

## Core ideas

- Soil from highly CKDu affected area showed higher concentration of colloids
- Colloid deposition increases with the decreasing flow rate and with increasing ionic strength
- Cd(II) attachment was pronounced at low flow rate and high ionic strength
- Colloid attachment is the key mechanism on colloid retention in porous media

Transport of colloids and colloid-facilitated heavy metals in agricultural soils: Could it be a potential causative factor for the chronic kidney disease with unknown etiology (CKDu) in Sri Lanka?

## Abbreviations:

AAS, Atomic Absorption Spectrometer; CFT, Colloid-Facilitated Transport; CKDu, Chronic Kidney Disease of unknown Etiology; CSQG, Canadian Soil Quality Guidelines for the Protection of Environmental and Human Health; EC, Electrical Conductivity; HOC, Hydrophobic Organic Compounds; NCP, North Central Province; PSD, Particle Size Distribution; TDS, Total Dissolved Solids; WDC, Water Dispersible Colloids; WHO, World Health Organization

## Abstract

Naturally-occurring colloids, particles of diameter  $< 10\text{ }\mu\text{m}$ , are ubiquitous in Geo-environments and can potentially facilitate transport of numerous contaminants in soil including harmful heavy metals, pesticides, pathogens etc. via Colloid-Facilitated Transport (CFT). The CFT of contaminants to groundwater is still an underrepresented transport domain and may lead to significant environmental and health problems related to groundwater contamination. Colloid mobilization, transport and CFT in various geomedias are highly sensitive to physico-chemical perturbations. This study investigated colloid transport and colloid-facilitated heavy metal transport in saturated porous media with a series of column experiments using soil colloids extracted from two areas affected by Chronic Kidney Disease of Unknown Etiology (CKDu) in North Central Province of Sri Lanka. Colloid breakthrough curves were obtained from the column studies to observe the colloid transport under different flow rates ( $0.5\pm0.05$ ,  $1.65\pm0.05$ ,  $4.10\pm0.05\text{ cm}^3/\text{s}$ ) and ionic strengths (NaCl - 0.01 M, 0.05 M, 0.1 M). The CFT was studied using Cadmium (Cd(II)) as a model contaminant together with colloidal suspension under selected scenarios for high colloid deposition. Elevated colloid concentrations were observed in high CKDu affected area compared to the low endemic area. The experimental results were numerically simulated on an advection-diffusion/dispersion modelling framework coupled with first-order attachment, detachment and straining parameters inversely estimated using HYDRUS 1D software. Experimental and simulated colloid breakthrough curves showed a good agreement, and recognized colloid

attachment as the key mechanism for colloid immobilization in selected soil. Both colloids and CFT of Cd(II) showed pronounced deposition under low flow rate and high ionic strength.

## Introduction

The Chronic Kidney Disease of unknown etiology (CKDu) is a common health problem worldwide, in which the loss of kidney function occurs slowly and progressively over a period of several years, leading to a terminal kidney failure. The disease was first identified among the Sri Lankan community in early 1990s (WHO, 2012) in the North Central Province (NCP) of Sri Lanka which extends over 10,400 km<sup>2</sup> in the dry zone of the country and has a well-established irrigation system facilitating agriculture. Within the next three decades (1990 - 2020), the problem drew significant local and international attention as the most prominent non-communicable health concern in the country. Pathetically, the farmers (especially males) from poor socio-economic backgrounds in NCP became the victims of the CKDu, evidence suggested this to be around 15% - 23% in Anuradhapura and Polonnaruwa districts (Jayathilake et al., 2016). The number of victims of CKDu, exceeded 30,500 by 2017 as diagnosed at eleven hospitals from 2003 - 2017 (Ranasinghe et al., 2019). As estimated by some studies, the incidence of CKDu in Sri Lanka has been doubling every four to five years (Wimalasena, 2015). In turn, over 150,000 people have been affected by the disease by 2015 and about 3% of them lose their lives annually (Kafle et al., 2019). The emerging COVID-19 pandemic since 2019 has apparently overshadowed the CKDu epidemic in the country, and has significantly obscured the seriousness of the problem during the last couple of years.

Although various hypotheses surround the causative factors of the CKDu including heat/dehydration, pesticides, infections, water contamination with agrochemicals/heavy metals etc., none of them has been comprehensively investigated or scientifically established yet. Although the literature is abounding with extensive studies carried out from morphological, epidemiological, socio-economical, hydrological, toxicological, and agricultural perspectives, a conclusive causative factor has not been pinpointed yet. In fact, later studies believed that the problem is of multifactorial origin, given the complexity in tracing specific causative factors. Nevertheless, many researchers presumed that the CKDu has a strong coexistence with the presence of contaminants such as agrochemicals in groundwater, which is the prime drinking source of the agricultural community in the NCP. Some researchers have indicated that the possible risk factors for CKDu could be toxins from the environment (Wanigasuriya et al., 2011) and intensive use of fertilizers and pesticides in agricultural lands (Bandara et al., 2010, Jayathilake et al., 2013). The use of heavy-metal-contaminated water for irrigation has also shown to contribute heavy metal accumulation in crops, which later has become severely toxic to the functioning of vital human organs, kidneys and liver. Studies have demonstrated that Cadmium (Cd) and Lead (Pb) damage the renal tubules and lead to severe neurotoxin effects on the human kidneys (Järup, 2003). Further, Bandara et al. (2008) observed

that the Cd content in milk produced in the NCP was in the range of 0.05-0.15 mg/l, leading to a hypothesis that the grazing land areas were polluted with heavy metals. The scientific focus, however, has diverted to some other factors as well, for example the elevated levels of Fluoride, and complex interactions between the ratio of dissolved Sodium and Calcium in the presence of Fluoride, etc. (Paranagama et al., 2018). However, the impact of none of these factors has been scientifically proved nor been disproved yet, and hence their potential contributions cannot be downplayed in searching for the most probable causative factors for the CKDu problem.

Until late nineteenth century, the strongly-sorbing contaminants, such as heavy metals, were believed to be immobile in the vadose zone. The classical constitutional mechanisms governing contaminant transport in the vadose zone both in liquid and gaseous phases, are primarily based on advection, diffusion and dispersion of dissolved constituents (Charbeneau, 2006) in which contaminants with high affinity to soil were considered to be retarded in the solid phase. However, frequent observations of groundwater appearance of strongly sorptive contaminants in surprisingly high concentrations raised the scientific concerns to explore an obscured mechanism whereby strongly – sorptive contaminants can potentially migrate to groundwater. The colloid-facilitated contaminant transport (CFT) was later introduced as one such hidden frontier which could facilitate highly sorptive contaminant transport to groundwater. Colloids are technically described as particles of characteristic dimensions falling in the window of 1 nm to 10  $\mu$ m, although the upper and lower limits remain vaguely defined across literature. Colloids constitute mineral fragments, microbes, plant decay debris which are ubiquitous in subsurface environments (Denovio et al., 2004). Characteristically, colloids are highly charged and possess large specific surface area, the two key attributes which enable colloids to potentially facilitate and transport numerous contaminants through soils including radionuclides, hydrophobic organic compounds (HOCs), heavy metals, pesticides, pathogens etc. Heavy metals, in particular, were found to be mobile with higher rates in the presence of colloids. For instance, Zn and Cu were reported to be transported 5 to 50 fold more in the presence of colloids than in the absence of it (Karanthanas, 1999). Similarly, Pb mobility increased 10 to 3000 times with colloidal transport than without colloids (Karanthanas, 2000). Further, studies evidenced the transport of Zn, Pb and Cu facilitated by readily dispersible soils (Zang et. al., 2005). Moreover, Chen et. al. (2005) observed colloid facilitated transport of Cs while Flury et. al. (2002) found that, colloids which mobilized in flow experiments with packed sediments can carry Cs along. Furthermore, Villholth et. al. (2000) noticed evidence of colloid-facilitated transport of prochloraz to subsurface drains when investigating in situ colloid mobilization and transport. In a study done by Bin et. al. (2011) using soil columns (collected from Montreal, Canada) found that water flooded incubation reduces release of Pb, Fe, Al, Ca from soil by decreasing either the mobility of soil colloids or the associations between Pb and mobile colloids. A study conducted by Armienta et. al. (2016) using a particular soil in Mexico revealed that concentrations

of As and other metals considered (Zn, Fe, V, Cd, Cu) showed an important transportation in the entire study zone. The measured concentrations of As, Cd, Cu, Pb, V, and Zn in both dry and rainy seasons presented greater values than those recommended for agriculture and industrial sites by the Canadian Council of Ministers of the Environment (CSQG 2007). Notably, the physico-chemical properties have a significant effect on colloid transport which leads to colloid-facilitated transport of contaminants.

Despite sufficient evidences, the presence of colloidal phase as a potential transport domain is still overlooked and its role on facilitated transport of harmful contaminants is grossly downplayed in recent numerical tools. The experimental studies on colloid-facilitated transport of heavy metals under varying physico-chemical conditions are sparse in literature, and almost nil within the local context. In particular, no scientifically based systematic study has been undertaken to investigate the role of colloids on carrying contaminants such as agrochemicals and heavy metals to groundwater in CKDu affected regions in the NCP of Sri Lanka.

This study investigated the role of colloids and CFT of Cd (II), as a potential contaminant, in the subsurface environment of highly-CKDu affected area (Madawachchiya) and a relatively low-endemic area (Horowapothana) in Sri Lanka under varying soil physicochemical conditions. We hypothesize that soil colloids and colloid-facilitated transport may potentially play a role in groundwater transport of agrochemicals and thereby constitutes another causative factor for the CKDu prevalence in the NCP of Sri Lanka. The colloid and contaminant breakthrough curves obtained from a series of column studies under differing physical and chemical conditions were simulated within a classical advection-diffusion/dispersion modelling framework with first-order attachment/detachment and straining coefficients inversely estimated from the experimental data.

## Materials and Methods

### Materials

River sand was used as the porous media in all column experiments. The fraction passing through 2-mm sieve and retained on 425  $\mu\text{m}$  was selected in column experiments. Before packing, the sand was thoroughly washed and oven-dried at 105°C for 24 hours. Particle size distribution (PSD) of river sand was obtained following the sieve analysis and hydrometer methods. The PSD was parameterized using the modified Rosin-Rammler function (Chamindu Deepagoda et al., 2016) as shown in Fig. 1, which yielded the mean particle size ( $D_{50}$ ) of 1.0 mm.

The water-dispersible soil colloids (WDC) were extracted from agricultural soils sampled at 50 – 100 cm depth from Medawachchiya (8.5375° N, 80.4910° E) and Horowapothana (8.6181° N, 80.8294° E), which represent CKDu high-endemic and low-endemic areas, respectively. The co-located two areas were particularly selected due to the similar climate and environmental conditions as well as common agricultural practices but with distinct contrast in number of CKDu

cases. The collected soils were oven-dried at 105°C for 24 hours and sieved through a 2-mm sieve. The soil was then mixed with feed solution (tap water) at a ratio of 3:20 (W/W%), followed by manual shaking and leaving undisturbed for 20 hours at 25°C to allow coarser particles to settle. Then, the WDC which were less than the Stokes diameter 10  $\mu\text{m}$  were siphoned from the supernatant.

The physicochemical characteristics of the both soil types are listed in Table 1. In addition, 0.001 M NaBr was used as an ionic tracer to mimic the water flow. To observe the effect of ionic strength for colloid transport and CFT, 0.01 M and 0.1 M NaCl was used. Further,  $\text{CdCl}_2$  was used to investigate the CFT of Cd(II). All the chemicals used in the experiments were of analytical grade.

The percentage deposition of colloids in porous media were calculated for selected scenarios by integrating the colloid breakthrough curves.

Table 1

## Methods

### Experimental Setup

The apparatus used for column experiments consisted two 1-m high Marriott tanks and an acrylic column with 74 mm in diameter and 22 cm in height. For colloid transport experiments, first Marriott tank was filled with the feed solution (tap water) and the second was filled with the colloidal solution and the ionic tracer (0.001 M NaBr). The ionic strength was regulated by adding an ionic solution (0.01 M, 0.1 M, and 1.0 M NaCl) to the second Marriott tank. For contaminant transport experiments, Cd(II) was added to the second Marriott tank with the colloidal solution in addition to the tracer. The column consisted two 1-cm gravel-filled layers, each packed at the top and the bottom, which ensured uniform distribution of the influent and effluent solutions across the sand layers. The pre-treated sand was wet-packed in 5-cm layers to a height of 20 cm. Wet-packing with gentle tapping ensured removal of entrapped air and enhanced quality of packing. A filter paper (425  $\mu\text{m}$ , Whatman Inc<sup>TM</sup>) was kept at the bottom of the sand column to avoid the clogging of outlet by sand particles. The two Marriott tanks and the sand-packed column were hydraulically connected via a three-way valve ensuring no air entrapment. Finally, water and water-dispersible colloidal solution was allowed to flow in downward direction through the packed sand column while continuous stirring to keep the colloids dispersed. Water and colloidal solution were applied to the sand column in the sequences of 1-2 pore volumes of water, 4 pore volumes of water dispersible colloidal solution and again 4-10 pore volumes of water, in the given order. Effluent samples were collected in pre-determined time intervals and analyzed for turbidity, total dissolved solids (TDS), electrical conductivity (EC) and pH.

A series of colloid transport experiments were conducted under the simulated irrigation conditions and three different flow rates and three ionic strengths of the soil water-colloidal system for both soil types. Table 2 shows the details of the experimental series. In addition, CFT of Cd(II) was investigated for

selected colloid transport experiments. For heavy metal analysis, the effluent received from the column outlet was filtered by 2- m filter paper (Whatman Inc<sup>TM</sup>). and analyzed using the Atomic Absorption Spectrometry (AAS) (AA-7000 series, Shimadzu Corporation).

**Table 2**

#### Numerical Modelling

To numerically simulate the transport of water and colloidal particles in porous media, the HYDRUS-1D computer software package (Šimůnek et al., 2006) was used. The HYDRUS-1D potentially simulates water, heat and multiple solutes movement in one dimensional variably saturated media. The classical advection-dispersion modelling framework for solute transport in saturated porous media was invoked here as the governing equation, combined with kinetic attachment, detachment, straining models to specifically account the dynamics of colloid movement in porous media. The complete governing equation for colloid transport can be expressed as:

$$\frac{\partial C}{\partial t} = -\nu_x \frac{\partial C}{\partial x} + D_L \frac{\partial^2 C}{\partial x^2} - (k_{att} C - \frac{\rho_b}{\theta} k_{det} S_{att}) - k_{str} \Psi_{str} C \quad (1)$$

where  $C$  ( $\text{nL}^{-3}$ ) is the colloid concentration in the aqueous phase at a distance  $x$  (L) and time  $t$  (T),  $\nu_x$  ( $\text{LT}^{-1}$ ) is the interstitial pore water velocity,  $D_L$  ( $\text{L}^2\text{T}^{-1}$ ) is the hydrodynamic dispersion coefficient,  $k_{att}$  ( $\text{T}^{-1}$ ) is the first-order attachment rate coefficient,  $\rho_b$  ( $\text{ML}^{-3}$ ) is the dry bulk density of the porous medium,  $\theta$  ( $\text{L}^3\text{L}^{-3}$ ) is the porosity of the porous medium,  $k_{det}$  ( $\text{T}^{-1}$ ) is the first-order detachment rate coefficient,  $S_{att}$  ( $\text{nM}^{-1}$ ) is the attached particle concentration,  $k_{str}$  ( $\text{T}^{-1}$ ) is the first-order straining coefficient and  $\Psi_{str}$  (-) is the dimensionless colloid straining function.

#### Results and Discussion

##### Characterization of porous media, soil and soil colloids

Figure 1 shows the PSD curve for porous media material. The symbols represent the experimental values and the solid lines show the modified Rosin-Rammler distribution function (Chamindu Deepagoda et. al., 2016) as shown in Equation 4 with parameters best fit the measured data

$$p(x) = 100 \left[ 1 - e^{-\ln 2 \left( \frac{x}{D_{50}} \right)^\beta} \right] \quad (2)$$

where  $x$  (L) is the particle size,  $D_{50}$  (L) is the mean particle diameter and  $\beta$  (-) is a dimensionless empirical constant of the distribution. Notably, a larger  $D_{50}$  value denotes a larger average grain size whereas larger  $\beta$  denotes a poor gradation and hence a uniformly-distributed grain size.

### Figure 1

The PSD of sand demonstrated an average grain diameter of 1mm (i.e.  $D_{50} = 1$  mm) with a good gradation ( $\sigma = 1.2$ ). The mean grain size and size distribution has a direct bearing on colloid transport (and hence CFT of contaminants) due to their explicit influence on hydraulic conductivity for fluid flow and mechanical filtration and straining on colloid immobilization, both of which are functions of the grain size. Impact of grain size of hydraulic conductivity has been widely presented in literature and their potential relations have been extensively discussed (e.g., Kozeny–Carman; Carman (1937); Hazen relationship ; Hazen (1911)), although only a limited studies on colloid deposition on grain size are available. Colloid attachment has been observed more in coarse-grained sand than fine-grained sand likely due to the more accessible attachments sites when the grain size increases. Straining, on the other hand, is negatively related to the grain size due to wide availability of small pore throats and grain-grain contacts in fine-textured media which provide potential sites for straining. Due to the enhanced close packing, well-graded porous media can better retain colloids than poorly-graded (or well-sorted) porous media.

Figure 2 shows the modified PSD of soil colloids, i.e., after the siphoned extraction of WDC from the solution, for both high and low endemic soils. The WDC extracted from the soil collected from the two areas have apparently the same size distribution, albeit a little larger peak was observed in low-endemic soils as compared to high-endemic soils. Nevertheless, the mean colloid diameter for both soil colloids were found to be around 10  $\mu\text{m}$ . Interestingly, the PSD of the two soils (before extraction; not shown) were also very close to each other, implying no difference in the soil type, characterized as sandy loam, in both locations.

### Figure 2

Initial colloid concentrations, measured in terms of turbidity, shown in the Figure 3 reveals that the high endemic soil yielded approximately 30% higher concentration (on average) of colloids compared to the low endemic soil under both chemical (changing ionic strength; Fig 3a) and physical (changing flow rate; Fig. 3b) conditions. Notably, the availability of more colloids in high-endemic soil suggests, though not necessarily prove, higher colloid-facilitated contaminant transport in high endemic soils which could potentially be linked to the higher prevalence of CKDu in the high endemic areas. It is worth emphasizing that high colloid abundance does not necessarily mean higher colloid-facilitated transport, since the latter depends largely on colloid-contaminant interactions as well. Due to the pronounced initial WDC concentrations, hereafter the high-endemic soil was considered as representative soil to discuss the results since the two soils did not differ significantly with regard to the breakthrough patterns.

### Figure 3

Effect of physical perturbations on colloid breakthrough

In general, the natural in situ-released soil colloids, mobilized from sand particles, get first dispersed and started leaching when the feed solution is applied. In all the column experiments, the column was initially flushed by water to remove the in-situ mobilized colloids from the porous media, which are loosely attached to sand surfaces and mobilized to the macro pores (de jong et al., 1998). Typical breakthrough curves obtained under the varying flow rates (for physical perturbations) are shown in the Figure 4. The breakthrough of colloids occurred after flushing one pore volume, and gradually reached a uniform plateau and remained steady during colloid application period. The symmetric breakthrough curves demonstrate the physical homogeneity within the flow domains under a given flow rate. Note that both soil colloids and the ionic tracer seem to have occurred simultaneously in all profiles, suggesting that the effect of size and charge exclusion phenomena were not pronounced. This could have occurred since the system mimics a clean bed removal with no pronounced colloid blocking or filter ripening processes (Kretzschmar et al., 1997). No noticeable tailing was observed in the receding curves of the colloids and the tracer. It can be clearly seen that high deposition of colloids occurred in low flow rate  $0.50 \text{ cm}^3/\text{s}$  (figure 4a) as compared to high flow rate (Fig. 4c) due apparently to the increased sedimentation and decreased hydrodynamic shear at low flow rates. The percentage colloid depositions were 28.9% , 16.3 % , and 5.5% for  $0.50 \text{ cm}^3/\text{s}$ ,  $1.6 \text{ cm}^3/\text{s}$ , and  $4.0 \text{ cm}^3/\text{s}$ , respectively. This is further confirmed by numerical simulations as described later. Due to the significant colloid deposition at low flow rate, it was selected to further investigate the effect of chemical perturbation as discussed below.

#### Figure 4

Effect of chemical perturbations on colloid breakthrough and deposition

Colloid breakthrough curves were observed for three ionic strengths (0.10 M, 0.05 M and 0.01 M NaCl) at low flow rate ( $0.50 \text{ cm}^3/\text{s}$ ). At low ionic strength, electrostatic repulsive forces between colloids and solid matrix reduces the collision efficiency, ( $\alpha < 1$ ) which leads to a low deposition (Fujita and Kobayashi, 2020). Conversely, at high ionic strength, electro-repulsive forces between colloids and solid matrix increases the collision efficiency ( $\alpha = 1$ ) leading to a higher deposition (Fujita and Kobayashi, 2020). This phenomenon is well demonstrated in Figure 5 by the colloid breakthrough curves and agreed well with the results in Ryan and Elimelech (1996). The observed percent deposition of colloids were 47.6%, 54.3 %, and 67.8% for the ionic strengths of 0.01 M, 0.05 M and 0.1 M NaCl, respectively. According to Kretzschmar et al. (1999), the decreasing ionic strength results in a large colloid peak in the effluent, indicating a strong increase in the particle release rate at low ionic strength. Also, particle release at low ionic strength is prominent only in the presence of monovalent counter ion at the colloid and matrix surface. When the surfaces are saturated with divalent counter ions, such as Cd(II), particle release remains low even at very low ionic strengths. Since chemical disturbances cause release of attached colloids, this tailing can be assumed to be a result of detachment of colloids due



to the decrease in ionic strength after shifting the pulse from colloidal solution to water. The sudden distinct peak, occurred around six pore volumes in Fig. 5b (0.05 M NaCl) and Fig. 5c (0.1 M NaCl) demonstrate release of detached colloids at high ionic strengths.

### Figure 5

This implies that some of the colloids are attached to sand grains reversibly. Colloids which are loosely retained in the secondary energy minima mobilize to pore waters, as changes in ionic strength eliminate the secondary energy minimum (Kuznar and Elimelech, 2007). Further, if the repulsive forces are high enough to lift up some weakly strained colloids, liberation of strained colloids may also have contributed to the late release of colloids. It was presumed here both detachment (of attached colloids) and liberation (of strained colloids) contributed the sudden release of colloids.

### Simulation of colloid transport

Hydrus - 1D (Šimůnek et al., 1998) was utilized to simulate the colloid transport in all column experiments. The critical scenarios related to the high colloid deposition were simulated to estimate the transport parameters for high-endemic soil as shown in Figure 6.

### Figure 6

Notably, the simulations were done in a two-step process. As the first step, the observed results for tracer were simulated using Hydrus 1D and estimated the hydraulic conductivity and hydrodynamic dispersivity in the sand-packed column under given condition. In the second step, first-order attachment, detachment and straining coefficients were simultaneously obtained inversely from the simulation of colloid breakthrough curves while the hydraulic conductivity and hydrodynamic dispersivity were fixed at the pre-determined values obtained from the tracer simulations. Both simulated colloids and tracer profiles were well fitted with the experimental data and the obtained fitted parameters are listed in Table 2. In soil colloids, attachment, straining and detachment coefficients may have significant figures due to the presence of metal oxides and organic matter, and some positively charged sites can also be expected (Chen et al., 2020). As explained in experimental breakthrough curves, higher colloid attachment coefficients were obtained for higher ionic strengths.

### Table 2

#### Colloid-Facilitated Transport (CFT) Experiments

Figure 7 shows the observed breakthrough curves of colloids and Cd(II) for the critical scenarios leading to high colloid deposition: low flow rate without change in ionic strength (7a), and low flow rate with high ionic strength (7b), both leading to high deposition in colloid transport. Note that the for heavy metal analysis, the colloidal fraction below 2  $\mu\text{m}$  was used, and hence the Cadmium

breakthrough curve exhibits the Cd(II) in dissolved phase and adsorbed on to  $< 2$  m colloidal phase.

### Figure 7

For the low flow rate without changing ionic strength (figure 7a), there is a comparatively low adsorption of colloids on the immobile solid matrix, and hence high colloid transport and colloid-facilitated Cd(II) transport in the system. Consequently, a very little Cd breakthrough in the dissolved phase and with  $< 2$  m colloidal particles was observed. When the clean water is flushed after the 5 pore volumes, a large amount of colloid-attached Cd(II) released to the dissolved phase, yielding a high colloid breakthrough after 5 pore volumes. Conversely, when high ionic strength colloidal solution is applied (Fig. 7b), colloid attachment to the solid matrix is comparatively high, yielding less colloid breakthrough with the colloidal phase and more Cd(II) release in the dissolved phase. However, when clean water is applied after 5 pore volumes, the attached colloids detached from the solid matrix and remobilized in the system, and therefore much of the Cd(II) ions were co-transported in the colloidal phase. Therefore, significant transport of Cd(II) in the dissolved phase was not observed. The observations, therefore, clearly indicate the impact of ionic strength in colloid-facilitated Cd(II) transport in the chemically-varied porous system. Table 3 indicates the parameters obtained from simulation of high-endemic soil colloids.

### Table 3

#### Simulated Experiment on CFT of Cd(II)

A computational domain representing a 2-m deep conceptual soil profile was defined to analyze the time taken for Cd(II) to reach the groundwater, which is presumed to be at a 2-m depth with no capillary fringe. The soil profile is assumed to be similar to the saturated porous media of the column experiment and capillary zone is of negligible height. Concentration of 10 mg/l Cd(II) was applied as influent at the surface for 24 hours period for two scenarios (0.50 cm<sup>3</sup>/s without NaCl and 0.50 cm<sup>3</sup>/s with 0.1 M NaCl) as mentioned in the CFT experiments. The simulation was done using the inversely estimated parameters as shown in Table 3.

### Figure 8

Figure 8 shows the two-dimensional graphical representation of colloid-facilitated Cd(II) transport in subsurface. As seen from figure 8, it can be clearly observed that the migration of Cd(II) significantly varied with the ionic strength of the colloidal solution. As expected, compared to the high ionic strength, Cd(II) transport is faster in the scenario without NaCl. A larger depth seems to have affected by Cd(II) retention in soil at higher ionic solution (0.1 M NaCl) as compared to 0 M NaCl solution, due to the pronounced Cd(II) attachment at higher ionic strength. Similar observations were made in the studies done by Chen et al. (2018) and Shi et al. (2022).

## Conclusions

This study investigated transport of colloid-sized soil particles of diameter  $< 10$   $\mu\text{m}$  extracted from soils sampled from two Sri Lankan sites in Medawachchiya (a high CKDu endemic area) and Horowpothana (a low CKDu endemic area) using saturated sand as the porous media. The laboratory column experiments for colloid transport were conducted under different flow rates (0.5, 1.6, 4.0  $\text{cm}^3/\text{s}$ ) and ionic strengths (0.01 M, 0.05M, 0.1 M NaCl). Medawachchiya soil yielded 30% higher concentration of colloids compared to the those from Horowapothana soils, but did not differ appreciably with respect to colloid breakthrough curves. Colloid deposition decreased with increasing flow rate, from 28.9% in 0.5  $\text{cm}^3/\text{s}$  to 5.5 % in 4.0  $\text{cm}^3/\text{s}$ , due to increased hydrodynamic shear at grain surfaces. Further, at a given flow rate, colloid deposition increased with increasing ionic strength, from 47.5 % at 0.01 M NaCl to 67.8% at 0.1M NaCl at low flow rate (0.5  $\text{cm}^3/\text{s}$ ), due to less net repulsive forces at high ionic strength. A distinct colloid peak was observed in the receding curve at high ionic strengths, due to remobilization of weakly deposited colloids. Numerical simulations conducted on an advection-dispersion framework combined with first-order colloid attachment, detachment and straining coefficients using HYDRUS 1D showed a good agreement with observed and simulated colloid breakthrough curves. Colloid-facilitated transport of Cd(II) as a model contaminant proved high Cadmium deposition under high ionic strength.

This study thus provided a new insight on tracing potential causative factors for the CKDu problem in Sri Lanka from a geo-environmental perspective invoking the potential of colloid-facilitated transport of contaminants, but with some limitations. Only one soil from high endemic area was selected for the analysis; a detailed study needs different soil types across the affected areas to further investigate hypotheses. We used river sand as the porous domain, but further studies including insitu soils as the porous media may provide useful implications. Cadmium (Cd (II)) only was used as a model contaminant, close investigations are necessary with other potential contaminants including agrochemicals (e.g., glyphosate) and their fate in the subsurface. A field-scale colloid transport study could provide more realistic observations than a laboratory-scale study to derive more meaningful conclusions.

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#### Captions of Figures and Tables

Figure 01. The Particle size distribution of the porous medium. Circles denote the

measured data while the solid line denotes the best-fit modified Rosin-Rammler parametric

function, Equation 4 ( $D_{50} = 1 \text{ mm}$ ;  $\sigma = 1.2$ )

Figure 02. The size distribution of the water-dispersible soil colloids extracted from both

high-endemic (red line) and low-endemic (blue line) soils

Figure 03. Initial turbidity (measured in Nephelometric Turbidity Units, NTU) of high

endemic (red line) and low-endemic (blue line) colloids used in column experiments under (a)

different ionic strengths and (b) different flow rates

Figure 04. Colloid breakthrough curves (in red color) for (a) low flow rate (0.50 cm<sup>3</sup>/s), (b)

medium flow rate (1.60 cm<sup>3</sup>/s) and (c) high flow rate (4.0 cm<sup>3</sup>/s) for high-endemic soil colloids. Breakthrough curves of the tracer is also illustrated (in blue color)

Figure 05. Experimental breakthrough curves for (a) low ionic strength (0.01 M), (b) medium

ionic strength (0.05 M), and (c) high ionic strength (0.10 M) for high-endemic soil colloids

Figure 06. Simulated breakthrough curves for low flow rate (a) without changing ionic strength

and (b) high ionic strength (0.10 M) for high-endemic soil colloids

Figure 07. Experimental breakthrough curves for tracer transport (blue line) colloid transport

(red line) colloid-facilitated transport of Cd<sup>2+</sup> (green color) for low flow rate (a) without

changing ionic strength (0M NaCl) and (b) high ionic strength (0.10 M NaCl) for high-endemic

soil colloids

Figure 08. Two-dimensional graphical representation of Cadmium (Cd II) transport with depth

against time in low flow rate (a) without changing ionic strength (0 M NaCl) and (b) high ionic

strength (0.10 M NaCl) for high-endemic soil colloids. Color contours denote Cd (II)

concentrations

Table 1: Physio-chemical properties of two soil types used for the study.

Table 2: Experimental series

Table 3: Parameters obtained from simulation of high-endemic soil colloids

Tables

Table 1: Physio-chemical properties of two soil types used for the study.

Soil Type	Particle Density (g/cm <sup>3</sup> )	Bulk Density (g/cm <sup>3</sup> )	Zeta pote
Medawachchiya (High-endemic)	2.65	1.70	-14.2 ± 4.1
Horowpothana (Low-endemic)	2.63	1.69	-18.4 ± 2.7

Table 2: Experimental series consisting of physical perturbations and chemical perturbations

Physical perturbations- Flow Rate (cm <sup>3</sup> /s)	Chemical perturbations- NaCl as ionic strength (M)
0.5 ± 0.05	0.01
1.6 ± 0.05	0.05
4.0 ± 0.05	0.1

\*0.001 M NaBr as tracer element in physical perturbations, 0.5 cm<sup>3</sup>/s as flow rate in Chemical perturbations

Table 3: Parameters obtained from simulation of high-endemic soil colloids

Parameter	Symbol	Value	
		For 0.50 cm <sup>3</sup> /s flow rate with 0 M NaCl	For 0.50 cm <sup>3</sup> /s flow rate with 0.1 M NaCl
Hydrodynamic dispersivity of Na <sup>+</sup> tracer (cm)	D <sub>L</sub>		
First order attachment coefficient (min <sup>-1</sup> )	k <sub>att</sub>		
First order detachment coefficient (min <sup>-1</sup> )	k <sub>det</sub>		
First order straining coefficient (min <sup>-1</sup> )	k <sub>str</sub>	32x10 <sup>-4</sup>	8.1x10 <sup>-4</sup>