

# Effects of colloidal humic acid on the transport of sulfa antibiotics through a saturated porous medium under different hydrochemical conditions

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## ABSTRACT

Colloidal humic acid (HA) acts as a vector that can facilitate the transport of contaminants in groundwater. However, investigations of factors that enhance the transport of sulfa antibiotics when there are colloids present remain incomplete to date. In this study, column experiments were performed under different conditions (particle size, pH, ionic strength, cation valence, colloidal concentration) using 0.25 mg/L sulfamerazine (SM) with or without colloids. The results showed that antibiotics were more easily deposited on the surface of porous media with a diameter of 0.22 mm than 0.45 mm. As the pH increased from 6 to 8, adding colloidal HA increased the maximum breakthrough concentration from 0.94 to 1 for SM. Adding colloidal HA at different NaCl concentrations decreased the maximum  $C/C_0$  ratio from 0.97 to 0.92. However, adding colloidal HA changed the  $C/C_0$  ratio more when the divalent cation ( $\text{Ca}^{2+}$ ) was present. Overall, increasing the colloidal HA concentration clearly caused the effluent sulfamerazine concentration to increase.

**Key words** | adsorption, colloidal humic acid, sulfa antibiotics, transport

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## INTRODUCTION

Antibiotics are widely used in livestock husbandry and fertilization with manure, which has led to the migration of large amounts of veterinary antibiotics through the soil and into groundwater (Sukul & Spiteller 2006). Sulfa antibiotics are a major class of antibiotics that have been found to pose potential threats to the health of humans and other biota in the environment (Yang *et al.* 2011). Accordingly, it is important to gain a thorough understanding of the transport behaviors of sulfa antibiotics in porous media.

Early studies assumed that sulfa antibiotics in groundwater would partition strongly to the stationary phase and be transported slowly in subsurface water. As a result, it was concluded that there was little risk of sites distant from a source becoming contaminated with sulfa antibiotics (Grolimund *et al.* 1996). However, colloids with large specific surface areas and high numbers of reactive surface functional groups have recently been found to be stable in

the mobile phase of groundwater and capable of being transported over long distances via groundwater flow (Zhou *et al.* 2016). Colloidal HA can act as a carrier for pollutants, facilitating the transport of pollutants in the groundwater (Zhu *et al.* 2014), which could allow potential environmental problems caused by pollutants to be enhanced when they are adsorbed onto the colloids and transported in groundwater. Therefore, failure to consider colloid-facilitated sulfa antibiotic transport will lead to underestimation of the transport velocities and distances of sulfa antibiotics (Knappenberger *et al.* 2014). Understanding antibiotic adsorption on natural colloids is essential for prediction of the behavior and toxicity of antibiotics in natural groundwater.

It has been shown theoretically that colloidal particles can facilitate the transport of contaminants such as metals (Grolimund *et al.* 1996), radionuclides (Severino *et al.* 2007), and viruses (Syngouna & Chrysikopoulos 2013)

through porous media, but there has been little attention given to colloids facilitating the transport of sulfa antibiotics. In this study, column experiments packed with glass beads were performed to determine the potential for colloidal HA to increase the degree to which sulfa antibiotics (SM) are transported in groundwater. The experiments were performed using different particle sizes (0.45 mm and 0.22 mm), pH values (6, 7, and 8), solution ionic strengths adjusted by  $\text{Na}^+$  and  $\text{Ca}^{2+}$  (0, 0.02, and 0.05 M), colloidal HA concentrations (0, 5, 10, and 20 mg/L), and flow rates (0.1, 0.2, 0.4 ml/min) to assess the influences of different conditions on colloid transport and deposition in the porous medium. The results of each column experiment were assessed by plotting a breakthrough curve (BTC), in which the antibiotic concentration in the effluent ( $C$ ) to the initial antibiotic concentration ( $C_0$ ) ratio was plotted against the pore volume in the system.

## MATERIALS AND METHODS

### Experimental materials

#### Porous medium

Column experiments saturated with glass beads that had a net negative charge and were sieved to provide an average diameter of 0.45 mm or 0.22 mm were used as a model medium. This medium excluded the uncertainties caused by heterogeneity and the properties of different interfaces in real soil and minimized the release of colloidal material. The glass beads used in the column experiments have been described in detail elsewhere (Liu *et al.* 2016).

#### Colloidal HA

Humic acid, which is found widely in aquifers, strongly affects the transport of sulfa antibiotics (Zhu *et al.* 2014). In this study, Suwannee River humic acid (SRHA) standard II purchased from the International Humic Substance Society (CO, USA) was used as the colloid in the experiments.

Based on a total organic carbon concentration in groundwater of about 5 mg/L and the chemical composition

of HA, the concentration of colloidal HA suspension was diluted to 10 mg/L. In addition, the colloidal HA suspension was diluted to 5, 10, and 20 mg/L to study the effects of HA concentration on the adsorption and transport of antibiotics. The suspensions were stored in the dark at 4°C and re-sonicated before use.

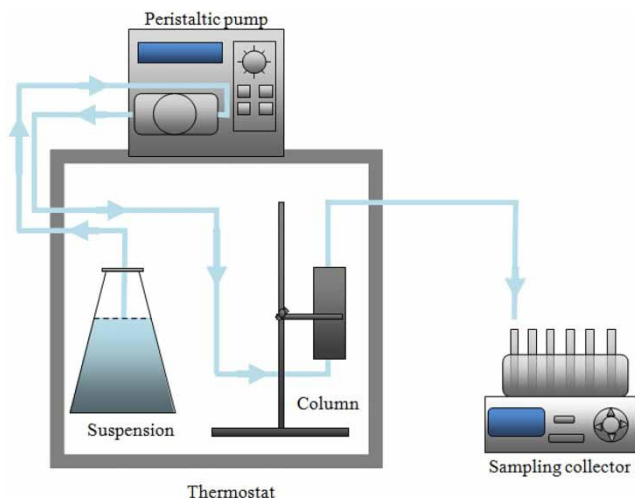
#### Sulfa antibiotics

Sulfamerazine (SM) purchased from Sigma-Aldrich (St Louis, MO, USA) was chosen because it is commonly used in veterinary medicine. SM has been found in groundwater at concentrations ranging from nanograms per litre to a few milligrams per litre (Heberer *et al.* 1998), with residual levels of 17.3 ng/L in Spain and 54 ng/L in the United States, respectively (Bartelt-Hunt *et al.* 2011; García-Galán *et al.* 2011). The method used to determine the sulfa antibiotic concentrations was linear over the range 0.005–10 mg/L, subject to accuracy and instrumental error, so an antibiotic concentration of 0.25 mg/L was used in the experiments. The prepared suspensions were stored at 10°C prior to use to simulate subsurface conditions.

#### Column transport experiments

The transport experiments performed in the columns each had an effective length of 6.0 cm and an internal diameter of 3.2 cm. Glass beads were added to each column to act as the porous medium and a filter membrane was retained at each end of the column to prevent the glass beads from being lost. In addition, a peristaltic pump (BT-100; Longer-Pump, Baoding, China) was used to control the flow rate of the suspension that was passed through the column. The effluent was collected using an automatic fraction collector at fixed time intervals, and the sulfa antibiotic concentrations in the effluent samples were determined. The column and the solution were kept in a refrigerator at 10°C to simulate the typical temperature of groundwater (Figure 1).

The column was slowly saturated by deionized water from the bottom of the column for 24 h until the pH of the effluent stabilized. The column experiments were conducted in two stages. First, the influent suspension (4 PV) was injected into the column during the adsorption stage



**Figure 1** | Schematic of the experimental system.

(stage 1), after which the input was switched to deionized water (3 PV) during the desorption stage (stage 2) until no antibiotics were detected in the effluent.

### Effluent analysis

Although sulfa antibiotics are found only at trace concentrations in groundwater, they pose great hazards to humans and other biota. The sulfa antibiotic concentrations in each

aliquot were measured using a solid-phase microextraction system and a high-performance liquid chromatograph (LC1200; Agilent Technologies, Santa Clara, CA, USA). An Agilent XDB C<sub>18</sub> chromatographic column (150 mm × 46 mm, 5 μm) was used with a mobile phase of 0.1% formic acid–methyl alcohol, a detection wavelength of 270 nm and a temperature of 30°C (García-Galán *et al.* 2011; Aixia *et al.* 2014).

## RESULTS AND DISCUSSION

### Characterization of the suspension

The zeta potentials and particle sizes were measured using a Zetasizer (Malvern Zetasizer, Nano ZS, Malvern, UK) (Table 1).

### Zeta potential of the suspension

The zeta potentials of the suspensions under different conditions ranged from −6.74 to −29.3 mV. The pH strongly affected the zeta potential, which was −18.7 mV when the pH was 6 and decreased to −28.8 as the pH increased to 8 when IS was 0. This could have been because more HA dissociated as the pH increased, resulting in fewer

**Table 1** | Characterization of the colloidal HA and sulfa antibiotic suspensions

pH	IS (M)	Colloidal HA (mg/L)	Pdl	Pk1 Mean diameter (nm)	Pk2 Mean diameter (nm)	Pk3 Mean diameter (nm)	Pk1 Area (%)	Pk2 Area (%)	Pk3 Area (%)	ZP (mV) (±0.8–2.5)
6	0.00	10	0.756	233.0	0	0	100	0	0	−18.7
7	0.00	10	1.000	316.0	0	0	100	0	0	−24.1
8	0.00	10	0.756	656.9	0	0	100	0	0	−28.8
7	0.00(NaCl)	10	1.000	316.0	0	0	100	0	0	−24.1
7	0.02(NaCl)	10	0.689	389.1	100	0	100	0	0	−22.0
7	0.05(NaCl)	10	1.000	438.6	87.56	5,560	85.2	10.6	4.2	−16.8
7	0.00(CaCl <sub>2</sub> )	10	1.000	316.0	0	0	100	0	0	−24.1
7	0.02(CaCl <sub>2</sub> )	10	0.928	845.8	0	0	100	0	0	−9.6
7	0.05(CaCl <sub>2</sub> )	10	0.257	1,490	0	0	100	0	0	−6.74
7	0.00	5	0.630	290.7	0	0	100	0	0	−21.9
7	0.00	10	1.000	316.0	0	0	100	0	0	−24.1
7	0.00	20	0.853	819.3	0	0	100	0	0	−29.3

Note: IS is the ionic strength, Pdl is the polydispersity index, Pk Mean and Pk Area are the colloid size and the contribution of the colloid to the total colloid load for up to three peaks, respectively, and ZP is the zeta potential.

negatively charged groups (such as phenolic and carboxylic groups). The carboxylic groups in HA will be dissociated under acidic and neutral conditions, but both carboxylic and phenolic groups will be dissociated under alkaline conditions. Therefore, the surface will become more negatively charged as the pH increases (Liu *et al.* 2016). However, the zeta potential increased as the ionic strength increased from 0 M to 0.05 M at pH 7. We concluded that compression of the electrostatic double layer at high ionic strength caused the electrostatic repulsion between particles to decrease in strength, allowing more aggregation to occur under unsteady conditions (Torkzaban *et al.* 2008). As a result, zeta potential decreased from  $-21.9$  to  $-29.3$  mV as the colloidal HA concentration increased.

### Size of suspended particles

As shown in Figure 2, most of the suspended particles were small (about 233 nm in diameter) under acidic conditions, but they were about 316 and 656.9 nm at pH 7 and 8, respectively. This was likely because increasing the pH caused more functional groups to dissociate, resulting in increased particle size. The particle size increased from 316 to 438.6 nm when the IS increased from 0 to 0.05 M in response to the addition of NaCl. These findings indicated that aggregates formed through weak electrostatic interactions (such as hydrogen bonds and metal bridges) when the ionic strength was high. However, when there were divalent cations ( $\text{Ca}^{2+}$ ), more negative charges were neutralized and particle size increased from 316 to 1,490 nm. In addition, the particle sizes were 290.7 and 316 nm when the colloid concentrations were 5 and 10 mg/L, respectively; however, the particles increased in size and multiple scattering occurred when the colloid concentration was 20 mg/L. Pranzas *et al.* (2003) suggested that colloidal HA takes a chain-like conformation and becomes relatively unstable at high colloidal HA concentrations.

### Column transport experiments

The colloidal HA facilitated the transport of sulfa antibiotics to different degrees under different hydrochemistries. Column experiments with or without colloidal HA were performed in each test (Table 2). For T9 and T10, with the

exception of the colloid concentrations, the experimental conditions were the same as for T1.

### Colloid-facilitated antibiotic transport at different particle sizes

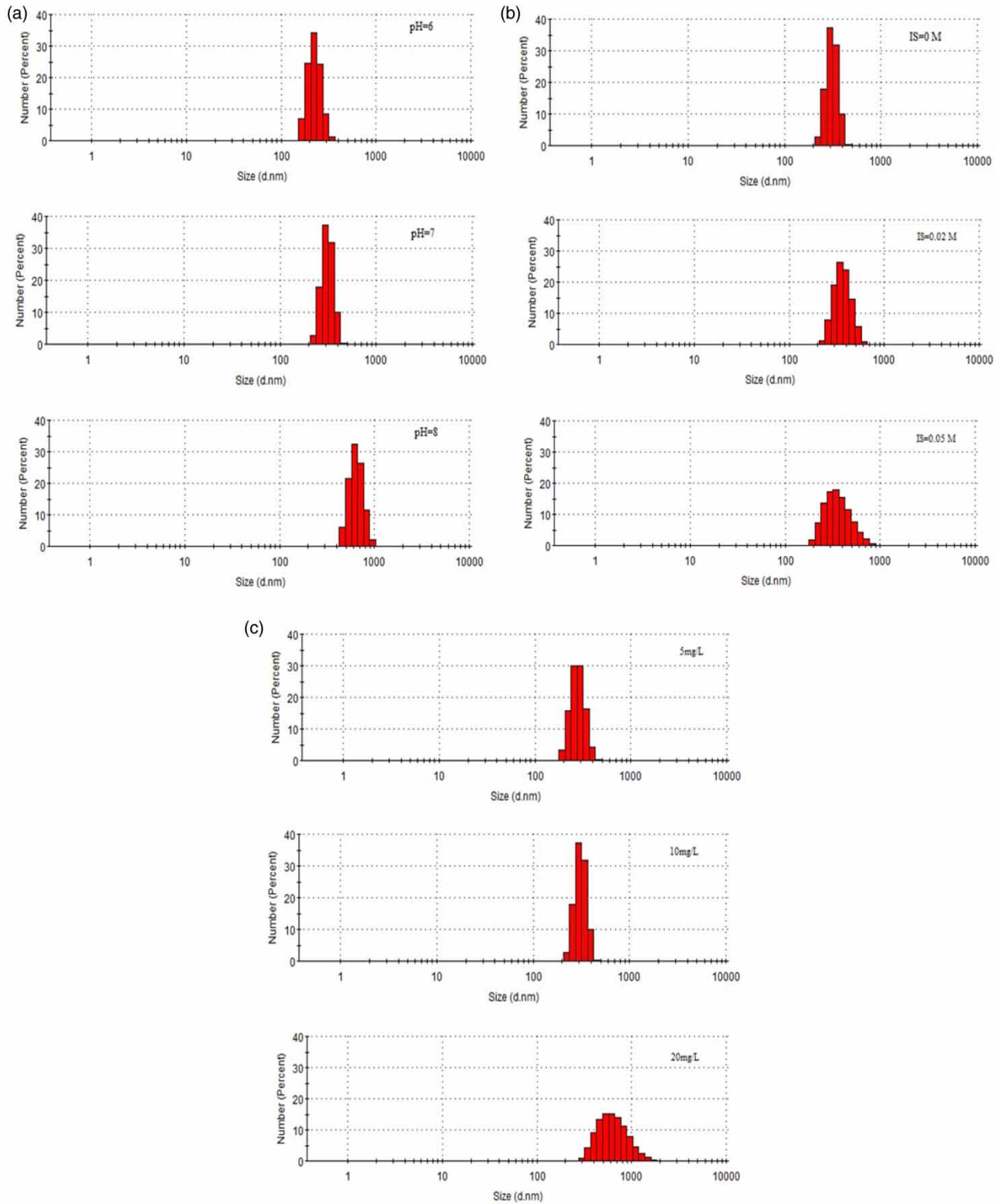
The effects of glass bead sizes on the transport of suspension were identified in column experiments (Figure 3(a)). When the diameter was 0.45 mm and no colloids were present, SM adsorbed on the media in the column showed 91% breakthrough. When the medium contained smaller-sized particles (T2), less antibiotic was transported through the column in the absence of colloid, with only 72% SM breakthrough occurring. These differences in behavior because of smaller particle size were expected because of the higher specific surface area and more effective adsorption sites for antibiotics.

Significant facilitated transport has been observed in the presence of colloids, resulting in more antibiotics in the effluent. When the pore size of the medium was 0.45 mm, the peak  $C/C_0$  increased by 6% for both SM. However, when the diameter of the porous medium decreased to 0.22 cm, the peak  $C/C_0$  increased by 15% for SM. Moreover, the deposition rate coefficient of antibiotics increased with decreasing pore size in the presence of colloids, which is in good agreement with previous studies that showed colloid deposition is affected by the size of solid collectors (Wang *et al.* 2016). Overall, these findings show that antibiotics in effluent increase as a result of colloidal HA.

### Colloid-facilitated antibiotic transport at different pH

More antibiotics were transported at pH 7 than at pH 6 or 8 (Figure 3(b) and 3(c)). The relative mass of the antibiotics recovered in stage 1 was about 78% at pH 7, and the maximum  $C/C_0$  ratio was 0.91. The relative mass recovered in stage 1 was between 2% and 8% lower at pH 6 and 8 than at pH 7, and the maximum  $C/C_0$  ratio was significantly lower at pH 6 and 8 than at pH 7.

The presence of colloidal HA increased the degree of antibiotic transport, and the SM breakthrough concentration at lower pH was lower than at higher pH, the  $C/C_0$  ratios being higher than 0.94 and close to 1. The SM broke through the column when colloids were present, and the relative mass recovered in stage 1 increased by 5% or more (and less SM



**Figure 2** | Size distributions of the suspension at (a) different pH values (IS = 0), (b) different ionic strengths (pH = 7), (c) different colloidal HA concentrations (pH = 7 and IS = 0).

**Table 2** | Experimental conditions for column experiments

Influence factors	Test no.	Size (mm)	pH	IS (mol/L)
Particle size	T1	0.45	7	0.00
	T2	0.22	7	0.00
pH	T3	0.45	6	0.00
	T1	0.45	7	0.00
	T4	0.45	8	0.00
IS (NaCl)	T1	0.45	7	0.00
	T5	0.45	7	0.02
	T6	0.45	7	0.05
IS (CaCl <sub>2</sub> )	T1	0.45	7	0.00
	T7	0.45	7	0.02
	T8	0.45	7	0.05

Influence factors	Test no.	Concentration of HA (mg/L)
Concentration of colloid	T1	0
	T9	5
	T1	10
	T10	20

was retained) when colloidal HA was present. These changes were attributed to the effects of surface charge on colloid deposition efficiency. The surface charge is a function of suspension pH, with the negative zeta potential of the suspension being  $-18.7$  mV when the pH was 6 and becoming increasingly negative as the pH increased in the presence of colloids. As a result, the repulsion between particles facilitated SM movement.

Colloidal HA serves as a carrier and facilitates the transport of antibiotics. There are two other mechanisms through which colloidal HA could facilitate the transport of SM at different pH values. The first involves steric hindrance. Specifically, increasing the pH will decrease the number of sites on the collector surface at which antibiotics could be adsorbed, so more SM molecules will be transported through the column. The other mechanism involves the effects of pH on transport of the colloidal HA itself (Akbour *et al.* 2013). More colloidal HA will be transported at higher pH values because less colloidal HA will become deposited on the solid collector (Tombácz 1999).

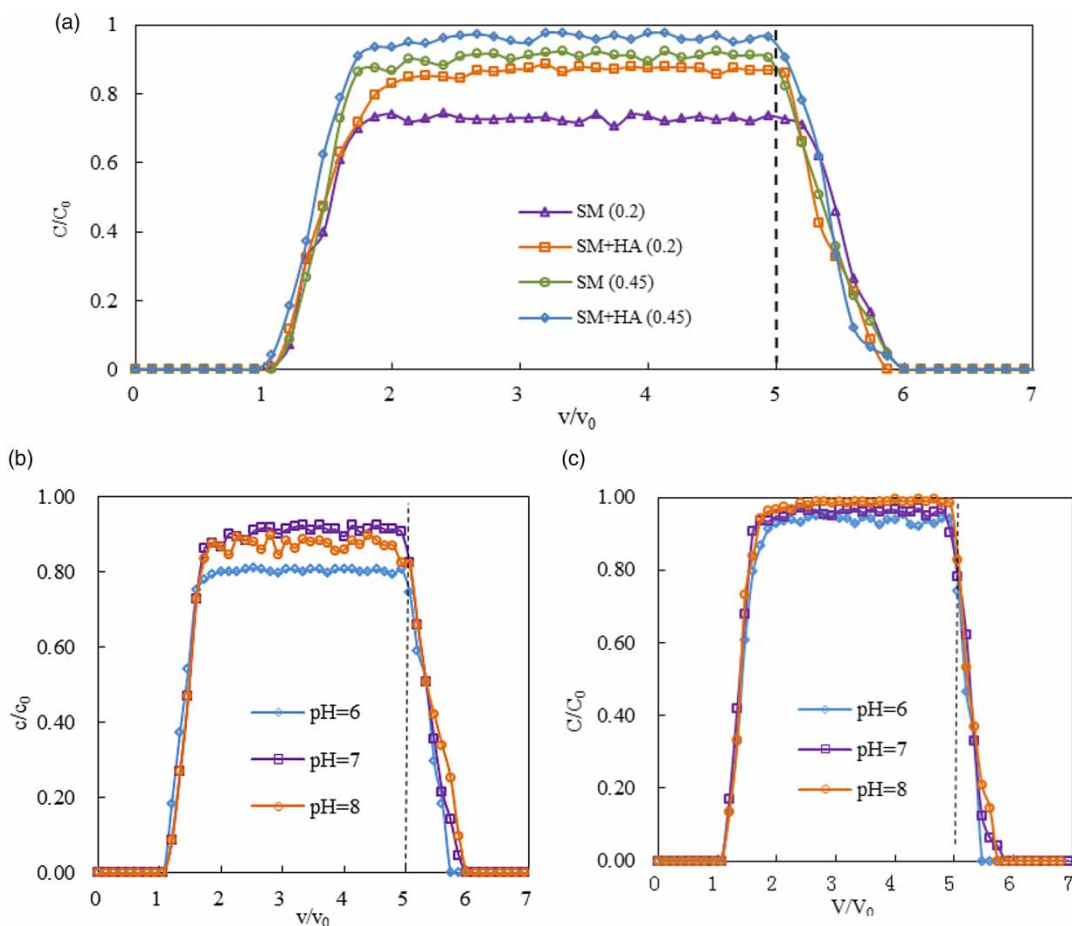
### Colloid-facilitated antibiotic transport under different electrical conditions

As shown in Figure 3(d) and 3(e), 77.9% of the applied masses of SM were recovered in the effluent during stage

1 when in the absence of colloids and electrolytes. The stable maximum  $C/C_0$  ratio was 0.91, and the degree to which the SM was transported in the absence of colloidal HA but in the presence of an electrolyte decreased as the ionic strength increased. Smaller amounts of SM broke through the porous medium and the relative mass recovered in the effluent was lower during the SM addition process. In addition, the maximum  $C/C_0$  ratio was lower, and more SM was retained by the column. Moreover, the presence of colloidal HA promoted SM transport, but the maximum  $C/C_0$  ratio was lower when the ionic strength was high than when it was low. Based on these findings, we concluded that colloidal HA facilitated the transport of SM. Moreover, the results observed when the ionic strength increased indicated that the risks associated with SM may be higher when groundwater has a low ionic strength than a high ionic strength.

These findings suggest that transport of SM was significantly facilitated by the presence of colloids. This is because increasing the ionic strength would increase the likelihood of ions in solution undergoing ion exchange, increasing competition between ions and the antibiotics. As a result, the amounts of antibiotics in the effluent would decrease and more would be retained by the column (Guo 2014). The presence of colloidal HA caused the  $C/C_0$  ratio to decrease as the ionic strength increased. The ionic strength likely affected the surface structures and properties of the colloidal HA, causing the electrical double layer thickness to decrease as the ionic strength increased. This would have led to less electrostatic repulsion between the HA and collectors, allowing the colloidal HA to be deposited on the surfaces of the collectors and decreasing the amounts of antibiotics that were present in the effluent because of facilitation by the colloidal HA. The deposited colloidal HA would then effectively act as adsorption sites, decreasing facilitation of antibiotic transport because they could more easily be deposited on the collectors.

Less facilitation of transport by colloidal HA occurred in the presence of  $\text{Ca}^{2+}$  than  $\text{Na}^+$  (Figure 3(f) and 3(g)).  $\text{Ca}^{2+}$  could simultaneously bind with colloidal HA and SM, decreasing the stability of the colloid and SM suspension. A molecular sieving effect may also have occurred because the pores would have been narrower than the diameter of an antibiotic molecule bound to  $\text{Ca}^{2+}$  (Xia *et al.* 2013).



**Figure 3** | Breakthrough curves for antibiotics eluted in the absence and presence of colloidal HA under different conditions: (a) particle size, (b) SM at different pH, (c) SM + HA at different pH, (d) SM (NaCl), (e) SM + HA (NaCl), (f) SM (CaCl<sub>2</sub>), (g) SM + HA (CaCl<sub>2</sub>), (h) SM at different colloid concentrations. (Continued.)

### Colloid-facilitated antibiotic transport at different colloidal HA concentrations

The SM concentration in the effluent in the presence of colloids was significantly increased relative to the SM solution without colloids because of association with mobile colloids (Figure 3(h)). Under different colloidal concentrations, the transport velocity of SM was similar, and maximum concentration was reached approximately 2.1 PV after the particles were introduced. When there was no colloid, 13.3% was retained, while 88.6% of SM was recovered in the effluent when the colloidal HA concentration was 5 mg/L, and the maximum  $C/C_0$  ratio was 0.96. Increasing the colloidal HA concentration to 10 mg/L increased the relative mass recovered in stage 1

to 88.4%, while the peak concentration became close to 1. Increasing the colloidal HA concentration to 20 mg/L did not obviously increase the relative mass recovered in stage 1 because 20 mg/L was higher than the critical colloidal HA concentration at which facilitation ceased to be promoted further. The deposition decreased, and more antibiotics were found in the effluent when the colloidal concentration increased. The mass retention generally decreased as the colloidal concentration increased. Taken together, these findings indicate that colloidal HA could facilitate the transport of SM, and that increasing the colloidal HA concentration increased the amount of facilitation that occurred. This likely occurred because the ability of colloidal HA to be transported was limited, and some of the HA was deposited.

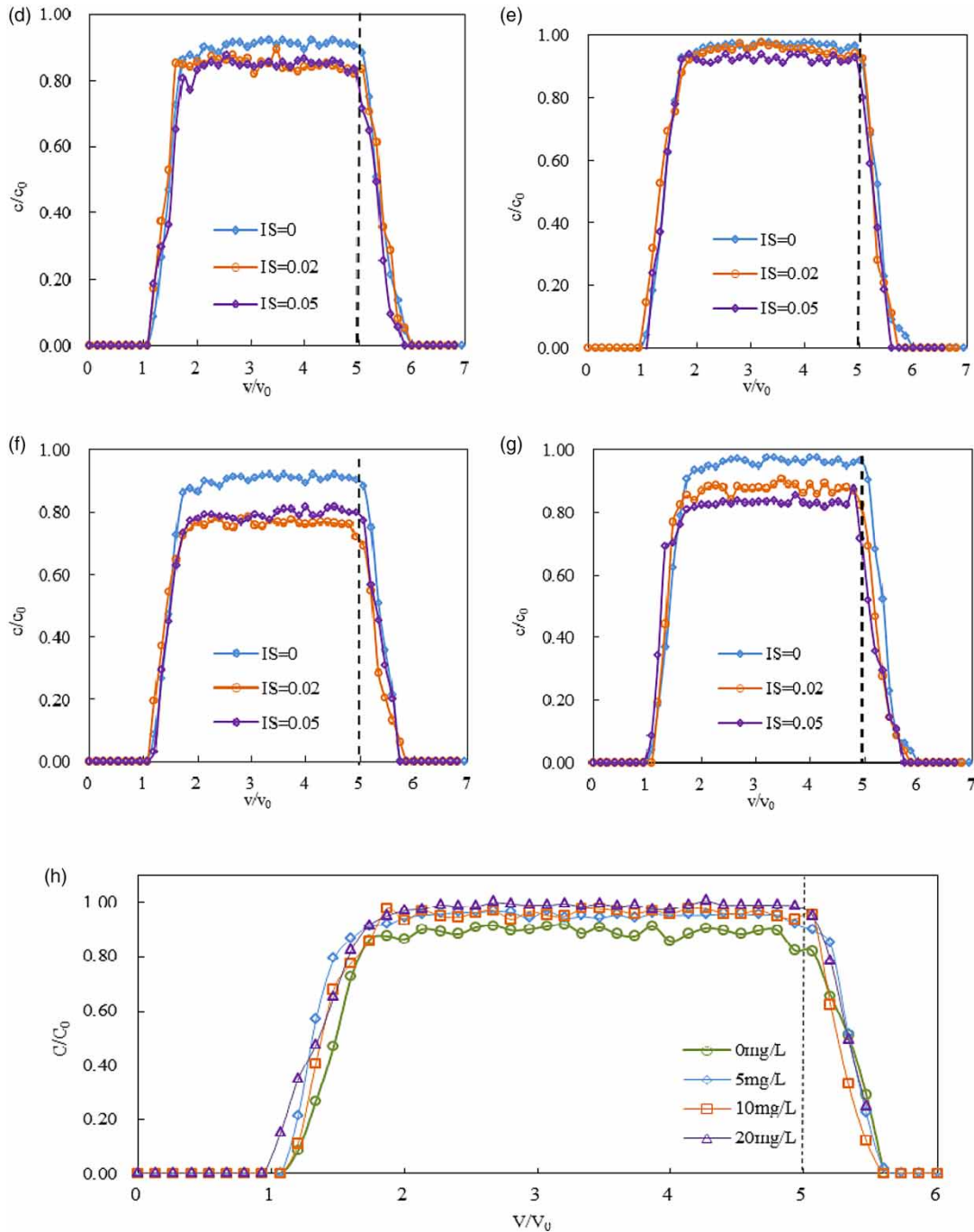


Figure 3 | Continued.

## CONCLUSIONS

The column experiments showed that the transport of sulfa antibiotics in a porous medium can be facilitated by colloidal HA.

This means that the presence of colloids in groundwater can increase the risks posed by SM in the groundwater to humans and other biota. The breakthrough curves clearly demonstrated that the mobility of the antibiotics–colloid suspension was



highly dependent on the pore size of the medium. The pH of the suspension was one of the important factors influencing the transport of SM. Because the zeta potentials of the suspension and porous medium were all negative, SM associated with colloids was excluded from the glass beads by electrostatic repulsive forces. The colloid-facilitated transport was higher at low IS because the deposition efficiency increased with ionic strength as a result of the thickness of the double layer decreasing. Increasing the colloidal HA concentration from 0 to 10 mg/L increased the peak  $C/C_0$  of the antibiotics that were transported by 60%; however, further increasing the colloidal HA concentration did not increase the amounts of antibiotics transported. Slow flow rates increased the reactive time of antibiotics in the column, which increased the deposition of the antibiotics and reduced their levels in the effluent.

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First received 19 October 2017; accepted in revised form 5 February 2018. Available online 20 February 2018