Influence of non-uniform flow on toxic elements transport in soil column percolation test

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ABSTRACT

The influence of flow channels on the leaching behavior of toxic elements in contaminated soil cannot be neglected in a column percolation test. This study presents a visualization of the flow channel formed in the soil and evaluated the relationship between the leaching behavior of soil components and flow. We conducted column percolation tests with two types of filling methods (Compaction and No compaction) and used X-ray computed tomography to visualize the soil structure and non-uniform flow. Additionally, the variations of flow in a cross section of water were evaluated using hydraulic conductivity based on differential pressure. Under No compaction, a flow occurred throughout the soil column at the beginning of the water passage, but a non-uniform flow emerged as the liquid–solid ratio increased; under Compaction, a non-uniform flow was formed from the beginning of water passage. The leaching behavior of the major components and toxic elements from soil with high adsorptive properties was significantly affected by the filling method up to a liquid–solid ratio of 2. These results suggest that the non-uniform flow formed in the column percolation test has a significant impact on the leaching of soil components.

1. Introduction

Column percolation tests are used to evaluate the leaching behavior of toxic elements in soil and recyclable resources (Kalbe et al., 2007; Katayama et al., 2020; Pantini et al., 2015; Tsiridis et al., 2015). In general, the column percolation test is a standardized method, e.g., ISO 21268-3 in 2019 and CEN/TS 1405 (2004), which is used by various international organizations (DIN, 2009; US EPA, 2013). Following the method of the column percolation test, the soil is filled in a column and a continuous vertical up-flow of water through the column is used for a certain period to contact the sample with the solvent. By measuring the solvent that passes through the soil, the leaching behavior of toxic element components in the soil can be determined.

The column percolation test method can result in a complex flow, which may affect the leaching behavior of toxic elements from samples during the test. Nakamura et al. (2014) and Yasutaka et al. (2017) simultaneously conducted standardized column percolation tests using the same sample at several institutions. According to the obtained results, the cumulative concentration of toxic elements resulted in an error of approximately 20% at a liquid–solid ratio of 10, regardless of the leaching trends. Furthermore, Naka et al. (2016) elaborated upon the shortening of the column percolation test, which revealed that the concentration and trend of the toxic elements at liquid–solid ratios ranging up to 1 did not correspond to those from multiple test results. The leaching behavior was not reproduced owing to the influence of multiple factors such as ion type, ion competition, initial concentration in the soil (i.e., soil composition), hydraulic conductivity, porosity, sorption/desorption mechanism, and contact area. Internationally standardized tests, such as the column percolation test, require improved reproducibility of toxic element concentrations and elution trends.

The flow of water through the soil is complicated by soil structure. As suggested by the Kozeny–Carman equation, the soil’s structure significantly contributes to hydraulic conductivity. The formation of complex structures produces non-uniform flow in the soil and causes complex solute transport from the soil (Filipović et al., 2020; Gerke, 2006; Luo et al., 2008). Additionally, the non-uniform flow in the column significantly contributes to the mass transfer in the soil (Liu and Jeng, 2019; Zhang and Chui, 2019). With respect to the leaching mechanism of toxic elements in the soil and recyclable resources obtained in the column percolation tests, the formation process of the non-uniform flow in the column should be considered.
Thus, the characteristics of the internal structure, such as the pore distribution and the resultant non-uniform flow, should be appropriately visualized using X-ray computed tomography (CT) (Luo et al., 2010; Munkholm et al., 2012) to understand the transport of solutes and non-uniform flow. Van Offenwert et al. (2019) and Sammartino et al. (2015) visualized the solute transport and non-uniform flow in soils using X-ray CT, which allowed the fast imaging of materials. The leaching behavior of soil components can be further investigated using the above-mentioned imaging technique to visualize the non-uniform flow in soil.

In this study, we visualized the non-uniform flow formed during the column percolation tests and investigated the variation in the leaching behavior of soil components with respect to the variations in the non-uniform flow using hydraulic conductivity as an indicator. This study conducted column percolation tests with different packing methods and used X-ray CT with iodine solution. The leaching trends of major components (9 elements) and toxic elements (7 elements) obtained from each column test and the changes in flow and hydraulic conductivity were evaluated.

2. Materials and methods

2.1. Materials

The column percolation tests were conducted using sandy soil in a manner similar to that in prior research (Nakamura et al. (2014) and Yasutaka et al. (2017)). The soil sample was air-dried at 25 °C for 14 d or a longer duration, and the particle size was adjusted to <2 mm.

The particle diameter distribution is presented in Figure 1. The water content, particle density, and particle size distribution were measured following the JIS A 1203 (2009), JIS 1202 (2009), and JIS A 1204 (2009) standardized methods, respectively. The moisture content and soil particle density of the soil samples used in this study were 3.12 wt% and 2.59 g/cm³, respectively.

An energy dispersive X-ray fluorescence spectrometer (EDX-720, Shimadzu Corporation, Kyoto, Japan) was used to quantitatively analyze the soil components, and the analysis method was quantified using an absolute calibration curve method (Yamasaki et al., 2011). The concentrations of the major components and toxic elements in the soil sample are listed in Tables 1 and 2.

2.2. Methods of column percolation tests

In this study, the column percolation test was conducted according to the ISO 21268-3 in 2019 and CEN/TS 1405 standards. The column percolation test is illustrated in Figure 2, and its experimental conditions are listed in Table 3.

The column represented in Figure 2 composed of acrylic (inside diameter: 5 cm; height: 30 cm). A pressure gauge was placed underneath the column to measure the differential pressure between the solvent flow from the pump and the atmospheric pressure. The column was saturated with water and subsequently allowed to stand for a duration of at least 66 h. Subsequently, distilled water was passed through the column at a flow rate of 12.5 ± 2 ml/min. Depending on the classification, 100–200 ml of the solution was collected at the top of the column and filtered using a membrane filter of 0.45 μm.

The leaching tendency of soil components due to the variation of non-uniform flow formed in the column was investigated. The non-uniform flow in the column was varied according to the difference in the soil structure resulting from the two distinct filling methods utilized in this study. The filling method pertained to compaction, wherein multiple layers were compacted using 125 g rammers per the ISO-TS 21268-3 standard (Compaction). The other method is to drop the sample from the top and fill it without compaction (No compaction). The weights of the soil filled for the Compaction and No compaction methods were 588.7 g and 543.5 g, respectively.

The concentrations of F⁻, Cl⁻, NO₃⁻, and SO₄²⁻ were measured using an ion chromatography technique (Metrohm 881 Compact IC proAnion), and the concentrations of Na, Ca, Fe, Mg, and Mn were measured using an ICP optical emission spectrometer (5110 ICP-OES; Agilent Technologies). The soil concentrations of the toxic elements Cr, Co, Ni, Cu, Zn, As, and Sb were measured using an ICP mass spectrometer (NexION®1000, PerkinElmer). Additionally, the pH and EC of the test solutions were measured.

The hydraulic conductivity of the column was determined using the differential pressure generated by the water flow. The hydraulic conductivity $K$ (m/s) was calculated using Darcy’s law as follows (refer to Eq. (1)).

$$K = \frac{Q \cdot \rho \cdot g \cdot A}{\Delta P \cdot L}$$  \hspace{1cm} (1)

where $Q$ is the flow rate (m³/s), $\rho$ is the density of water (kg/m³), $g$ is the acceleration of gravity (m/s²), and $A$ is the cross-sectional area of the column (m²), $L$ is the column length (m), and $\Delta P$ is the pressure difference between atmospheric pressure and the bottom of the column (Pa).
2.3. Visualization of soil structure and non-uniform flow by X-ray CT

The X-ray CT images of the columns were obtained using an X-ray CT scanner (ScanXmate-D225RSS270, Comscantecno Co., Ltd.) to visualize the pore distribution in the column (voltage: 120 kV; X-ray tube current: 150 μA; voxel size: 48 μm × 48 μm × 48 μm).

The non-uniform flow in the column represents the water flow through the pores. As depicted in Figure 3, we used potassium iodide aqueous solution (KI solution) to visualize this non-uniform flow. Koestel and Larsbo (2014) utilized a KI solution in conjunction with X-ray CT imaging techniques to visualize the flow in soil. The KI solution iodide exhibited a high tendency to absorb X-rays, and the CT numbers of water and a KI solution were remarkably distinct. Therefore, the visualization of the non-uniform flow formed in the soil using X-ray CT could be rendered possible. In this study, a X-ray CT imaging method was employed by leveraging these particular properties of KI solution to visualize the non-uniform flow formed during the percolation column test. As reported in an existing study, the concentration of the KI solution was 15 wt% (Watanabe et al., 2019).

The flow visualization method in this study utilizing the KI solution is illustrated in Figure 3. In this study, we visualize and discuss the difference between the flow at the beginning of the test and the flow after the end of the test (four cases total) using columns filled with soil before and after the column percolation test. To visualize the flow path, KI solution was injected from the bottom end into a column saturated with distilled water, and an X-ray CT scan was performed. The distillate was then drained to push away some of the KI solution present in the column, and an X-ray CT scan was performed again. The KI solution was passed through the column in the same volume of distilled water needed to saturate the column. The KI solution is used to visualize the excellent flow formed; the point of difference between the CT value of the KI solution and the water remaining in the column soil indicates the flow in the column. The injection rate of KI solution and water was 12.5 ± 2.5 mg/min, as in the column percolation test. It is presumed that where the KI solution was washed away with distilled water, the KI solution in the high flow area was removed compared to the surrounding area, resulting in a significant decrease in the CT number. Thus, we evaluated the difference in CT number before and after rinsing out the KI solution with distilled water (CTN_{KI-Water}), and the three-dimensional distribution of CTN_{KI-Water} is described in Eq. (2).

\[
\text{CTN}_{\text{KI-Water}} = \text{CTN}_{\text{KI}} - \text{CTN}_{\text{water}}
\]

where CTN_{KI-Water} is the difference in the CT number between KI solution and distilled water, CTN_{KI} is the CT number after the injection of KI solution, and CTN_{water} is the CT number after washing the KI solution with distilled water. CTN_{KI-Water} can be less than 0 in column. It means the area where KI solution and distilled water do not pass through, or the area where distilled water passes through and KI solution remains. Therefore, when CTN_{KI-Water} is less than 0, the flow is extremely small compared to the region where CTN_{KI-Water} is large. The region where CTN_{KI-Water} < 0 is defined as CTN_{KI-Water} = 0. Thereafter, the following

**Table 3. Basic experimental conditions.**

| Parameter                   | Value |
|-----------------------------|-------|
| Column diameter [cm]        | 5     |
| Column height [cm]          | 30    |
| Sample state [-]            | Air dry |
| Flow rate [ml/h]            | 12.5 ± 2.5 |
| Initial equilibration (time after saturation) [h] | 66 |
| Packing method [-]          | Compaction, No compaction |
| Filling amount of Compaction [g] | 588.7 |
| Filling amount of No Compaction [g] | 543.5 |
| Eluent [-]                  | Distilled water |
| Filter paper [μm]           | 0.45  |

Figure 2. Diagram of column percolation test (left: experimental image; right: photo of experiment).

Figure 3. Method to visualize non-uniform flow formed in column.
calculation was performed for a medium voxel size of 384 μm × 384 μm × 384 μm, which was created from the original voxel data in the image reconstruction process, because a large amount of data was produced at the original voxel size (48 μm × 48 μm × 48 μm).

In order to compare the flow before and after the passage and with different filling methods, CTND/M was calculated by dividing CTN_{KI-Water} by the maximum value of CTN_{KI-Water} (CTN_{Max}) (Eq. (3)). The CTN_{Max} was 3200 HU and 2800 HU before and after the passage of Compaction, and 4000 HU and 3000 HU before and after the passage of No compaction.

\[
CTN_{D/M} = \frac{CTN_{KI-Water}}{CTN_{Max}}
\] (3)

The position corresponding to relatively large CTN_{D/M} signifies the position at which the KI solution was rinsed out using a considerable

Figure 4. Photo of soil packed in column and internal structure observed under X-ray CT.

Figure 5. Non-uniform flow formed by Compaction. When CTN_{D/M} is close to 1, it becomes the priority channel in the column. (a) the beginning of water connection, (b) the end of water connection.
amount of distilled water, and the corresponding flow that presumably possessed a high flow rate. The three-dimensional distribution of CTN<sub>D/M</sub> was rendered using Voxler<sup>®</sup> 3 (HULINKS), a three-dimensional data visualization tool.

3. Results and discussion

3.1. Visualization of soil structure in a column under X-ray CT

This study used X-ray CT to obtain the soil structure inside the column for the two filling methods (Compaction and No compaction) prior to the water passage. Figure 4 shows the soil before water was passed through, and the corresponding soil structure observed by X-ray CT. The photographic observation of the column revealed that (a) the soil was composed of a layered structure resulting from Compaction, whereas (b) the soil without compaction exhibited no layered structure.

In the X-ray CT image, the CT number was expressed in grayscale, and a higher CT number corresponds to a color proximate to white. In the case of Compaction, a layered structure produced by the compaction can be observed in the soil structure inside the column. In contrast, the structure of the soil without compaction was observed under the X-ray CT image, which revealed that the particles existed in the form of a mountain inside the column, including large voids adjacent to the column wall. This was caused by the modified soil structure formed inside the column owing to the variations in the filling method of the soil in the column.

3.2. Visualization of non-uniform flow and hydraulic conductivity during column percolation test

The three-dimensional visualization of the non-uniform flow in the column before the percolation column test obtained using Eq. (2) and Eq. (3) is depicted in Figures 5 and 6. A greater change occurred before and after water flow in No compaction than in Compaction, according to a comparison of Figures 5 and 6. In the compacted soil presented in Figure 5, the non-uniform flow inside the column did not vary much before and after the water passage. In addition, a non-uniform flow was formed near the column wall. As observed in Figure 6, the column filled with the No compaction soil displayed the flow throughout the column at the beginning of the column percolation test, after which a non-uniform flow was formed near the column wall.

In the filling method without compaction in Figure 4(b), the soil in the column is relatively loose compared to that with compaction. Therefore, the flow was not restricted by the soil structure formed in the column. In the soil packed by compaction in Figure 4(a), the soil was packed in layers and immobilized in the column by compaction. Therefore, the flow in the column changed little before and after the column test compared to No compaction. In Compaction, the flow formed in the column is not significantly different before and after the percolation column test.

Figure 6. Non-uniform flow formed under No compaction condition. When CTN<sub>D/M</sub> is close to 1, it becomes the priority channel in the column. (a) the beginning of water connection, (b) the end of water connection.

Figure 7. Variations in hydraulic conductivity in column percolation test.
The hydraulic conductivity was calculated using Eq. (1). Figure 7 shows the change in hydraulic conductivity with the two filling methods, and it can be seen that the trend changes significantly at a liquid-solid ratio of 2. This indicates a change in the flow direction in the column. The hydraulic conductivity of the No compaction is $1.5 \times 10^{-3} \text{ m/s}$ at the beginning of the water passage, which immediately and rapidly

Figure 8. Trends of (a) pH and (b) EC in column percolation test.

Figure 9. Leaching trends of major elements (a) $F^-$, (b) $Cl^-$, (c) $NO_3^-$, (d) $SO_4^{2-}$, (e) Ca, (f) Fe, (g) Mg, (h) Mn, and (i) Na in column percolation test.
decreased after the initiation of water passage, attaining up to $2.1 \times 10^{-6}$ m/s at a liquid–solid ratio of 1 and $1.7 \times 10^{-6}$ m/s at a liquid–solid ratio of 2. The hydraulic conductivity of No compaction varied approximately 100 times between the liquid–solid ratios of 0–2. The hydraulic conductivity of the Compaction was $1.2 \times 10^{-5}$ m/s at the immediate beginning of the water passage, which gradually decreased to $3.1 \times 10^{-6}$ m/s after the water passage up to a liquid–solid ratio of 2.

The flow was determined by the soil structure and column percolation tests were performed using two types of filling methods (Compaction and No compaction). Three-dimensional visualization of the non-uniform flow showed that there was a large difference in the start of the water flow depending on the filling method. This difference was caused by the distribution of flow rates in the column. However, after the water flowed up to a liquid–solid ratio of 10, the difference in the non-uniform flow between the filling methods disappeared. Based on the variations in the hydraulic conductivity evaluated by the differential pressure measured in the column percolation test, the non-uniform flow formed in the column was affected by the variations in the filling method up to a liquid–solid ratio of 2. The previous studies suggested that the variations in the soil filling method and soil moisture content affected the hydraulic conductivity (Poulsen and Blendstrup, 2008; Van Verseveld and Gebert, 2020). In addition, a relationship exists between the hydraulic conductivity, pore distribution, and flow in the soil (Alaoui et al., 2011; Arya et al., 1999). Therefore, the variations in the filling method influenced the hydraulic conductivity and the non-uniform flow in the column until the liquid–solid ratio of 2 was obtained in this study.

The compaction presented in Figure 5 did not cause any variations in the flow. The soil in the column was immobilized by tightening with rammers. In the column percolation test, the water passed from the bottom end of the column, presumably changing the structure of the unconfinned, packed soil due to flow. The soil in the column flows like that in No compaction due to the fixation of the soil structure and the removal of air by the passage of water. Finally, the permeability and non-uniform flow, which were fluctuating at the start of the water flow, were similar at the end of the column infiltration test.

### 3.3 Changes in leaching tendency due to different packing methods in column percolation test

The leaching tendency of various elements in the soil was primarily caused by the pH and EC of the solution. The pH and EC obtained from the column percolation tests are presented in Figure 8. In the case of Compaction, the EC reached more than 80 mS/m immediately after the water passage and rapidly decreased to 10 mS/m up to a liquid–solid ratio of 1. In particular, the EC gradually increased at the solid–liquid ratio of 2–10, and at a liquid–solid ratio of 10, and it was almost equal to that of the No compaction. In the case of No compaction, the EC attained 60 mS/m up to a liquid–solid ratio of 2, and thereafter, gradually decreased to a steady state of 20 mS/m with the passage of water. The pH of water was slightly higher in the No compaction soil than the Compaction soil for liquid–solid ratios within 0–2, and the numbers were identical thereafter. This increases the frequency of soil-water contact when the permeability is high. Conversely, when the hydraulic conductivity is low, the frequency of soil-water contact decreases. Therefore, pH and EC are expected to be higher when the hydraulic conductivity is high in the early stages of water flow.

The leaching trends of the major components and toxic elements obtained from the column percolation tests are shown in Figures 9 and 10.
The leaching trends of the major components in the column percolation tests portray the characteristics of each soil constituent. (a) F in Figure 9 illustrates the various leaching trends depending on the filling method. In case of Compaction, the concentration fluctuated at the initiation of the water passage, but it increased subsequently. In case of No compaction, the concentration increased up to a liquid–solid ratio of 2, and thereafter, reached a steady state. The initial concentrations of (b) Cl, (d) SO\textsubscript{4}\textsuperscript{2−}, and (i) Na varied with the filling method, they displayed similar trends, and the concentrations decreased from the beginning of water passage to a liquid–solid ratio of 2, after which it attained a steady state. Moreover, the leaching tendency of (c) NO\textsubscript{3} and (f) Fe relied on the filling method. In particular, (c) NO\textsubscript{3} leached at a constant concentration with the water passage in the No compaction, whereas it leached at a high concentration at the initiation of the water passage in the Compaction. (f) Fe decreased to a liquid–solid ratio of 2 in case of No compaction, and eventually, reached a steady state. In case of Compaction, Fe was leached at a constant concentration upon the water passage. The concentrations of (e) Ca, (g) Mg, and (h) Mn decreased with a liquid–solid ratio of 2 in both the filling methods. This element has a similar trend to hydraulic conductivity, with lower concentrations for lower permeability (L/S = 2–5) and similar concentrations for the same hydraulic conductivity (L/S = 5).

As depicted in Figure 10, the leaching trend of (a) Cr and (c) Ni decreased to a liquid–solid ratio of 2 and became steady thereafter in case of No compaction. In comparison, the compacted soil portrayed a similar trend, but the reduction in the concentration up to a liquid–solid ratio of 2 was greater than that in No compaction. (d) Cu has a lower concentration in No compaction than in Compaction, contrary to the filling method for (a) Cr and (c) Ni. For (b) Co and (f) As, the leaching tendency in No compaction was reduced by water passage, but no significant variation was observed in the concentration in case of Compaction. Moreover, (e) Zn and (g) Sb did not display any variations in the leaching tendency or concentration depending on the soil filling method.

This study examined the relationship between the leaching trends of soil components obtained from the column percolation tests with two types of filling methods and variations in the flow in the column. Based on the results of this study, the leaching concentration and leaching tendency varied depending on the filling method. In particular, the concentration of certain soil components may increase with the altered hydraulic conductivity at an altered liquid–solid ratio of 2 owing to the water passage. Specifically, if the hydraulic conductivity is high in the early stages of water flow, pH and EC will increase. The leached concentrations of many elements increase accordingly. Conversely, some elements are more easily adsorbed and decrease in concentration due to their ionic strength. Thus, the non-uniform flow formed during the column percolation test significantly influenced the leached components.

4. Conclusion

In this study, the column percolation tests with two types of filling methods were conducted to visualize the non-uniform flow formed by the soil structure. This study discussed factors related to flow, such as non-uniform flow and hydraulic conductivity, as well as changes in the leaching behavior of soil components.

The characteristics of the flow formed during the column percolation test relied on the filling method. In particular, a non-uniform flow was formed on the column wall for the compaction soil, whereas a non-uniform flow spread in the column at the beginning of the water passage through the No compaction soil. This indicated that the flow in the soil varied during the column percolation test owing to the distinct soil-filling method. Upon comparing the hydraulic conductivity, the No compaction soil varied approximately 100 times, and the Compaction altered 10 times as the water flowed in the column.

The leaching concentration and tendency of soil components varied considerably depending on the two types of filling methods used in the column percolation tests. This finding suggested that the influence of the flow exhibited a distinct tendency to increase or decrease based on the element.

This column percolation test is necessary to evaluate the leaching trend and concentration of toxic elements from the target and to reproduce the results. The flow becomes unstable in its early stage, which affects the leaching concentrations and trends of soil components. The flow then immediately stabilizes and, for many elements, leaching concentration and trends are no longer affected by differences in filling methods. Therefore, the results obtained by the column percolation test are not expected to significantly affect the leaching concentration and trends. The concentrations and trends of toxic elements by column percolation tests according to standardized methods can be a characteristic of the elements contained in the sample, whose reproducibility is guaranteed by official regulations.

However, there are various non-uniform flows with complex leaching behavior of soil components in the geotechnical environment. Therefore, in order to understand the leaching concentration and trends of toxic elements in the actual environment, standardized column tests are considered insufficient. Thus, the leaching behavior of toxic elements should be evaluated at contaminated sites by, for example, collecting soil cores with retained soil structure for direct use in column percolation tests.

Declarations

Author contribution statement

Kyouhei TSUCHIDA: Performed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper.
Kengo NAKAMURA: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.
Noriaki WATANABE: Conceived and designed the experiments; Performed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper.
 Takeshi KOMAI: Analyzed and interpreted the data; Wrote the paper.

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Data availability statement

The data that has been used is confidential.

Declaration of interests statement

The authors declare no conflict of interest.

Additional information

No additional information is available for this paper.

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