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## Co-transport of Pb (II) and Cd (II) in saturated porous media: effects of colloids, flow rate and grain size

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### ABSTRACT

The transport of Pb(II) or Cd(II) in subsurface has been studied in the literature; however, their co-transport in porous media in presence of colloids has not been clearly understood. In this work, a series of column experiments were conducted to study Pb(II) and Cd(II) co-transport in saturated porous media under various experimental conditions with different combination of colloidal solution (montmorillonite colloid, manual loessial soil colloid and humic acid), flow rate (0.1, 0.5 and 1.0 ml min<sup>-1</sup>) and sand grain size (0.4–0.8mm and 0.8–2.0mm). The results showed that increase of flow rate and grain size promoted the mobility of Pb(II) and Cd(II), furthermore, the presences of mobile colloids also enhanced the mobility of the two heavy metals, meanwhile, Cd(II) showed higher mobility than Pb(II) in the columns. Findings from this work enhanced current understanding of the competitive transport and colloid-facilitated transport of heavy metals in saturated porous media.

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flow rate; sand grain size

## 1. Introduction

Lead (Pb) and cadmium (Cd) ions are toxic heavy metals in the environment, and anthropogenic activities are their main contaminants sources. Because Pb (II) and Cd(II) have huge impact on the environment, their potential risks to the soil and groundwater systems should be concerned. The transport of heavy metals through the soils can seriously pollute the groundwater and drinking water aquifers [1]. It is thus necessary to advance current understanding of Pb(II) and Cd(II) transport behaviors in porous media for the planning of rational pollution control strategies [2].

Soil colloids are particles with effective diameters less than 10 µm and serve as a critical reactive component. Soil colloids play an important role in facilitating Pb(II) transport in soils [3]. Colloids such as clay minerals and humic acid (HA) can accelerate the transport of heavy metals in soils [4–6]. A variety of inorganic and organic colloids exist in natural soils. The high surface area of colloid particles make them capable to adsorb various organic and inorganic contaminants including heavy metals [7]. The colloids thus can act as mobile media to advance the transport of sorbed heavy metals in soils [3]. For example, colloidal montmorillonite can govern Pb(II) and Cd(II) transport because of large specific surface area, high cation exchange capacity and reaction amphoteric hydroxyl groups [8]. Humic acid (HA) displays both macromolecular and colloidal characteristics [9]. The

presence of carboxylic and phenolic groups result in HA carries negative charges predominantly, it thus has a high affinity for metallic cations in aqueous solutions [10]. Manual loessial soil colloids contain minerals, organic matter and other substances and thus exhibit the properties of inorganic and organic colloids [11].

Previous studies have reported that mobile colloids can facilitate heavy metals transport from soils to groundwater and the presence of montmorillonite and HA have significant impact on groundwater Pb (II) pollution [12,13]. HA and inorganic soil components can strongly bind Pb(II) contaminants, and HA and montmorillonite colloids thus are important sinks for heavy metals in soils [14,15]. Previous studies have shown the relationship between a single heavy metal and colloids, but there are less works focused on the co-transport of multiple heavy metals with soil colloids in porous media. It is therefore critical to conduct a systematic study on colloid deposition and released in porous media and their association with both Pb(II) and Cd(II).

The main factors controlling the transport of heavy metals in porous media can be classified as follows: chemical factors such as pH, ionic strength and heavy metal type; and physical factors such as flow rate, medium grain size and influent concentration [16]. The isotherms of heavy metals competitive adsorption suggest that there is different selective adsorption sequence for multi-metal adsorption, which can be

interpreted by their electronegativity, hydrolytic ability, Misono softness parameter, first hydrolysis constant [17,18]. Increased flow rate and decreased sand grain size also have strong impact on the fate and transport of heavy metal contaminants in porous media [19–21].

The main purpose of this work was to investigate the effect of colloids on the co-transport of Pb(II) and Cd(II) in saturated porous media under different physical conditions. Quartz sand was used as porous media for the column experiments. The specific objectives of the research were as follows: (1) compare Pb(II) and Cd(II) transport with different types of colloids, (2) determine the effect of flow rate and grain size on retention and transport of Pb(II) and Cd(II) with different types of colloids

## 2. Materials and methods

### 2.1. Materials

Lead ( $\text{Pb}(\text{NO}_3)_2$ , Tianjin Tianli Chemical Reagent Company, Tianjin, China) and cadmium ( $\text{CdCl}_2$ , Aladdin industry corporation, Shanghai, China) mixed solution was prepared with deionized (DI) water at a concentration of  $50 \text{ mg L}^{-1}$ . Manual loessial soil was pulverized and passed nylon sieve with 1 mm diameter. Montmorillonite (Shanghai Fengxian Fengcheng Chemical Reagent Company, Shanghai, China) colloids and manual loessial soil colloids were prepared according to the procedure of Gao et al. [22]. 10g of manual loessial soil or montmorillonite was suspended in 200 mL DI water. After 24 hours shaking, the suspension was placed in ultrasonic bath for 30 minutes and then allowed to sit for 24 hours. The fraction of colloids in suspension were collected as experimental colloids and the concentrations were determined gravimetrically. The solutions were diluted to  $500 \text{ mg L}^{-1}$  of manual loessial soil colloid or montmorillonite colloid with DI water before each experiment.

HA colloid (Tianjin Guangfu Fine Chemical Research Institute, Tianjin, China) was prepared following the method described by Hu et al [23]. The HA content of the stock solution was measured to be  $30 \text{ mg L}^{-1}$  by UV-visible spectrophotometry (UV-2800, UNICO Instrument Co., Ltd., USA) based on a known dissolved mass concentration [23]. The zeta ( $\zeta$ ) potential of the colloids was determined by measuring the electrophoretic mobility with a Zeta-Plus (Zetasizer nano ZS90, Malvern Instruments, UK) [24]. Quartz sand (Shaanxi Zhouzhi County Quartz Sand Company, China) with a size ranges of 0.4–0.8 mm and 0.8–2.0 mm was used in this study as the porous media. Before experiment, the quartz sand was washed sequentially by 0.1 M NaOH solution and 0.1 M HCl solution, followed by washing with DI water until there was no impurities and metal oxide. After washing, the quartz sand was dried in an oven at  $105^\circ\text{C}$  for 4 hours.

### 2.2. Column experiments

A plexiglass column of 3 cm in diameter and 15 cm in height was wet-packed with quartz sand [25]. Approximately 160 g of quartz sand was used to pack one column with a porosity of 0.45. Once packed, flow in the column was induced in an upward direction using a peristaltic pump. The saturated column was leached with DI water for 3 h to precondition it and to remove impurities. The transport experiment consisted of two injection stages. At stage one, contaminant solution was injected into the column for about 2.5 pore volumes (PVs) at a steady flow rate ( $0.1, 0.5, 1.0 \text{ ml min}^{-1}$ ). At stage two, the column was leached with manual loessial soil colloid, montmorillonite colloid, colloidal HA and DI water (blank control) for 5 pore volumes (PVs) at the same flow rate as the stage one, respectively. In addition, 0.01 M KCl as a conservative tracer test was conducted following the same procedures.

### 2.3. Effluent analysis

Effluent samples were collected from the outlet in glass test tube every 20 mL with an automatic fraction collector (BS-16A-LCD, Shanghai Huxi Analysis Instrument Factory Co., Ltd., China). Manual loessial soil colloid, montmorillonite colloid and HA colloid concentrations were measured by UV-visible spectrophotometry (UV-2800, UNICO Instrument Co., Ltd., USA) at 350 nm [3], 410 nm [26] and 254 nm [27], respectively. Pb(II) and Cd(II) concentrations were analyzed by an atomic absorption spectroscopy (AAS, Z-2000, Hitachi, Japan). All treatments were conducted in duplicates and the average values were reported.

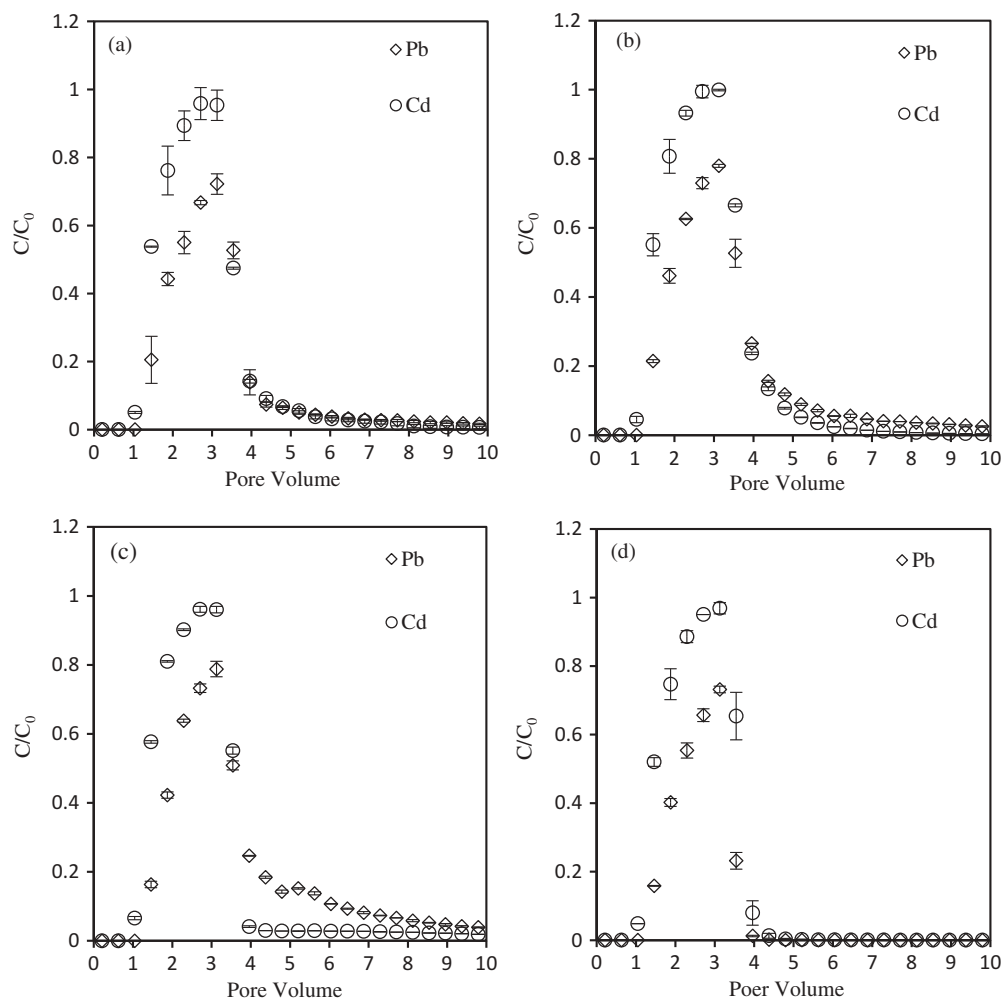
### 2.4. Statistical analysis

Data were analyzed by variance using Statistical Analysis System. A t-test was used to determine the statistical significance ( $P < 0.05$ ) between Pb(II) and Cd(II) under different experimental conditions. The colloid data under different flow velocities and quartz sand grain sizes were tested in the same method.

## 3. Results and discussion

### 3.1. Effect of different colloids on the co-transport of Pb(II) and Cd(II) in saturated quartz sand column

The co-transport behaviors of Pb(II) and Cd(II) in the porous media with different types of colloidal solution (montmorillonite colloid, manual loessial soil colloid, and HA) are presented in Figure 1. The flow rate was  $0.5 \text{ mL min}^{-1}$  and quartz sand with particle diameters ranging from 0.4 mm to 0.8 mm was used in these experiments. The y coordinate shows the ratio of outlet to the inlet heavy metal concentration ( $C/C_0$ ), and



**Figure 1.** Breakthrough curves of Pb and Cd in saturated porous media at different colloid types at flow rate of  $0.5 \text{ ml min}^{-1}$  (a) montmorillonite colloid; (b) manual loessial soil colloid; (c) HA, (d) DI water).

**Table 1.** Conditions and results of transport experiments of Pb(II) and Cd(II) with different colloids.

Colloid	Heavy metal	Recovery rate (%)		Zeta (mV)
		Heavy metal	Colloid	
Montmorillonite colloid	Pb	62.32%	80.21%	-40.47
	Cd	86.44%		
Manual loessial soil	Pb	73.84%	74.21%	-30.70
	Cd	93.97%		
Humic acid	Pb	79.64%	83.40%	-52.77
	Cd	87.15%		
DI water	Pb	45.81%	-	-
	Cd	81.26%		

the abscissa shows the number of PVs of the solution that had passed the porous media. Under the same experimental condition, Cd(II) was first detected in the effluent at about 1.25 PVs, while Pb(II) appeared later than Cd(II) at about 1.7 PVs.

Cd(II) had a higher peak concentration than Pb(II), and the breakthrough peak of Cd(II) appeared earlier than that of Pb(II). The different phenomenon of the breakthrough curves (BTCs) suggested that the mobility of Cd(II) was higher than that of Pb(II) in saturated porous media. The recovery of Cd(II) from the sand column was higher than that of Pb(II) under the same

conditions (Table 1). Under the condition of four types of colloidal solutions, a total of 86.44%, 93.97%, 87.15% and 81.26% of the Cd(II) and 62.32%, 73.84%, 79.64% and 45.81% of Pb(II) eluted from the quartz sand for montmorillonite colloid, manual loessial soil colloid, HA and DI water, respectively. In addition, the BTCs of Pb(II) and Cd(II) showed gradual falling to  $C/C_0 = 0$ , but Pb(II) concentrations reduced more slowly. This phenomenon was particularly obvious when the column was flushed by HA. The recovery rate of Pb(II) differed significantly ( $P < 0.0001$ ,  $F = 15,208.3$ ) and Cd(II) also differed significantly ( $P < 0.0001$ ,  $F = 3382.8$ ), which were under the same experimental conditions. The phenomena were ascribed to the stability constants of heavy metals, chelating ability, different electronegativity, mass charge ratio and other factors [28], which led to different ways of adsorption of Pb(II) and Cd(II) on quartz sand. The adsorption of Pb(II) occurs on variable charge sites in binary system. The adsorption of Cd(II), however, occurs predominantly on permanent charge sites in the system. Because of its lower hydrolytic ability, Cd(II) does not compete effectively for variable charge surfaces, and so its adsorption is

more restricted to permanent charge sites [17]. As a result, Pb(II) has higher selectivity of sorbents than Cd(II) [29], which explains why Pb(II) showed lower mobility than Cd(II) in saturated porous media in this work.

Aqueous colloidal solution affected the co-transport of heavy metals in porous media. Compared with DI water, montmorillonite colloid, manual loessial soil colloid and HA all enhanced the mobility of Pb(II) and Cd(II). In presences of the three colloidal solutions, the recovery rates of Pb(II) were 62.32%, 73.84% and 79.64%, respectively, higher than that (45.81%) of DI water. The same recovery rate trend happened to Cd(II), the recovery rate of Cd(II) increased from 81.26% to 86.44%, 93.97% and 87.15%, respectively (Table 1). This indicated that HA had stronger abilities to remobilize Pb(II) and Cd(II) from quartz sand surface than montmorillonite colloid. This was mainly due to the large number of functional groups on the surface of manual loessial soil colloid and HA, such as carboxyl, phenol, carbonyl and hydroxyl groups attached to aliphatic or aromatic carbons [30]. The presence of such carboxyl and phenol groups in HA provided a large site for the adsorption of Pb(II) and Cd(II) [31]. At the same time, the Zeta potential of the manual loessial soil colloid was larger, which implied that the colloidal silica colloids with Pb(II) and Cd(II) repulse more due to the positive charge on the surface, making the recovery of Pb(II) and Cd(II) greater.

Pb(II) BTCs of HA reduced slower at the end of experiment than other types of colloids, indicating that the remobilization process of Pb(II) from quartz sand surface was slow. There was no significant difference of Cd(II) peak breakthrough with montmorillonite colloid, manual loessial soil colloid, HA and DI water. But under the influenced by HA and manual loessial soil colloid, the peak appeared earlier than montmorillonite colloid and DI water. This phenomenon could be explained that HA and manual loessial soil colloid were more effective than montmorillonite colloid and DI water to remobilize Cd(II) from the sand surface. Cd(II) BTCs with manual loessial soil colloid also showed the remobilization process of Cd(II) from the sand took a long time; resulting in manual loessial soil colloid was more likely to transport Cd(II) in porous media. As observed, Pb(II) was more easily adsorbed on the medium and difficult to be desorbed, the conclusion was validated by the calculation of recovery rates.

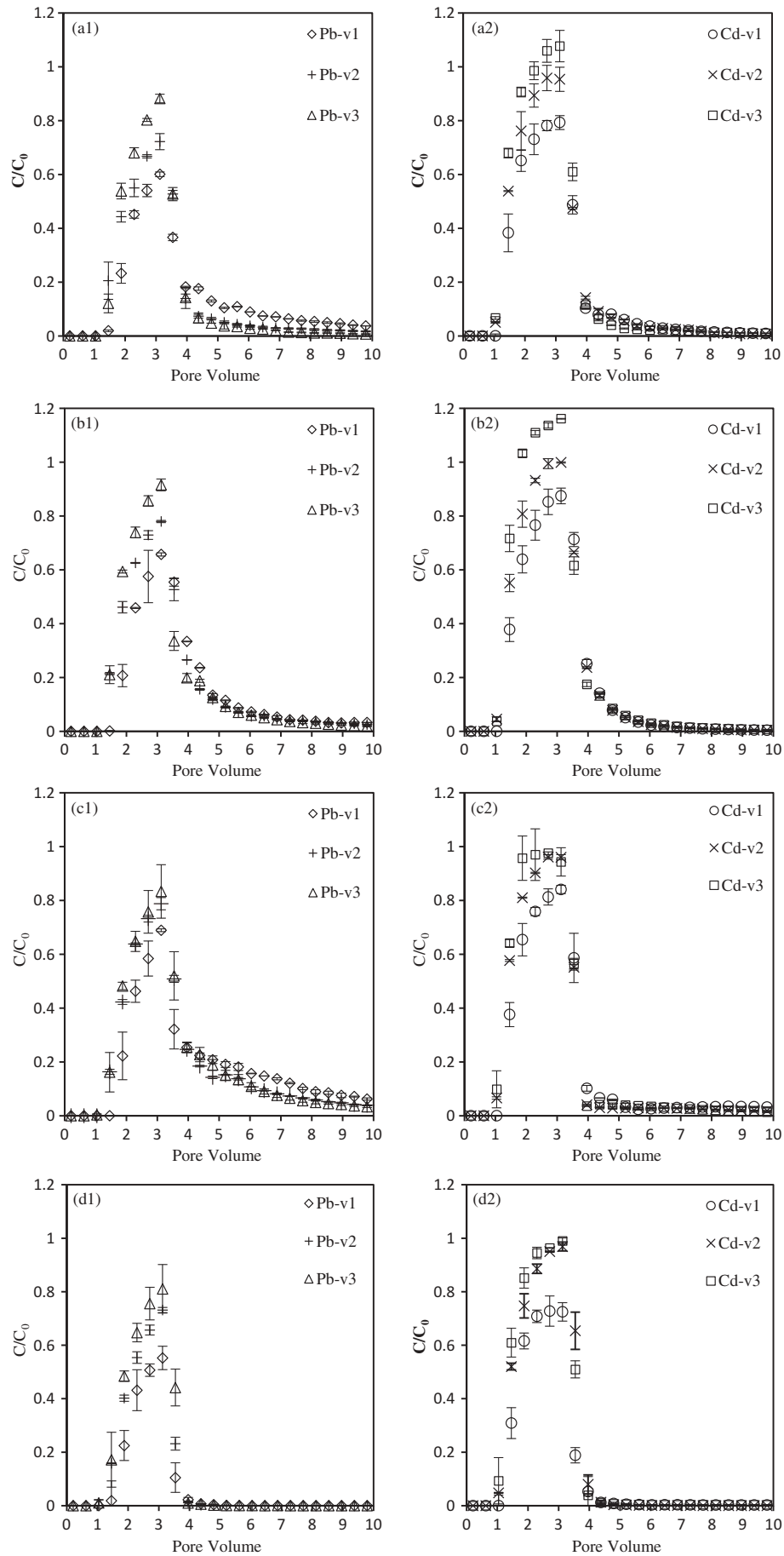
Compared with the three colloidal solutions under the same experimental condition, the transport of colloid in the quartz sand column revealed different behaviors, the recovery rate of montmorillonite colloid, manual loessial soil colloid and HA were 80.21%, 74.21% and 83.4%, respectively (Table 1). The recovery rate of the three colloids differed significantly ( $P < 0.05$ ,  $F = 214.2$ ). The surface charge characteristics of colloidal particles could significantly affect the

mobility of colloids in porous media. The colloidal particle and sand surfaces are often negatively charge [32]. The zeta potentials of montmorillonite colloid, manual loessial soil colloid and HA were  $-40.47$  mV,  $-30.7$  mV and  $-52.77$  mV, respectively (Table 1). The electrostatic repulsion between the quartz sand and colloidal molecules is strongly impacted by the value of zeta potential. Electrostatic repulsion decreased as zeta potential increased, leading to increasing retention of colloids. The manual loessial soil colloid with highest zeta potential led to its highest retention in the quartz sand column in comparison to the other two colloids. The zeta potential of HA was the minimum and thus showed the lowest retention. The retention amount of montmorillonite colloid was between those of manual loessial soil and HA and the trend matched the calculation of colloid recovery rates.

### 3.2 Effect of flow rate on the co-transport of Pb(II) and Cd(II) in saturated quartz sand column

The effect of flow rate on the BTCs of Pb(II) and Cd(II) in the porous media is showed in Figure 2. The transport of Pb(II) and Cd(II) in the column was accelerated by faster flow rate. Under the same experimental conditions, when the flow rate increased from  $0.1$  to  $1.0$  ml min<sup>-1</sup>, the recovery rates of heavy metals and colloids increased. In addition, the mobility of Pb(II) and Cd(II) with colloidal solutions showed the same trend of recovery rates. At the end of BTCs, Pb(II) concentration followed by slow gradual falling until  $C/C_0 = 0$  as the flow rate decreased. For example, after being flushed by the montmorillonite colloidal solution with an increase of flow rate (Figure 1(a)), the peak concentrations of Pb(II) and Cd(II) increased. After increasing flow rate, the recovery rates of Pb(II) and Cd(II) in the column increased from 58.26% to 67.14% and 72.97% to 94.23%, respectively. Meanwhile, the recovery rate of montmorillonite colloid increased from 71.23% to 81.56% (Table 2). The recovery rate of Pb(II) under the three flow velocities differed significantly ( $P < 0.0001$ ,  $F = 3368.4$ ). Similar result was observed in Cd(II) recovery rate under the same conditions ( $P < 0.0001$ ,  $F = 8403.3$ ).

Among these colloidal solution transport experiments, the BTCs and recovery rates showed the same trend as flow velocity increased. The recovery rates of Pb(II) and Cd(II) with the colloidal solution (manual loessial soil colloid, HA or DI water) were all consistent with the trend of montmorillonite colloid, which were differed significantly ( $P < 0.0001$ ). From previous research, we know that the rate and extent of colloid detachment increase as the flow velocity increase [21]. The reason for this phenomenon was attributed to that increase of flow velocity led to a decreased contact time between heavy metals and quartz sand



**Figure 2.** Breakthrough curves of Pb(II) and Cd(II) in saturated porpus media at different flow rates (a) montmorillonite colloid; (b) manual loessial soil colloid; (c) humic acid; (d) DI water; v1 = 0.1 ml min<sup>-1</sup>, v2 = 0.5 ml min<sup>-1</sup>, v3 = 1.0 ml min<sup>-1</sup>.



**Table 2.** Recovery rate of Pb(II), Cd(II) and colloids with different flow rates.

Colloid	Heavy metal	Flow rate (ml min <sup>-1</sup> )	Recovery rate (%)	
			Heavy metal	Colloid
Montmorillonite colloid	Pb	0.1	58.26%	71.23%
	Cd	0.1	72.97%	
	Pb	0.5	62.32%	80.21%
	Cd	0.5	86.44%	
	Pb	1	67.14%	81.56%
	Cd	1	94.23%	
Manual loessial soil	Pb	0.1	63.53%	13.46%
	Cd	0.1	81.41%	
	Pb	0.5	73.84%	74.21%
	Cd	0.5	93.97%	
	Pb	1	77.83%	75.24%
	Cd	1	100.00%	
Humic acid	Pb	0.1	73.12%	83.07%
	Cd	0.1	77.17%	
	Pb	0.5	79.64%	83.40%
	Cd	0.5	87.15%	
	Pb	1	82.77%	84.14%
	Cd	1	95.34%	
DI water	Pb	0.1	31.24%	–
	Cd	0.1	56.39%	
	Pb	0.5	45.81%	–
	Cd	0.5	81.26%	
	Pb	1	55.71%	–
	Cd	1	84.45%	

surface in the columns, resulting in decreasing of adsorption from heavy metals to porous media surface to enhance the mobility of Pb(II), Cd(II) and colloids in the columns. Increasing flow velocity may also decrease colloid straining efficiency to increase their mobility in the porous media [33,34]. Results of this work clearly demonstrated that the value of  $C/C_0$  became higher when flow rate increase, which also enhanced the recovery rates of Pb(II), Cd(II) and colloids in experiments.

Experimental results also showed that flow velocities had different influences on three types of colloidal solutions. At the flow rate of 1.0 ml min<sup>-1</sup>, the peak of  $C/C_0 > 1$  was shown in Figure 2(a,b). Such irregularities might come from the competition between Pb(II) and Cd(II) and their different affinities towards quartz sand surface. The first-order hydrolysis constant of Pb(II) ( $pK_1 = 7.9$ ) was higher than that of Cd(II) ( $pK_1 = 10.1$ ), leading to the stronger hydrolytic ability of Pb(II) than that of Cd(II), the mobility of Cd(II) was faster than that of Pb(II) in the porous media, resulting earlier breakthrough of Pb(II) [35].

### 3.3. Effect of grain size on the co-transport of Pb(II) and Cd(II) in saturated quartz sand column

The BTCs showed grain size affected the transport of Pb(II) and Cd(II) in porous media. Under the same experimental conditions (Figure 3(a)), when sand grain size range increased from 0.4–0.8 mm to 0.8–2.0 mm, the plateaus of the  $C/C_0$  curves of Pb(II) and Cd(II) both increased and the recovery rates of the two heavy metals and colloids also increased (Table 3). Compared with that of Cd(II), the mobility

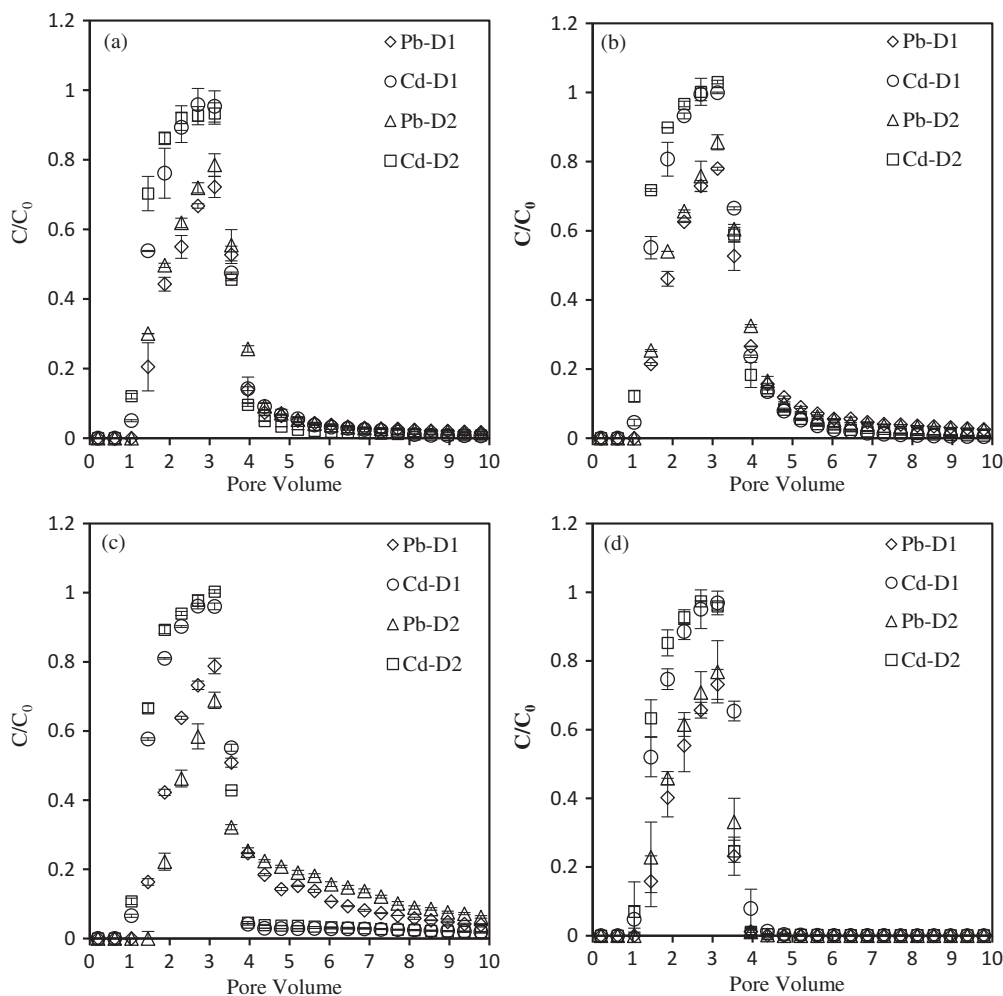
of Pb(II) was affected more obviously as the grain size increased. This was mainly because the outflow of the colloid increased and the recovery rate of the heavy metal increased with the sand size increasing. The larger the particle size of the medium, the less strain the colloid was subjected to by the medium, and it was not easily deposited on the surface of the medium [21].

Increasing the grain size in the column, the recovery rates of montmorillonite colloid increased from 62.32% to 70.58% for Pb(II) and 86.44% to 87.78% for Cd(II). The recovery rates of Pb(II) ( $P < 0.0001$ ,  $F = 3280.1$ ) and Cd(II) ( $P < 0.05$ ,  $F = 49.195$ ) with the medium of two grain sizes differed significantly. Peak effluent concentration decreased with the decreasing of sand grain size and the recovery rate of montmorillonite colloid increased from 80.21% to 83.38% (Table 3). This was also because that the increase of the medium size made the pores increase, the colliding probability between the colloid and the quartz sand decreased, and the recovery rate of the colloid increased [21].

The recovery rate of montmorillonite colloid and manual loessial soil colloid differed significantly ( $P < 0.05$ ), but the recovery rate of HA did not ( $P > 0.05$ ,  $F = 14.567$ ). In the cases of other colloidal solutions, the BTCs and recovery rates showed the same trend as increasing sand grain size. With smaller grain size, the quartz sand specific surface area increases, it thus can adsorb more heavy metal ions. Straining is another important mechanism of colloid retention in porous media. Under this mechanism, the retained colloids are found to be dependent on the grain size distribution [36]. In this study, when sand grain size decreased from 0.8–2.0mm to 0.4–0.8mm, the quartz sand pore size decreased, resulting in enhanced straining of colloids and thus hindered their transport in porous media [36–38].

## 4. Conclusion

This work was conducted to determine the effects of colloidal solution, flow rates and sand grain size on the co-transport of Pb(II) and Cd(II) in saturated porous media. Experimental results showed that (1) colloidal solution promoted the co-transport of Pb(II) and Cd(II), which were influenced by colloids surface charge characteristic, zeta potentials and competitive adsorption of heavy metals; (2) higher flow rate decreased the adsorption of Pb(II), Cd(II) and colloids on quartz sand surface, gave rise to high mobility of heavy metals and colloids in sand columns; (3) increasing grain size decreased quartz sand pore size and thus enhanced straining of montmorillonite and manual loessial soil colloids, which influenced the retention and transport of Pb(II), Cd(II). This implied that the migration of heavy metals carried by the



**Figure 3.** Breakthrough curves of Pb(II) and Cd(II) in saturated porous media at different grain sizes at flow rate of  $0.5 \text{ ml min}^{-1}$  (a) montmorillonite colloid; (b) manual loessial soil colloid; (c) HA; (d) DI water; D1 = 0.4–0.8mm, D2 = 0.8–2.0mm).

**Table 3.** Recovery rate of Pb(II), Cd(II) and colloids with different grain sizes.

Colloid	Meavy metal	Grain size (mm)	Recovery rate (%)	
			Heavy metal	Colloid
Montmorillonite colloid	Pb	0.4–0.8	62.32%	80.21%
	Cd	0.4–0.8	86.44%	
	Pb	0.8–2.0	70.58%	83.38%
	Cd	0.8–2.0	87.78%	
Manual loessial soil	Pb	0.4–0.8	73.84%	74.21%
	Cd	0.4–0.8	93.97%	
	Pb	0.8–2.0	79.15%	81.04%
	Cd	0.8–2.0	99.65%	
Humic acid	Pb	0.4–0.8	79.64%	83.40%
	Cd	0.4–0.8	87.15%	
	Pb	0.8–2.0	82.46%	84.92%
	Cd	0.8–2.0	91.02%	
DI water	Pb	0.4–0.8	45.81%	–
	Cd	0.4–0.8	81.26%	
	Pb	0.8–2.0	52.05%	–
	Cd	0.8–2.0	78.43%	

natural colloids due to the competitive adsorption between different heavy metals may lead to the diffusion of certain heavy metals far more than other heavy metals, resulting in great environmental risks; secondly, it is also necessary to pay attention to the effect of changes in the subsurface environment on the promotion of colloidal metal migration. This work

provides a insight into the co-transport behavior of different heavy metals competing for natural colloids.

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## Disclosure statement

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