porous media (Ren et al., 2021). Based on this, the aggregation and the deposition experiment of MPs were conducted to clarify the mechanism of the effects of ionic intensity and pH on the transport capacity of MPs.

Recently proposed models demonstrate that the aggregation of nanoions can alter the average adhesion rate and affect its mobility (Katzourakis and Chrysikopoulos, 2015). Interestingly, aggregate kinetics experiments of MPs in Fig. 2a showed that there were no significant changes in the hydrated particle sizes of MPs in different hydrochemical environments, and that no notable aggregation phenomena occurred between MPs particles during the transportation process. Also, the results of the settling experiments demonstrate that MPs are relatively stable in different solution environments (Fig. S8). Previous studies have indicated that the critical aggregation concentration (CCC) for 50 nm NPs is 225 mM and for 500 nm MPs is 770 mM in Na^+ systems (Liu et al., 2021). Therefore, it can be inferred theoretically that the 1 μm MPs used in this study had a larger CCC value, and MPs are less likely to cause aggregation in this experiment. As a result, it is reasonable to conclude that the hydrochemical environment in this study affects the transport behavior of MPs primarily by altering the interaction between MPs and porous media rather than by changing the interaction between MPs.

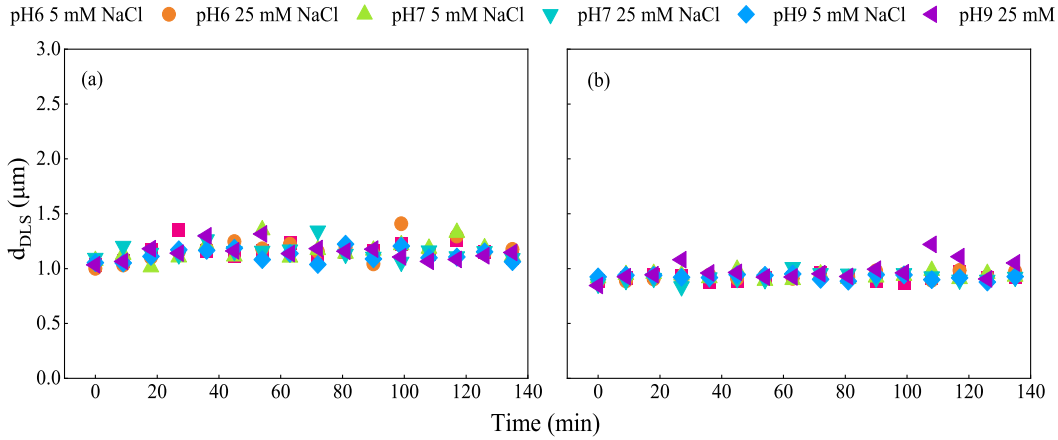

Fig. 2. The homogenous aggregation kinetics curves of MPs (a) and the heterogenous aggregation kinetics curves of MPs with kaolin colloids (b) in NaCl solution.

Deposition experimental results (Fig. 3a) theoretically inferred that the relatively higher ionic strength and lower pH could inhibit the transport ability of MPs by increasing affinity between MPs and porous media, which is consistent with the observations in Fig. 1. According to the classical DLVO theory, the increase in cationic strength leads to an accumulation of counterions around the negatively charged surfaces of MPs and porous media, which further screen the surface negative charges and lower the zeta potential (Table 1) (Wu et al., 2020). The decrease in surface electronegativity means that the electrostatic repulsion between the MPs and the surface of the porous medium decreases, resulting in unsustainable suspension stability for the MPs. A drop in pH shows that more H^+ probably decreases the surface negative potential of MPs and porous materials, resulting in the MPs to have a higher tendency to remain on the surface of quartz sand, rather than moving through the sand columns (Dong et al., 2018; Zhou et al., 2022).

Fig. 4 exhibited how the XDLVO energy between MPs and quartz sand changed with pH and ion intensity. For instance, at low ionic concentrations, the repulsion energy barrier between MPs and quartz sand rises from 1276 kT to 2101 kT as the solution pH rises from 6 to 9; at pH 6, the repulsion energy barrier between MPs and quartz sand falls from 1276 kT to 608 kT as the ionic concentration rises from 5 mM to 25 mM. Combining the results of XDLVO calculations and deposition experiments, it can be concluded that the decrease in pH and increase in ionic strength reduces the repulsive energy barrier between MPs and the porous medium, making it easier for MPs to be deposited on the quartz sand surface and thus inhibiting the transport of MPs in the quartz sand columns (Table 2).

3.2. Individual transport of MPs in goethite-coated sand column

In the typical pH range of natural water environments, the surface of goethite tends to carry a positive charge. Therefore, the presence of surface charge heterogeneity is a common characteristic shared by all materials coated with goethite, which typically carry a negative charge (Chen et al., 2021; Chen et al., 2019). This charge heterogeneity can influence electrostatic interactions between two types of charged particle surfaces as well as particle transport and aggregate. It has been demonstrated that the electrostatic attraction regulated by the interactions between goethite-coated sand and particles determined the particle deposition (Han et al., 2014; Lin et al., 2011; Wang et al., 2013). Comparing Fig. 1a and c, Fig. 1b and d, it can be concluded that the goethite coating on the sand surface inhibits the MPs transport ability in porous media. Under low ionic strength of 5 mM, the mobility of MPs decreased from 45.9 % to 29.6 % (pH value of 6), from 66.8 % to 31.5 % (pH value of 7), and from 80.3 % to 59.7 % (pH value of 9), respectively; under high ionic strength of 25 mM, the mobility of MPs decreased from 32.8 % to 18.5 % (pH value of 6), from 36.5 % to 18.9 % (pH value of 7), and from 40.4 % to 20.4 % (pH value of 9), respectively. Another reason is that goethite coordinated with water molecules are easily hydrolyzed to form a hydroxylated surface, which

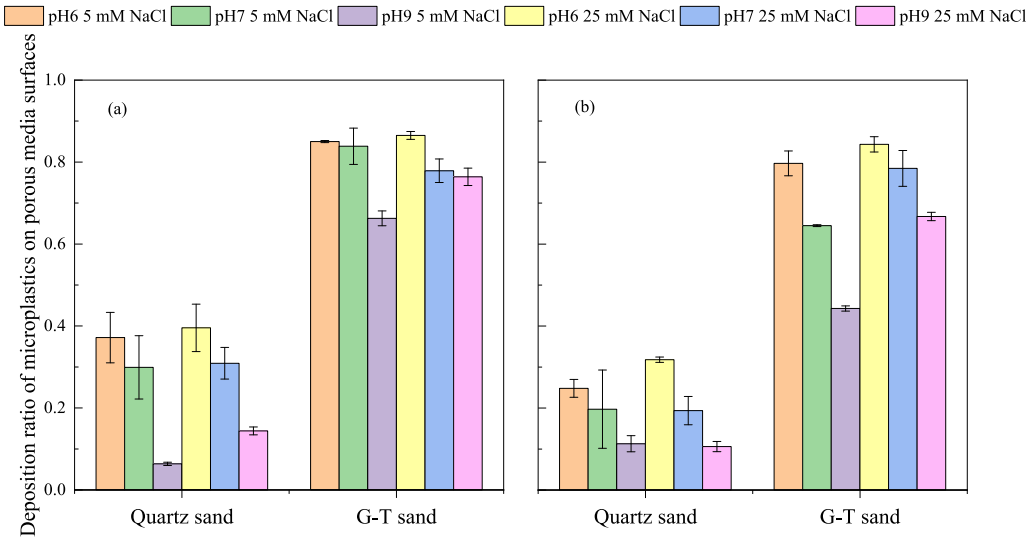

Fig. 3. Deposition ratio of microplastics on the surface of porous media when alone (a) and when coexisting (b) with kaolinite colloids.

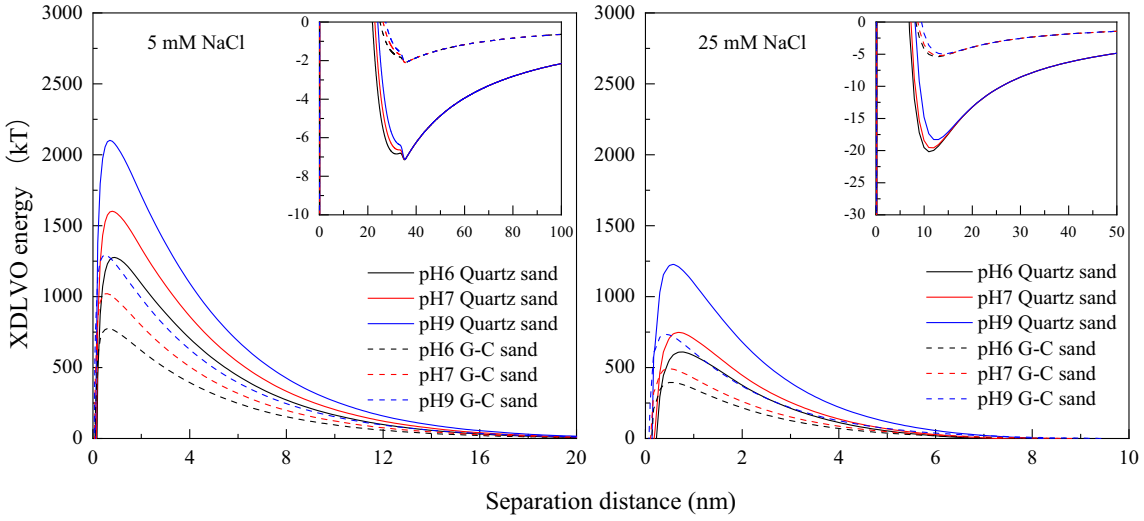

Fig. 4. XDLVO energy spectrum between MPs and porous media at different pH and ionic intensity conditions in quartz sand and G-C sand columns.

exhibits amphoteric oxidation properties and then keep MPs on its surface (Ma et al., 2022). Consequently, the decrease in MPs transport capacity should be mainly attributed to charge heterogeneity, and the effects of pH and ionic intensity on these properties should be responsible for different inhibitory effects. The results of the transport experiments were also corroborated by the deposition experiments of MPs on porous media. At low ionic strength of 5 mM, the deposition ratios of MPs on quartz sand were 32.8 % (pH value of 6), 24.5 % (pH value of 7), and 6.1 % (pH value of 9) respectively, and the deposition ratios of MPs on goethite-coated sand were 84.8 % (pH value of 6), 80.7 % (pH value of 7), and 67.5 % (pH value of 9) respectively; at high ionic strength of 25 mM, MPs deposited on the surface of quartz sand at ratios of 35.5 % (pH value of 6), 28.2 % (pH value of 7) and 13.7 % (pH value of 9), and on the surface of goethite-coated sand at ratios of 86.5 % (pH value of 6), 77.9 % (pH value of 7) and 76.4 % (pH value of 9), respectively. Therefore, MPs are more likely to accumulate on the surface of goethite-coated sand, causing MPs penetration capability to be weaker than in quartz sand.

Table 2
Summary of experimental results of MPs transport through the columns.

Porous medium	Cations	Ionic strength	pH	MPs	Kaolinite	Recovery
		mM		(mg/L)	(mg/L)	(%)
Quartz sand	Na^+	5	6	10	0	45.9
Quartz sand	Na^+	5	7	10	0	66.8
Quartz sand	Na^+	5	9	10	0	80.3
Quartz sand	Na^+	25	6	10	0	32.8
Quartz sand	Na^+	25	7	10	0	36.5
Quartz sand	Na^+	25	9	10	0	40.4
Quartz sand	Na^+	5	6	10	50	53.8
Quartz sand	Na^+	5	7	10	50	70.2
Quartz sand	Na^+	5	9	10	50	85.0
Quartz sand	Na^+	25	6	10	50	36.1
Quartz sand	Na^+	25	7	10	50	39.1
Quartz sand	Na^+	25	9	10	50	53.7
G-C sand	Na^+	5	6	10	0	29.6
G-C sand	Na^+	5	7	10	0	31.5
G-C sand	Na^+	5	9	10	0	59.7
G-C sand	Na^+	25	6	10	0	18.5
G-C sand	Na^+	25	7	10	0	18.9
G-C sand	Na^+	25	9	10	0	20.4
G-C sand	Na^+	5	6	10	50	32.1
G-C sand	Na^+	5	7	10	50	40.0
G-C sand	Na^+	5	9	10	50	65.4
G-C sand	Na^+	25	6	10	50	28.3
G-C sand	Na^+	25	7	10	50	40.4
G-C sand	Na^+	25	9	10	50	55.1

Recovery: mass recovery.
G-C sand: goethite-coated sand.

The XDLVO interaction energy results showed in Fig. 4 that the goethite coating on the quartz sand surface leads to a reduction of its zeta potential electronegativity, which makes the repulsive energy barrier between it and MPs lower. Taking the solution conditions at IS5, pH 6 as an example, the goethite coating caused the repulsion barrier between quartz sand and MPs to decrease from 1275 kT to 774 kT. Furthermore, the secondary energy minimum between goethite-coated sand and MPs was lower than that between quartz sand and MPs under the same hydrochemical conditions. The minimum value of secondary energy between MPs and quartz sand is −7.3 kT, and between MPs and goethite-coated sand is −2.2 kT for the solution conditions of IS5 and pH 6, for example. The results of XDLVO interaction energy calculations indicate that goethite-coated sand reduces the transport ability of MPs and makes MPs more inclined to be retained in the sand column in the form of irreversible deposition (Chen et al., 2021). In contrast, the mobility of MPs in the quartz sand column is higher, and the MPs deposited on the surface of pure quartz sand are more likely to re-enter the pore water and flow out of the sand column under the action of water scouring force (Tufenkji, 2006).

Additionally, the previous research has proven that the surface roughness and specific surface area of porous media also impact the transport capacity of particles, with rougher surfaces providing more deposition sites for particles, narrowing the pores in the sand column, and increasing the physical "straining" effect (clogging of pores by particles or aggregates), thus retarding the transport of MPs and resulting in lower BTCs (Chen et al., 2019; Duster et al., 2017; Liu et al., 2019; Qi et al., 2019; Wang et al., 2017). Therefore, the specific surface area and the surface roughness of quartz sand and goethite-coated sand were measured by BET and X-ray energy dispersive spectroscopy (SEM-EDS), respectively. It was found that the specific surface area of goethite-coated sand was about three times larger than that of quartz sand. The SEM-EDS chemical analysis revealed that the increase in the surface roughness of quartz sand is primarily due to the formation of hydroxy iron oxide on its surface. However, the ratio of d_p/d_c was only about 0.001 (d_p and d_c are the hydrodynamic diameter of the MPs aggregates and the diameter of the sand grains, respectively), which was significantly lower than the threshold value for the occurrence of straining (d_p/d_c = 0.002). Regardless of whether quartz sand or goethite-coated sand is used as the porous medium, the pore size is not small enough to cause blockage by MPs, and the physical "straining" effect of colloids cannot take place in the present experimental system (Lu et al., 2021b).

3.3. Co-transport of MPs and kaolinite in quartz sand columns

It has been discovered that kaolinite is crucial for the movement of MPs through porous media [21, 54]. Ye et al. discovered that kaolinite has a pronounced inhibitory effect on MPs' mobility when mixed with quartz sand and packed into the column; the rough surfaces and deposition sites provided by the positively charged edges of kaolinite should be responsible for its inhibition of MPs transport (Ye et al., 2022). If kaolinite colloidal particles are served as accompanying transport substances for MPs, vigorous hetero-aggregation between MPs and kaolinite particles could be observed (Vu et al., 2022). In other words, kaolinite particles may obstruct the transport of MPs in porous media when they co-present in suspension, which is mainly attributed to the surface charge properties of kaolinite in aqueous solution. In fact, more extensive research has revealed that the kaolinite-induced effects on the transport of MPs in porous media are ultimately determined by the charge properties of the MPs surface (Li et al., 2020). In the presence of kaolinite particles, the transport of negatively charged MPs would increase, while the transport of positively charged MPs would be negligible as ionic intensity increased (Zhou et al., 2022). Previous studies have primarily focused on the impact of kaolinite colloids on the transport of MPs through heterogeneous coalescence, while overlooking the role of colloid competition for deposition sites. While these studies provide valuable insights, they do not fully capture the complex interplay between colloids and MPs in natural systems. A more comprehensive understanding of the transport behavior of MPs requires a more detailed approach. Obviously, the study includes a range of environmental conditions that more closely resemble natural systems, providing a more realistic assessment of the fate and transport of MPs in the environment. To figure out the impact mechanism of MPs' transport in complex environments, it is necessary to analyze the changes in transport characteristics as the environment transitions from simple to complex. To achieve this, the BTCs for MPs transport alone and co-transport with kaolinite under the same hydrochemical conditions were compared. By comparing Figs. 1 and 5, it is clear that the presence of kaolinite colloids slightly enhances the transport of MPs in quartz sand in a wide range of hydrochemical environments. For example, under low ionic strength of 5 mM, the mobility of MPs increased from 45.9 % to 53.8 % (pH value of 6), from 66.8 % to 70.2 % (pH value of 7), and from 80.3 % to 85.0 % (pH value of 9), respectively; under high ionic strength of 25 mM, the mobility of MPs increased from 32.8 % to 36.1 % (pH value of 6), from 36.5 % to 39.1 % (pH value of 7), and from 40.4 % to 53.7 % (pH value of 9), respectively.

The coexisting kaolinite colloids in suspension may alter the transport and deposition behavior of MPs in porous media through multiple mechanisms of interaction (Chen et al., 2012; He et al., 2021; He et al., 2020; He et al., 2018; Li et al., 2019). First, the interaction between MPs and kaolinite colloids changes the size of MPs, which affects the transport and deposition behavior of MPs in quartz sand. The SEM and TEM images of MPs and kaolinite colloids (Fig. S5, S6) showed that the smaller kaolinite colloids have been adsorbed onto the surface of MPs and form MPs-kaolinite heteropolymers. The multimode size distribution (MSD) curves of MPs and MPs with kaolinite colloids are shown in Fig. S9. For MPs, the MSD curves followed positively skewed distribution. In contrast, the MSD curves for the MPs and kaolinite colloids show two peaks, with the larger peak representing the particle size distribution of the MPs-kaolinite colloid heterogeneous aggregates. Obviously, the particle size of MPs-kaolinite heterogeneous aggregates is much larger than that of MPs, causing the former to be more easily clogged in the pores of porous media (Chowdhury et al., 2012; Legg et al., 2014; Yang et al., 2022). For polydisperse system, the MSD curves usually present a more objective interpretation of the heterogenous sample (Xu et al., 2020). In addition, the results of the aggregation and settlement experiments show that the coexistence of kaolinite colloids does not destabilise the intercolloids in the suspension (Fig. 2a, S8). Obviously, the effect of coexisting kaolinite colloids on the transport behavior of MPs in quartz sand cannot be explained by considering only the changes in hydrated particle sizes of MPs.

As was already mentioned, one more significant factor influencing the transport of MPs in porous media is their surface charge characteristics. The zeta potentials of MPs and MPs-kaolinite heteropolymers in suspension were thus measured under varying hydrochemical conditions, which are listed in Table 1. It can be seen in Table 1 that presence of the kaolinite colloids greatly increases the electronegativity of the surface of MPs-kaolinite hetero-polymers. Comparing the XDLVO interaction energies of MPs in the individual transport system and the cotransport system (Figs. 4 and 6), it is evident that the repulsion energy barrier in the MPs-kaolinite cotransport system is greater than that in the individual MPs transport system. Consequently, the strong electrostatic repulsion between hetero-aggregates and quartz sand makes it difficult for them to deposit in a sand column (Dong et al., 2019; Zhang et al., 2021b; Zhao et al., 2021).

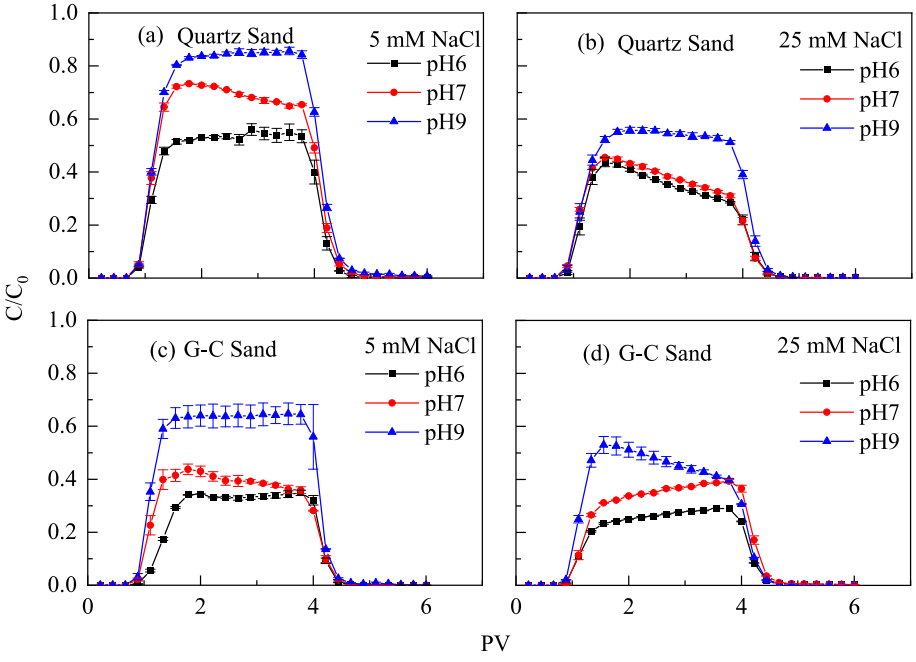

Fig. 5. The BTCs of MPs transport in porous (a, b) sand and (c, d) G-C sand columns under various pH and ionic strength conditions as kaolinite participates in.

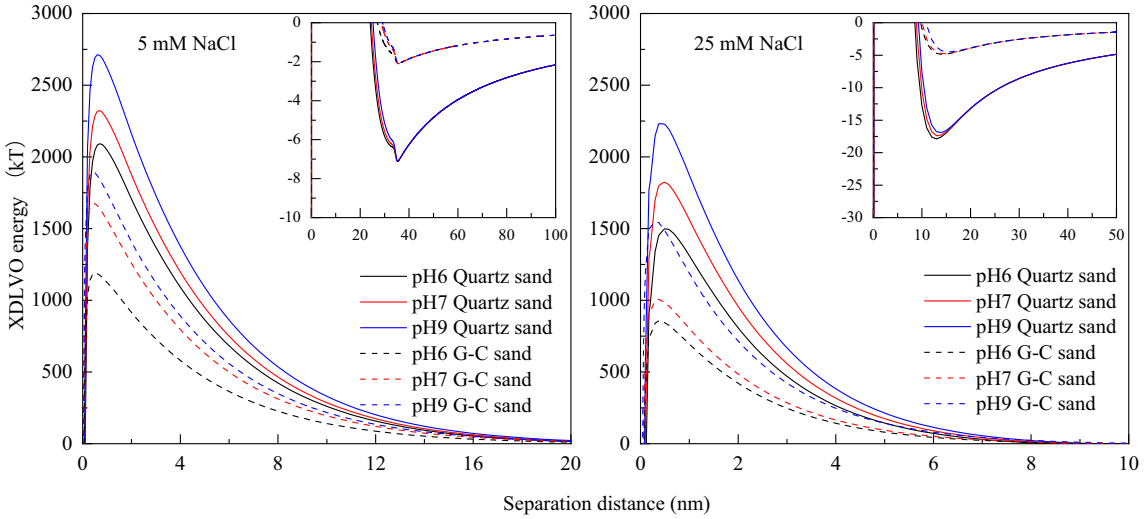

Fig. 6. XDLVO energy spectrum between MPs and porous media at different pH and ionic intensity conditions in quartz sand and G-C sand columns as kaolinite takes part in.

Furthermore, competition for deposition sites is an important factor that can influence solute transport in column experiments. When coexisting materials are present in a solution, they may occupy part of the deposition sites and inhibit colloidal deposition on the porous media surface. This means that some of the particles in the solution will be unable to deposit onto the porous media and will instead remain suspended in the flowing fluid. The extent of this inhibition will depend on factors such as the concentration of the coexisting materials, the surface charge characteristics of the particles, and the flow rate of the fluid (Chen et al., 2019; Li et al., 2020; Liu et al., 2015). To verify whether coexisting kaolinite colloids would compete with MPs for the limited deposition sites on the quartz sand surface, additional porous media deposition site competition experiments were performed under background Na^+ strengths of 5 mM and 25 mM at pH 7. The experimental results showed in Fig. 7 that kaolinite colloid deposited on the surface of quartz sand enhanced the mobility of MPs from 66.8 % and 36.5 % to 72.9 % and 47.5 % under ionic strength 5 mM and ionic strength 25 mM conditions, respectively. Evidently, pre-covering quartz sand with kaolinite colloid can increase the transport of MPs, demonstrating that kaolinite colloid deposited on the surface of quartz sand will occupy particular deposition sites and promote the transport of MPs (Li et al., 2020).

3.4. Cotransport of MPs and kaolinite in goethite-coated sand columns

The above paper has demonstrated that when quartz sand is used as the porous media, the contribution of coexisting kaolinite colloids to the MPs' transport ability was not significant. Intriguingly, the promotion of kaolinite to MPs' transport becomes noticeable and is highly correlated with the background ionic strength if the surface of quartz sand is covered with goethite. As shown in Fig. 5, at an ion strength of 5 mM, the mobility of MPs was elevated by only 2.5 % (pH 6), 8.5 % (pH 7), and 5.7 % (pH 9) compared to their transport alone in Fig. 1; however, at an ion strength of 25 mM, the mobility of MPs was elevated by 9.4 % (pH 6), 21.5 % (pH 7), and 34.7 % (pH 9) compared to their own transport alone. These results suggest that kaolinite colloids are more effective in promoting MPs transport in goethite-coated sand at the relatively higher ionic strength.

When analyzing the transport of MPs in the system, it is important to consider the main mechanisms that may be at play. Specifically, the presence of kaolinite colloids in suspension and goethite coating on the quartz sand should be taken into account. First, enhanced electrostatic repulsion between goethite-coated sand and MPs facilitated the transport of MPs. As analyzed above, the presence of kaolinite colloids in MPs-kaolinite hetero-polymers increases the electronegativity of the surface of the particles. This increased electronegativity results in a stronger electrostatic repulsion between the hetero-polymers and the surrounding goethite-coated sand. The repulsion helps to keep the hetero-polymers suspended in the fluid phase and prevents them from adhering to the surface of the goethite-coated sand.

Second, kaolinite colloids compete with MPs for limited deposition sites on the surface of porous media (Han et al., 2013; Xu et al., 2018). At a relatively higher background ionic strength of 25 mM, the deposition of

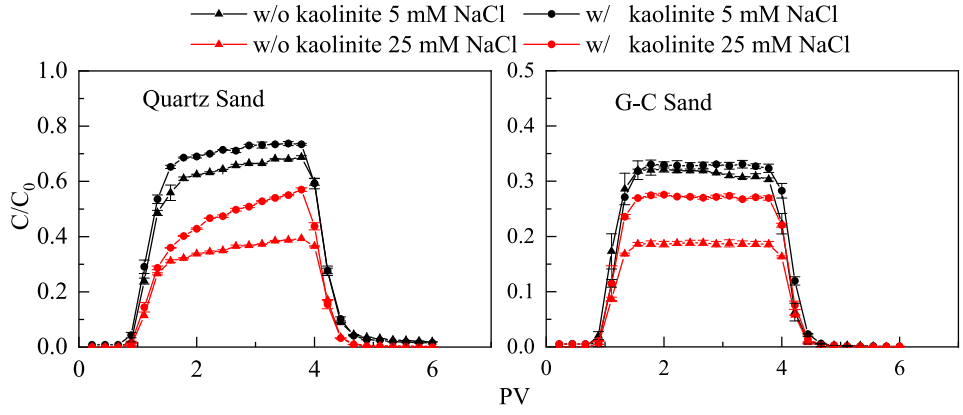

Fig. 7. The BTCs of MPs transport in kaolinite-covered ("w/") and kaolinite-uncovered ("w/o") porous media.

kaolinite colloids led to an increase in the mobility of MPs from 18.9 % to 27.3 %; however, at a relatively lower background ionic strength, this promotion effect was not significant, and the mobility of MPs only increased by 1.5 % (Fig. 7). This indicates that kaolinite colloids compete with MPs for deposition sites on the surface of goethite-coated sand, and that this competitive effect is more pronounced at high background ion intensities. The inhomogeneous goethite coating increases the complexity of the quartz sand surface, making it easier for kaolinite colloids to be deposited (Fig. S10) [25]. An elevated background ionic strength compresses the double electric layer of kaolinite colloids and lowers the repulsive energy barrier, allowing a large number of kaolinite colloids to occupy the limited deposition sites on the surface of the goethite-coated sand, thus inhibiting the deposition of MPs. Deposition experiments have likewise demonstrated that in a wide range of aqueous chemical environments, the deposition ratio of MPs on goethite-coated sand is weakened by the presence of kaolinite colloids (Fig. 3, S10). The experimental results demonstrated that for different porous media, the kaolinite colloids differ significantly in their promotion effect on the transport capacity of MPs. The results of the study further emphasize the contribution of kaolinite colloids deposited in the sand column to the transport of MPs.

4. Conclusion

It was demonstrated in this study that both the increase in ionic strength and the decrease in pH reduce the transport ability of MPs in porous media. Additionally, goethite-coated sand, when used as a porous medium, significantly reduces the transport capacity of MPs as compared to pure quartz sand. It is mainly due to the fact that goethite coating reduces the energy barrier between the porous medium and MPs, and increases the specific surface area and complexity of the porous medium. Coexisting kaolinite colloids facilitate MPs' transport, which is mainly attributed to the increase in electrostatic repulsion between MPs and porous media as well as the competition with MPs for deposition sites caused by kaolinite colloids. There was a significant difference between the contributions of kaolinite colloids to the transport capacity of MPs in different porous media. When quartz sand is utilized as the porous medium, kaolinite colloids are weak in promoting the transport ability of MPs and vary significantly for various hydrochemical conditions when goethite-coated sand is utilized. Kaolinite colloids occupy more deposition sites, thereby enhancing the transport capability of MPs at high background ionic strengths. The individual and co-transport with kaolinite of MPs in pure quartz sand and goethite-coated sand rationalized by XDLVO theory. The findings of this exploration shed insight into the potential and variable environmental behavior of MPs in multicomponent soil and groundwater systems.

CRediT authorship contribution statement

Bokun Chang: Conceptualization; Data curation; Formal analysis; Validation; Roles/Writing - original draft.

Bing He: Formal analysis; Investigation; Methodology.

Gang Cao: Formal analysis; Methodology.

Zhiying Zhou: Data Curation; Investigation.

Xiaoqi Liu: Resources; Investigation.

Yajun Yang: Investigation; Validation.

Chenyang Xu: Project administration.

Feinan Hu: Investigation; Validation.

Jialong Lv: Funding acquisition; Resources; Project administration; Supervision; Writing - review & editing.

Wei Du: Funding acquisition; Resources; Supervision; Writing - review & editing.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.163832.

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