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The paper was published in the proceedings of the 20th International Conference on Soil Mechanics and Geotechnical Engineering and was edited by Mizanur Rahman and Mark Jaksa. The conference was held from May 1st to May 5th 2022 in Sydney, Australia.

Column test for arsenic attenuation by sandy soil amended with calcium-magnesium composite of different particle sizes

Test en colonne pour l'atténuation de l'arsenic par un sol sableux amendé avec un composite calcium-magnésium de différentes tailles de particules

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ABSTRACT: The attenuation performance of basic and amended soils against arsenic is studied by column tests using flexible-wall permeameters. The amended soils had one of two particle size fractions of a calcium-magnesium composite (<2.0 or 2–9.5 mm). The agent content was 5% (50g/kg-soil by dry weight). Arsenic solution with a concentration of 0.1 mg/L was used as the permeant for the tests. Results show that the amended soils have better arsenic attenuation performance than basic soil. Breakthrough, taken as $C/C_0 = 0.1$, occurred after about 150 pore volumes of flow for the basic soil. The breakthrough was delayed when amended soils were used for the tests; it took about 430 and 860 pore volumes of flow when the agent was 2.0–9.5 mm and <2.0 mm in diameter, respectively. By using the experiment results, arsenic transport in the attenuation layer was discussed based on a simple case scenario using a one-dimensional advection-dispersion analysis. Assuming a 30-cm thick, saturated attenuation layer created with amended soil and the influent arsenic concentration is 0.1 mg/L, the effluent arsenic concentrations are expected to exceed the regulatory standards in Japan (0.01 mg/L) after 56 or 91 years when the agent employed is 2.0–9.5 mm and <2.0 mm in diameter, respectively.

RÉSUMÉ: La performance d'atténuation de sols basiques et amendés contre l'arsenic est étudiée par des tests en colonne utilisant des perméamètres à paroi flexible. Les sols amendés avaient une des deux fractions granulométriques d'un composite calcium-magnésium (<2.0 ou 2–9,5 mm). La teneur en agent était de 5% (50g/kg de sol en poids sec). Une solution d'arsenic d'une concentration de 0.1 mg/L a été utilisée comme perméant pour les tests. Les résultats montrent que les sols amendés ont une meilleure performance d'atténuation de l'arsenic que le sol de base. La percée, prise comme $C/C_0 = 0.1$, se produit après environ 150 volumes de pores d'écoulement pour le sol de base. La percée a été retardée lorsque les sols amendés ont été utilisés pour les tests; il a fallu environ 430 et 860 volumes de pore d'écoulement lorsque l'agent était de 2.0–9.5 mm et <2.0 mm de diamètre, respectivement. En utilisant les résultats des expériences, le transport de l'arsenic dans la couche d'atténuation a été discuté sur la base d'un scénario simple utilisant une analyse unidimensionnelle d'advection-dispersion. Dans l'hypothèse d'une couche d'atténuation saturée de 30 cm d'épaisseur créée avec un sol amendé et d'une concentration d'arsenic de 0.1 mg/L dans l'influent, les concentrations d'arsenic dans l'effluent devraient dépasser les normes réglementaires au Japon (0.01 mg/l) après 56 ou 91 ans lorsque l'agent utilisé a un diamètre de 2.0–9.5 mm et <2.0 mm, respectivement.

KEYWORDS: attenuation layer method, geogenic contamination, advection-dispersion analysis.

1 INTRODUCTION

Naturally occurring or geogenic toxic elements are widely distributed and occur in numerous types of natural rock and sediment, from marine clays to igneous and hydrothermally altered rocks (Inui *et al.* 2014; Li *et al.* 2017a; Tabelin *et al.* 2018). The way to properly manage the geogenically contaminated soils and rocks, after excavating them, has become an important consideration in many construction sites since it involves the risk of the leaching of unacceptable concentrations (Inui *et al.* 2014; Tabelin *et al.* 2014; Li *et al.* 2017b). The most straightforward way is the disposal as wastes. However, it should be noted that the concentrations are usually only slightly higher than the regulatory standards (Ito & Katsumi 2020), so they may not lead to serious environmental risks. Therefore, it is highly recommended to utilise the geogenically contaminated soils and rocks, after excavating them, taking appropriate countermeasures against the risk due to the geogenic contamination, rather than the simple disposal of them.

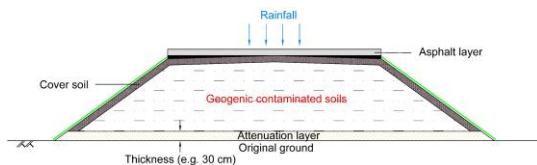


Figure 1. Schematic diagram of attenuation layer underlain geogenically contaminated soils and rocks in an embankment (Gathuka *et al.* 2021).

Many construction sites have been utilising geogenically contaminated soils and rocks for embankment and other geotechnical applications, taking different countermeasures. The widely applied countermeasures include encapsulation using geomembrane, chemical treatment to stabilise the contaminants and compaction to minimise the infiltration. Recently, installation of an attenuation layer has been anticipated as a countermeasure with a different concept from these other methods (Tatsuhara *et al.* 2012; Nozaki *et al.* 2013; Mo *et al.* 2020). Geogenically contaminated soils and rocks are embanked on top of the attenuation layer, as shown in Figure 1. The constituent materials of the attenuation layer sorb the contaminants of concern during leachate flow through the attenuation layer. A constituent material of the attenuation layer is the stabilising agent; it is chosen to stabilise the contaminants of concern as well as strengthen the attenuation/sorption capacity of the host soil (Nozaki *et al.* 2013; Gathuka *et al.* 2021; Kato *et al.* 2021a). Several benefits exist for this method. One of them is that it enables the construction of stable earthen structures with a simple and traditional manner. However, many issues remain to be solved to enhance the reliability of the attenuation layer method and widen the scope of its applications. Evaluating the attenuation performance of constituent materials of the attenuation layer is an important issue, which has been studied by several researchers, including by the authors (Mo *et al.* 2020; Gathuka *et al.* 2021; Kato *et al.* 2021b).

This study proposes using granular agents rather than powder ones as constituent materials for the attenuation layer. Employing granular agents in the field has several benefits: reduced dust and clumping, better soil grading, improved mechanical properties and can assure the attenuation layer has a certain permeability. However, there lies a concern in decreased attenuation performance caused by the smaller specific surface area, which requires evaluation of attenuation performance of the granular agents. Based on this need for evaluation, column tests using flexible-wall permeameters were carried out in this study to discuss the attenuation performance of basic and amended soils against arsenic, which is an important contaminant in geogenic contamination in Japan (Tabelin & Igarashi 2009; Takahashi *et al.* 2011; Tabelin *et al.* 2012; Inui *et al.* 2013; Yokobori *et al.* 2015). The tests were used to monitor the effluent arsenic concentrations at several liquid-to-solid (*L/S*) ratios, including much smaller ratios than those in batch sorption tests. Smaller ratios can better represent the in-field interactions of leachate with the constituent materials. Two particle size fractions of a stabilising agent were considered for preparing the amended soils in this study, namely, <2.0 mm and 2.0–9.5 mm. Similar to our previous study (Gathuka *et al.* 2021), the stabilising agent chosen for amending the soils was a calcium-magnesium (Ca-Mg) composite. The agent content was 5% (equivalent to 50g/kg-soil by dry weight). This calcined mineral was previously considered for the solidification/stabilisation method (Bobeja *et al.* 2012) and is now being considered for the attenuation layer method (Mo *et al.* 2020; Gathuka *et al.* 2021) due to its attenuation performance. By using the results obtained from the experiments, arsenic transport in the attenuation layer was discussed based on a simple case scenario using a one-dimensional advection-dispersion analysis.

2 MATERIALS AND METHODS

2.1 Materials

Decomposed granite soil obtained from a local market in Kyoto was used as the clean parent material. The soil was selected because it is widely distributed in Japan. It was sieved with a 2-mm opening screen to achieve soil particles smaller than 2 mm that were required for the tests. Table 1 summarises its basic properties.

Ca-Mg composite, manufactured by Sumitomo Osaka Cement Co. Ltd., was the stabilising agent. Table 2 summarises its chemical composition. Figure 2 shows the two particle size fractions of the Ca-Mg agent that were considered for the tests, namely <2.0 mm (or fine) and 2.0–9.5 mm (or coarse). The specific surface area (SSA) of the fine and coarse Ca-Mg agent was 3.7 and 3.6 m²/g, respectively. The SSA was measured by the nitrogen adsorption method based on Brunauer-Emmett-Teller (BET) theory.

Table 1. Basic properties of the soil particles smaller than 2 mm.

Parameter	Value	Method of measurement
Particle density	2.67 g/cm ³	JIS A 1202 (2009)
Particle size distribution		JIS A 1204 (2009)
Sand fraction [0.075–2 mm]	85.9%	
Silt fraction [0.005–0.075 mm]	7.7%	
Clay fraction [<0.005 mm]	6.4%	
Chemical composition		X-ray fluorescence analysis—XRF
SiO ₂	45.5%	(EDX-720, Shimadzu)
Fe ₂ O ₃	20.6%	
CaO	6.6%	
Al ₂ O ₃	16.9%	
Others	10.4%	

Table 2. Chemical composition of the Ca-Mg composite. The chemical composition was obtained in accordance with JIS M 8851 (2006).

Chemical composition	Value (wt.%)
Ig. loss (1050 ± 50 °C)	31.18
CaO	44.29
MgO	22.02
Al ₂ O ₃	0.09
Fe ₂ O ₃	0.07
P ₂ O ₅	0.07
Total S	0.01



Figure 2. Appearance of the <2.0 mm (left) and 2.0–9.5 mm (right) Ca-Mg composite.

2.2 Preparation of test specimens

Test specimens of the basic and amended soil were prepared with the following steps. The amended soils had one of two agent sizes (<2.0 or 2.0–9.5 mm). The agent content was 5% (=50 g/kg-soil by dry weight basis). Firstly, the dry soil was poured into a steel bowl. An appropriate amount of agent was added to the soil when preparing the amended soils. Secondly, distilled water was added using the appropriate amount necessary to achieve the optimum water contents of 10.1 and 11.0% for basic and amended soil, respectively, determined by the A-a method of JIS A 1210 (2009). Lastly, mixing was completed using a soil mixer. During the mixing, the machine was temporarily stopped from time to time to scrape the sides and bottom of the bowl.

The specimens were compacted in three equal heights in a steel mould of 6-cm inner diameter and 7-cm height with a degree of compaction of 95%. The compacted specimens had a dry density of ~1.90 g/cm³. After compaction, each specimen was saturated for about 48 h in a vacuum deaerator. The saturated specimen was removed from the steel mould, whilst not causing it any physical damage, and set on the flexible-wall permeameter.

2.3 Column tests

Column tests using flexible-permeameters were applied to evaluate the attenuation performance of basic and amended soils against arsenic (see Figure 3). Arsenic solution with a concentration of 0.1 mg/L was prepared using NaAsO₂ salts and applied as the permeant for the tests. The solution was deaerated for 24 h before it was used in the tests. Arsenic concentrations of geogenic contamination are low but smaller than 10 times the regulatory standard (Ito & Katsumi 2020), hence the use of the relatively low concentration for the tests. Cell pressure of 50 kPa was applied as confining pressure.

The arsenic solution was continuously permeated through the specimens at an average flow rate (*Q*) of 4.5 cm³/h using a peristaltic pump with the temperature of the room set at ~20 °C. Assuming flow is steady-state, the applied flow rate is equivalent to the Darcian velocity ($q = Q/A$, where *A* is the cross-sectional area of the specimen) of 4.42×10^{-5} cm/s. Under these flow conditions, the water residence time ($t_c = V_v/Q$, where *V_v* is the volume of voids in the specimens), which refers to the duration the water is in contact with the solid materials, was estimated to be about 12 h. Effluents were collected for several pore volumes of flow (PVF) and filtered using a membrane with a particle size retention of 0.45 μm.

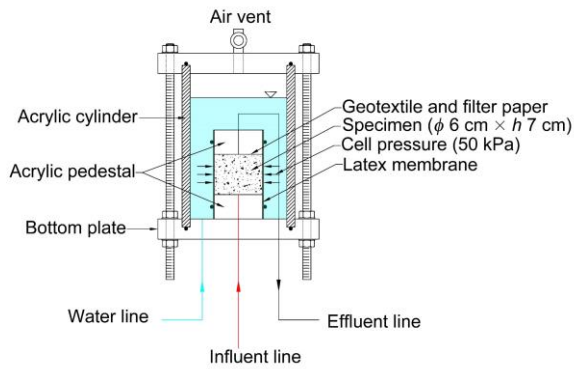


Figure 3. Schematic of flexible-wall permeameter used for column test.

Water chemistry filtered effluents were analysed. The pH and electrical conductivity (EC) were measured by Horiba F-45 pH/EC meter. Oxidation-reduction potential (ORP) was measured by Horiba F-73 pH/ORP meter and converted to E_h . Arsenic concentrations were analysed using a Shimadzu AA-6800 Atomic Absorption Spectrometer. The concentrations of several co-existing ions (Al, Ca, Fe, K, Mg and Na) were measured using an Agilent Technologies ICP-OES 710 Inductively Coupled Plasma Optical Emission Spectrometer.

2.4 Advection-dispersion analyses

The breakthrough curves observed from the column tests were modelled using the following one-dimensional advection-dispersion equation recommended by van Genuchten & Parker (1984) for a finite column accounting for sorption:

$$C = C_0 / 2 \left\{ \begin{array}{l} \text{erfc}[(Pe/4RT)^{1/2}(R-T)] \\ + \exp(Pe) \text{erfc}[(Pe/4RT)^{1/2}(R+T)] \end{array} \right\} \quad (1)$$

where R is the retardation factor, T is the pore volumes of flow, $Pe (= v_s L/D, \text{ where } L \text{ is the column length})$ is the Peclet number, and D (cm^2/s) is the dispersion coefficient. D is typically estimated using the longitudinal dispersivity λ , which is roughly approximated to be $0.1L$ (Gelhar *et al.* 1992), and the molecular diffusion coefficient D_m , which for arsenic is estimated to be $0.905 \times 10^{-5} \text{ cm}^2/\text{s}$ (Lide 2001), i.e. $D = D_m + \lambda|v_s|$.

The modelled breakthrough curves were shifted so that the initial rise in effluent arsenic concentrations coincided with the experimental ones. The most representative R was the one for which the residual sum of the squares becomes minimum.

3 RESULTS

3.1 Arsenic attenuation

Figure 4 shows breakthrough curves obtained from the column tests. The changes in arsenic concentration in effluent were evaluated with respect to the pore volumes of flow. PVF was calculated by dividing the cumulative volume of the effluent collected during the test by the volume of the voids in the specimen. The basic soil is confirmed to have a certain attenuation performance and could reduce the arsenic concentrations. Breakthrough, taken as $C/C_0 > 0.1$, occurred after ~ 150 PVF. Therefore, even if the attenuation layer cannot attenuate the arsenic, the original ground can be expected to attenuate the arsenic naturally.

The amended soils exhibited better arsenic attenuation performance than basic soil. The breakthrough was delayed when

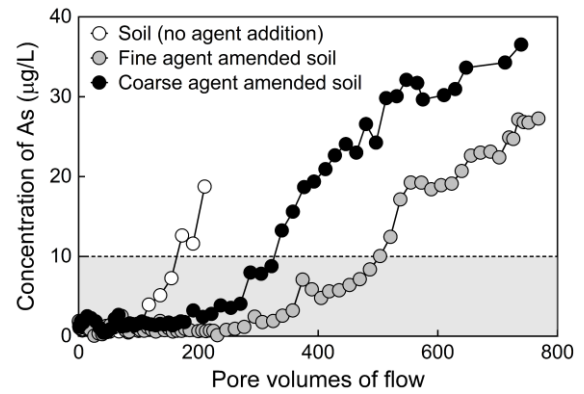


Figure 4. Concentrations of arsenic in effluents during column tests for basic and amended soil. The grey shaded area denotes the acceptable concentrations in Japan.

amended soils were used for the tests. Effluent arsenic concentrations exceeded the regulatory standards after ~ 430 PVF for the coarse agent amended soil, and ~ 860 PVF for the fine agent amended soil. Better performance was noted for the fine agent amended soil because of a large surface area for attenuation reactions provided by the fine agent particle size.

Table 3 summarises the solute transport parameters obtained by following the steps and discussed previously in Section 2.3. D varies between $1.1 \times 10^{-4} \text{ cm}^2/\text{s}$ and $1.2 \times 10^{-4} \text{ cm}^2/\text{s}$, and $Pe = 9.38 \pm 0.02$. Therefore, the advection process will play the dominant role in the solute transport phenomena. Arsenic will be transported by the water flowing in response to the hydraulic gradient (i).

Figure 5 shows the predicted breakthrough curves. Partition coefficient K_d was used as an index for quantifying the attenuation performance of the basic and amended soils. K_d was calculated from R , given that $R = 1 + \rho_d K_d/n$, where n is the porosity of the specimen. K_d was $37.8 \text{ cm}^3/\text{g}$ for the basic soil, $93.2 \text{ cm}^3/\text{g}$ for the coarse agent amended soil, and $150.5 \text{ cm}^3/\text{g}$ for the fine agent amended soil. Results show that coarse and fine agents could increase the attenuation performance of the soil by about 2.5 and 4 times, respectively, even if the agent content was 5%.

Table 3. Summary of the solute transport parameters used in this study.

Agent size	R	K_d (cm^3/g)	D (cm^2/s)	Pe
No agent	320	45.8	1.20×10^{-4}	9.39
<2.0 mm	982	150.5	1.13×10^{-4}	9.36
2.0–9.5 mm	650	93.2	1.17×10^{-4}	9.38

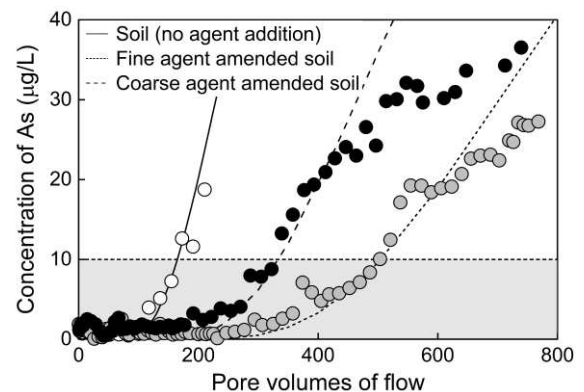


Figure 5. Modelled breakthrough curves from column tests. The grey shaded area denotes the acceptable concentrations in Japan.

3.2 Changes in pH levels

Figure 6 shows that basic soil can increase pH, which might be due to the exchangeable base cations buffering mechanism. As shown in this figure, by introducing the Ca-Mg agent into the soil matrix, the capacity of the soil to increase pH became more effective in the order of decreasing agent particle size. The fine agent offers very high capabilities to increase pH. In the initial stages, fine agent amended soil raised the pH from 6 to 10. However, with increasing volumes of flow, this property gradually decreased. Under similar conditions, the coarse agent amended soil could increase the pH from 6 to 9. Similarly, the pH buffering property of the amended soil decreased with increasing volumes of flow.

4 DISCUSSION

4.1 Expected attenuation mechanism

Figure 7 shows the chemistry of the effluents from the column tests in terms of E_h , pH and concentrations of As. Using an E_h -pH diagram for the As-O-H systems (Lu & Zhu 2010), it is recognised that the dissolved as in the effluents will most likely exist as a negatively charged HAsO_4^{2-} and/or H_2AsO_4^- oxyanion according to their pH. The results imply that the As-applied as NaAsO_2 [or as (III)] was oxidised to As(V). However, it is unclear if it was due to chemical reactions occurring inside the specimens or due to the testing method not considering anaerobic conditions. The As species and their associated compounds have less mobility and toxicity and are stable over a wide pH range (Bothe & Brown 1999).

Considering that the soil contains substantial amounts of Fe and Al-based chemicals (refer to Table 1), it is most likely carrying variable charges (Bolan *et al.* 1999). These variable charges will change according to the pH. When the soil particles have net positively charged surfaces, they will attract these negatively charged oxyanions. Furthermore, the calcium ions released from the soil are expected to react with the dissolved arsenic in the solution to generate low solubility substances such as CaHAsO_3 . McBride (1994) indicated that this mechanism relies on the concentrations of cations released from the soil material.

On the other hand, attenuation mechanisms by the Ca-Mg agent are expected to be mainly related to the precipitation reactions and essentially from the formation of oxides, hydroxides and/or carbonate phases (e.g., Itaya *et al.*, 2013). Itaya *et al.* (2013) showed that low-solubility Ca-As, such as $\text{CaHAsO}_4(\text{H}_2\text{O})$, will be generated during the Ca-Mg agent interactions with the dissolved arsenic.

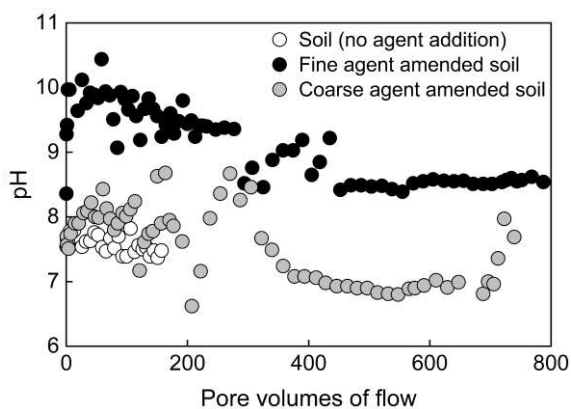


Figure 6. Trends in pH of effluent during column tests.

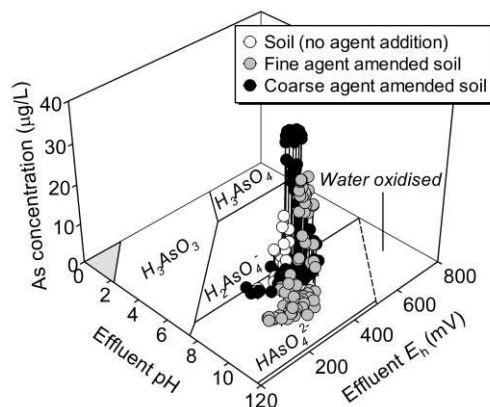


Figure 7. Chemistry of effluents in terms of their E_h , pH and concentration of As. The E_h -pH diagram is for the As-O-H system (edited from Lu and Zhu, 2010). The grey shaded area denotes the solid phase.

Therefore, if the attenuation layer is created with amended soil, both the soil and the Ca-Mg agent will function to attenuate against the arsenic. Therefore, the mechanisms mentioned above might co-occur. Furthermore, the magnitude of these mechanisms and how long they will continue to occur will depend on time and the materials. The results suggest that low-solubility Ca-As will be released because of the agent. This mechanism is expected to dominate during the buffering of the metal concentrations.

4.2 Arsenic transport in attenuation layer

A simple evaluation under assumed conditions is here discussed. The attenuation layer of thickness L is assumed to be saturated, and has a hydraulic conductivity (k) of 1×10^{-4} cm/s. Assuming seepage is steady-state, hydraulic gradient ($I = h_w/L$, where h_w is the leachate head) of 1, soil properties (f , k , K_d , D) are homogeneous and constant over time, and suction existing at the bottom of the layer is ignored, the effluent arsenic concentration exiting the layer [$C(L,t)$], with time (t) was estimated using the advection-dispersion equation by considering the initial condition ($C(x,0) = 0$) and boundary conditions ($C(0,t) = C_0$; $C(L,t) = 0$). Breakthrough time (t_b) in years was calculated by considering an assumed percolation (P) of 500 mm/year and cumulative infiltration ($CI = T \cdot L \cdot f$), i.e. $t_b = CI/P$.

Figure 8 shows estimates of breakthrough time for various layer thicknesses, given by the relationship of As concentration at exit of layer [$C(L,t)$] to As concentration at inlet [$C(0,t) = 0.1$ mg/L] ratios i.e. C/C_0 . As shown in the figure, for a 30 cm-thick layer of sandy soil amended with 5% coarse agent or fine agent, it will take approximately 56 and 91 years respectively for the As values to exceed the limits set by SCCL. Although the fine agent can better improve the As attenuation function of soil, the coarse agent can also be applied as a material for the attenuation layer—less, but long-term reactivity is expected. The breakthrough time can be increased by employing more agent, reducing the percolation into an embankment, installing a thicker layer, and so on

5 CONCLUSIONS

The main achievements obtained in this study can be summarised as follows:

- The amended soils have better arsenic attenuation performance than basic soil. Using partition coefficient K_d as an index, the soil's attenuation performance could be improved by at least two times, which is the case of using a 2.0-9.5 mm agent to amend the soil. The effect of agent

addition on attenuation performance of soil is much more significant when <2.0 mm agent is used to amend the soil.

- Ca-Mg agent addition could significantly improve the capability of natural soil to increase pH. Soil amended with <2.0 mm agent exhibited the highest pH buffering capacity. In the initial stages, it could increase pH levels from 6 to 10, but this property decreased with continuous permeation.
- The precipitation mechanism is expected to be one of the main arsenic attenuation mechanisms when using amended soil. The precipitates generated are less mobile and toxic, and stable over a wide pH range.

6 ACKNOWLEDGEMENTS

The authors would like to express their thanks to Mr Kenji Kuninishi and Mr Shintaro Hayashi (Sumitomo Osaka Cement, Co., Ltd.) for providing the Ca-Mg composite used as the agent in this study.

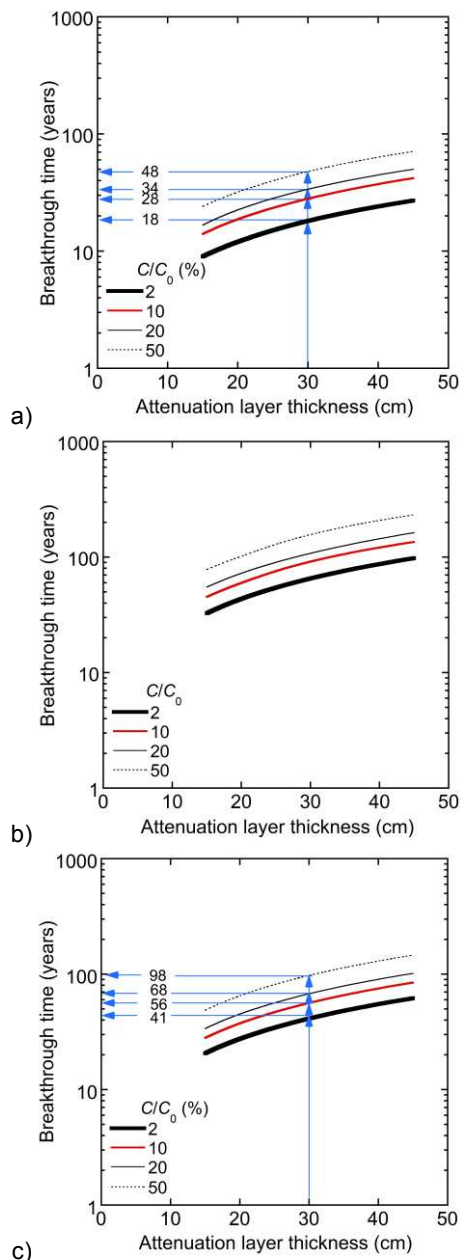


Figure 8. Breakthrough times for (a) the basic soil, (b) the fine agent amended soil, and (c) the coarse agent amended soil.

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