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Geoenvironmental assessment of Ariake clay for its potential use as a landfill barrier material

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Abstract: An assessment of the potential use of a typical Japanese marine clay, Ariake clay, as a soil barrier material in landfills was presented. Two types of heavy metals, Cd^{2+} and Pb^{2+}, were selected as key contaminants for laboratory diffusion test. Using commercially available contaminant transport software, the effective diffusion coefficients and partition coefficients of both Cd^{2+} and Pb^{2+} were back-calculated from the results of the diffusion test. The effective diffusion coefficients were found to be consistent with previously published values, indicating that the presented testing method was acceptable. The high retained percentage of Cd^{2+} and Pb^{2+} in the Ariake clay specimen indicated that the Ariake clay might be used as a good barrier material in landfills. With the derived effective diffusion coefficients and partition coefficients, the influences of Cd^{2+} and Pb^{2+} on the underlying aquifer in an assumed landfill were numerically assessed using the 1D diffusion transport theory.

Key words: Ariake clay; barrier; landfill; heavy metal

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有明粘土作为垃圾填埋场隔离层的环境岩土工程评价

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摘　要：通过室内实验并应用现有的计算软件，反算出在文中所述的试验条件下，两种重金属离子（Cd2+, Pb2+）在有明粘土中的有效扩散系数和分配系数。在此基础之上，对假定的垃圾填埋场于其下卧含水层的水质影响进行了计算分析和评估。

关键词：有明粘土；隔离层；填埋场；重金属

0　Introduction

Heavy metals such as Cd^{2+} and Pb^{2+} are commonly found in leachate from landfills. Due to the toxicity and migration, these heavy metals may cause impact to the aquifer underlying the landfill and thereby may pose potential risk to human health. An effective way for reducing this contamination risk is to use soil barriers (e.g., natural clayey soils) with low permeability to control the migration of heavy metals. To evaluate the application of clays for barrier materials, some key transport parameters, such as effective diffusion coefficients and partition coefficients of heavy metals, need to be evaluated. To determine these parameters, usually a column test, either diffusion test or diffusion-advection test, is required (Rowe et al. 1988; Shackelford and Daniel 1991a).

There is a tendency in Japan to build waste sites in costal areas, and the regional marine clays of interest may potentially be used as soil barrier materials. Du et al. (2000) indicated that the Ariake clay, which is a typical regional marine clay located at the Kyushu of Japan, seemed to be a good barrier material because of its high sorption capacity and low effective diffusion coefficient. However, relatively few literatures have been published concerning assessment on potential use of Ariake clay as a barrier material to reduce contamination risk of heavy metals. As a result, uncertainty in using Ariake clay as landfill barrier exists, especially as an industrial landfill barrier where heavy metals are commonly encountered.

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The objective of this study is to assess the potential utilization of Ariake clay as a barrier material in industrial landfills. For this purpose, a laboratory diffusion test and analysis method for determining the effective diffusion coefficients of two key contaminants, Cd^{2+} and Pb^{2+} in the Ariake clay are presented. The test data are applied for evaluating the impacts of Cd^{2+} and Pb^{2+} in an assumed landfill on the water quality of the underlying aquifer.

1 Soil description

Ariake clay is a typical marine sedimentation product accumulated in the Ariake Bay over the past 10,000 years (Ariake Bay Research Group, 1965). The details of its geological history and depositional geochemistry are provided by Ariake Bay Research Group (1965) and Ohtsubo et al. (1995), respectively. In this study, the Ariake clay was sampled at about 3 m in depth from the ground surface at the Kawasoe Machi, Saga Prefecture, Kyushu of Japan. Some geotechnical and chemical properties are shown in Table 1.

Table 1 Geotechnical and chemical properties of Ariake clay

Quantity	Value
Specific gravity, G_s	2.68
Natural water content, w_n /%	173
Liquid limit, w_L /%	115
pH	7.87
Clay particle fraction (<2μm) /%	46
Silt particle fraction /%	49
Sand particle fraction /%	5
Primary clay mineral	smectite
CEC (meq/100 g)	34
Specific surface area /($m^2 \cdot g^{-1}$)	69
Ignition loss /%	10.2

2 Testing method

The key chemical species were selected as cadmium (Cd) and lead (Pb) because these two heavy metals are commonly encountered in industrial landfills. The synthetic leachate was comprised of a solution containing $CdCl_2$, $PbCl_2$ and KCl. Prior to the diffusion test, the Ariake clay specimen was prepared by consolidation. About 1.7 kg of disturbed Ariake clay at the natural water content was poured into an acrylic cylinder with inner diameter of 10 cm (Fig. 1). A porous plate with thickness of 0.4 cm and porosity of about 0.42 was placed above the soil. A perforated stainless steel pate with thickness of 0.4 cm and open space coverage (open space volume/total volume) of 80 % was placed above the porous plate, and was connected with a stainless steel rod used to transfer a vertical pressure of 57 kPa that was applied from a device similar to an odometer test apparatus. This consolidation pressure is thought to be slightly higher than the preconsolidation pressure of the corresponding undisturbed soil. To prevent drying of the clay surface and to keep the pore fluid close to the original one, about 10 mL of pore water extracted from the other samples of the same clay were added on the clay periodically. During the consolidation, the valve for the plastic tubing at the base was kept open to drain the pore water. After consolidation, which lasted for about 1 month, the valve was closed and the residue pore fluid above the perforated steel plate was removed immediately. The synthetic leachate was introduced into the cylinder and the diffusion test was started. The properties of the soil specimen and the solution used for the diffusion test are listed in Table 2. These parameters will be used for the analysis of the results of the diffusion test as presented later in this paper.

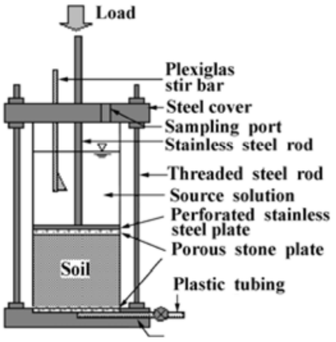

Fig. 1 Schematic of the diffusion test apparatus

Table 2 Properties of soil specimen and solution

Thickness, H /cm	10.2
Dry density, ρ_d /($g \cdot cm^{-3}$)	0.77
Porosity, n	0.71
Water content /%	91
Saturation degree/%	100
Hydraulic conductivity, $k \times 10^{-9}$ ($m \cdot s^{-1}$)	1.7
Height of solution, H_f /cm	10.2
Initial concentration, C_0 / ($mg \cdot L^{-1}$)	61.8(Cd^{2+}), 22.1(Pb^{2+})
pH of solution before diffusion test	5.88
pH of solution after diffusion test	7.83
Solution collected, $q_c \times 10^{-10}$ (per area per unit time, m/s)	3.5
Duration of test/d	54

Throughout the test, the concentrations of Cd^{2+} and Pb^{2+} in the source reservoir were periodically monitored by withdrawing 1-mL samples from the solution through the sampling port at three-day intervals (see Fig. 1). The same volume of distilled water as that of the sampled solution was added to the source reservoir to keep the constant head of source solution. The dilution caused by the addition of the distilled water was automatically taken into account as explained subsequently. At the completion of the diffusion, which lasted for 54 d, the solution in the cylinder was drained away and the apparatus was disassembled. The soil specimen was extruded and sliced into seven sublayers using a stainless steel thread. The pore water in each sublayer was obtained by squeezing the soil at a pressure of about 12.8 kPa for approximately 12 h. The concentrations of Cd^{2+} and Pb^{2+} in the squeezed pore water were analyzed using a HITACHI Z-6000 atomic adsorption spectrophotometer.

3　Analysis of results and discussions

The migration of Cd^{2+} and Pb^{2+} in the Ariake clay was evaluated considering the one-dimensional governing equation for diffusive transport in saturated soil as expressed by the Fick' second law, or

$$(n+\rho_d K_P)\frac{\partial C}{\partial t}=nD_e\frac{\partial^2 C}{\partial x^2}\quad ,\tag{1}$$

where ρ_d = the dry density of the soil, n = the porosity of the soil, C = the concentration of contaminant at the point and time of interest, t = the time, x = the distance (in this study $x = 0$ represents a plane that is the interface between the source solution and the Ariake clay), D_e = the effective diffusion coefficient defined by Shackelford and Daniel (1991a) and K_p = the partition coefficient.

The upper boundary condition imposed by the source solution was the finite mass condition or expressed as

$$C(t)=C_0-\frac{1}{H_f}\int_0^t f_T(\tau)\mathrm{d}\tau-\frac{q_c}{H_f}\int_0^t C(\tau)\mathrm{d}\tau\quad .\tag{2}$$

The base was impermeable such that the lower boundary was modelled as a zero-flux boundary condition, or

$$f_B(t)=-nD_e\left(\frac{\partial C(t)}{\partial x}\right)_B=0\quad ,\tag{3}$$

where C_0 = the initial concentration of Cd^{2+} or Pb^{2+}, H = the constant height of source solution, q_c = the volume of fluid per unit cross-sectional area of the soil per unit time removed from the source solution and replaced bythe soil as given by

$$f_T=-nD_e\frac{\partial C}{\partial x}\quad ,\tag{4}$$

distilled water, f_T = the mass flux from the source into and $f_{B(t)}$ = the mass flux entering into the base at specific time t. A solution to Eqs. (1) – (4) is presented in the form of the commercial software program, *Pollute V* 6.3 (Rowe and Booker, 1994) which is based on the finite-layer technique (Rowe and Booker, 1985). This solution is done by the following steps: 1) simplifying the governing and boundary condition equations by performing Laplace transform and solving the transformed governing equation analytically, and 2) inverting the Laplace transform numerically. Unlike finite element and finite difference formulations, *Pollute V* 6.3 does not require the use of a "time-marching" procedure. Using various combinations of D_e, and the partition coefficient, K_p, theoretical concentration – time curves for the source solution were generated and then compared with the experimental data. The values of D_e and K_p which together give the best fit "by eye" to the experimental data, were selected as the back-calculated values. Rowe and Booker (1985) have shown that only a single combination of D_e and K_p provides the best fit. All of the input parameters required for *Pollute V* 6.3 are summarized in Table 2. The experimentally measured data and best-fit curves for concentration variation with time in the source solution are shown in Figs 2 – 3. The back-calculated values of D_e and K_p for Cd^{2+} and Pb^{2+} are 4×10-10 m^2/s and 2 mL/g, and 4×10-10 m^2/s and 100 mL/g, respectively. The values of effective diffusion coefficients are well in the range of values reported in the literature for Cd^{2+} of 3.0×10-10 m^2/s to ~ 4.2×10-10 m^2/s (Shackelford and Daniel, 1991b), indicating the testing method used in this study is reasonable.

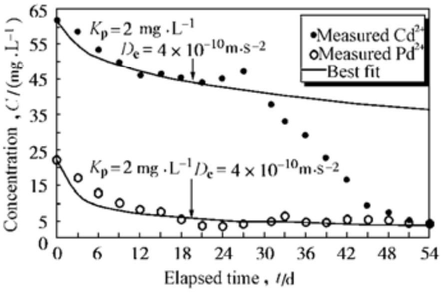

Fig. 2 Concentrations of Cd^{2+} and Pb^{2+} versus time in the source solution

From Fig. 3, it can be seen that the predicted curve for Pb^{2+} fits the experimental data better relative to that for Cd^{2+}. For Cd^{2+}, after about first 31 d, a substantial drop in the concentration was observed by the end of the diffusion test. However, only the data before the elapsed time of 31 d were used for back-calculating the D_e and K_p for Cd^{2+}. This considerable drop of concentration of Cd^{2+} may be partly due to the increase of pH in the source solution during the diffusion test. Before starting the diffusion test, the measured pH of the solution was 5.88 whereas it increased to 7.83 at the end of the test (see Table 2). It may be inferred that during the test (probably after 31 d), the pH of the solution may have increased to values (e.g., pH >7) that are favorable for the formation of hydroxy species of cadmium (e.g., $Cd(OH)_2$), and thereby the concentrations of Cd^{2+} decreased considerably. The modeling process used in this study does not take into account such a geochemical change and therefore the experimental concentrations were lower than the predicted concentrations after from 31 d of testing. Yong et al. (1992) indicated that that hydroxy species for Cd only began to form at pH ≥7 and at pH ≥4 for Pb. This difference may explain why a substantial drop in concentration for Cd^{2+} was observed in the source solution whereas no such drop was observed for Pb^{2+}. This drop in concentration for Cd^{2+} also implies that the sorption capacity of the Ariake clay for Cd^{2+} may have increased during a long-term period, which is a beneficial aspect associated with the use of Ariake clay as a barrier in industrial landfills.

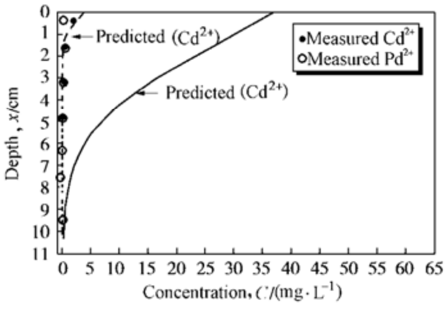

Fig. 3 Concentrations of Cd^{2+} and Pb^{2+} versus soil depth

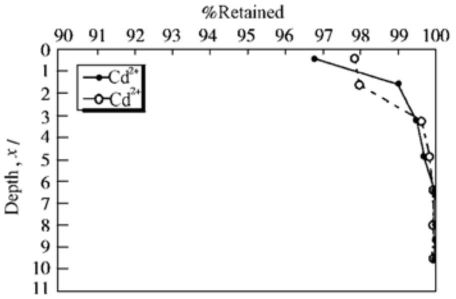

Fig. 4 Retained percent of Cd^{2+} and Pb^{2+} versus depth in the soil

The retained percents of Cd^{2+} and Pb^{2+} at different depths of specimen were calculated using

$$\%\ \text{Retained} = (1 - C_0/C')\times 100 \qquad (5)$$

where C_0 is the initial concentration and C' is the measured concentration in pore fluid at each depth of specimen. The retained percents of Cd^{2+} and Pb^{2+} distributed in the specimen are depicted in Fig. 4. It can be seen that both Cd^{2+} and Pb^{2+} have been greatly retained in the soil, with retained percents being larger than 95%. At depths deeper than 3 cm, the retained percents even exceeded 99%. The retained percent of Cd^{2+} even reached 99.96% at the depth of about 9.5 cm. Therefore it is concluded that Ariake clay has much high ability for retaining Cd^{2+} and Pb^{2+}, indicating that it may be used as a good barrier material for attenuating heavy metals in industrial landfills.

4 Implication of test data to practice

In the construction of modern landfills, the hydraulic conductivity of clay barriers is restricted to low value. For example, the US Environmental Protection Agency (US EPA) requires the hydraulic conductivity of clay barrier in municipal solid waste landfills should be less than 10^{-9} m/s. The main possible reason for restricting the hydraulic conductivity of clay barriers to low value is mainly to ensure that diffusion of contaminants in clay barriers is predominated over advection. Evidences have shown that when the clay barrier has low hydraulic conductivity or negligible hydraulic gradient, the diffusion of contaminants through the clay barrier would be dominated over the advection (Cooks and Quigley, 1984; Johnson et al., 1986). Under this condition, the effective diffusion coefficient would be a more important parameter relative to the hydraulic conductivity in a performance-based design of clay barriers in landfills. To better illustrate this aspect, the laboratory test data are applied to such a field scenario: a landfill with size of 100 m × 100 m underlain with an Ariake clay barrier with thickness of 5 m and the hydraulic conductivity of 1.7×10^{-9} m/s (the same as that in the diffusion test). A confined aquifer is underlying the Ariake clay barrier (Fig. 5). The geoenvironmental properties of the Ariake clay used in the landfill is assumed to be identical to the one used in the laboratory diffusion test. In fact, this clay barrier corresponds to the regulatory Japanese standard

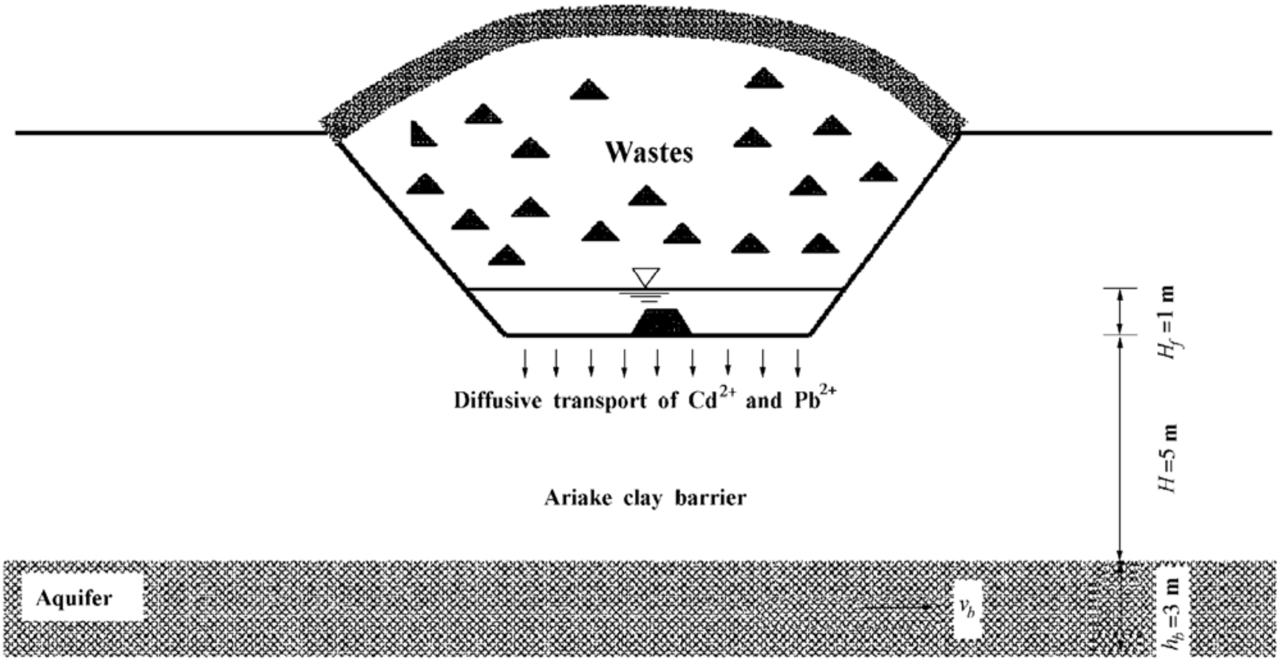

Fig. 5 Assumed field scenario: a landfill with a Ariake clay barrier and an underlying aquifer

landfill barrier system, Type-A, prescribed by the Japanese Ministry of Healthy and Welfare. Suppose there are heavy metals like Cd^{2+} and Pb^{2+} in the landfill and their concentrations reach peak (C_0) when the landfill is closed. Due to the migration of Cd^{2+} and Pb^{2+} from the landfill, during the post-closure there is a potential risk that the groundwater quality in the aquifer may be contaminated by Cd^{2+} and Pb^{2+}. Therefore an assessment of impacts of the landfill on the underlying aquifer is necessary, and may provide important information on the monitoring problem during the post-closure. In this study, the impacts are evaluated in terms of the variation of concentrations of Cd^{2+} and Pb^{2+} with time in the aquifer. The evaluation length is assumed to be 1000 years, which is enough for most practical situations. Because evidence has shown that in most well-constructed landfills, the hydraulic gradient across the clay barrier is very low (< 0.02) (Rowe et al., 1995), it is reasonable to assume that diffusion of contaminants in the Ariake clay barrier is dominated over advection. As a result, it is assumed that advection is neglected, and only the diffusion is of concerning. The diffusive transport of Cd^{2+} and Pb^{2+} in the Ariake clay barrier is modeled using the governing Eq. (1). The upper boundary is imposed by the leachate in the landfill, representing a finite mass condition, which is thought to be realistic for the analysis of impacts within the post-closure (Rowe, 1988), or

$$C(t)=C_0-\frac{1}{H_f}\int_0^t f_T(\tau)\mathrm{d}\tau \quad , \tag{6}$$

where H_f = equivalent height of leachate in the landfill, which only corresponds to the mass of contaminants that transport into the Ariake clay via diffusion (excluding the mass collected by the Leachate Collection System). The other parameters are the same as those defined in Eq. (2). The lower boundary is imposed by the confined aquifer. It is assumed that contaminants entering the aquifer will be mixed uniformly in the thickness h_b, and transport of contaminant in the aquifer with porosity of n_b is predominated by advection. The horizontal mechanical dispersivity α_H in the aquifer is assumed to be zero. This assumption may give the most conservatively predicted impact (Rowe and Booker, 1992). Based on the conservation of mass, the concentration in the aquifer at time of interest, $C_b(t)$, is then expressed by

$$C_b(t)=\int_0^t \frac{f_b(t)}{n_b h_b}\mathrm{d}t-\int_0^t \frac{C_b v_b(t)}{n_b L}\mathrm{d}t \quad , \tag{7}$$

where v_b = the horizontal Darcy velocity in the aquifer and L = the length of landfill in the direction of the velocity v_b, and f_b = the mass flux of contaminant entering the aquifer. The program, *Pollute V* 6.3 is used to conduct the analysis. All of the input parameters are summarized in Table 3. For simplicity, the effective diffusion coefficients, partition coefficient and other geotechnical properties listed in Table 3 (e.g., porosity and dry density) are assumed to be identical to those of the laboratory diffusion test presented earlier in this paper. The background concentrations of Cd^{2+} and Pb^{2+} in the aquifer are assumed to be zero. The horizontal Darcy velocity is assumed to be 25 m/yr according to the unpublished data from the Saga Prefecture, Japan.

Table 3 Input parameters for numerical analysis

Dry density, ρ_d/(g • cm^{-3})	0.77
Porosity, n	0.71*, 0.4**
Partition coefficient, K_p/(mL • g^{-1})	2 (Cd^{2+}), 100 (Pb^{2+})
Hydraulic conductivity, k /(m • s^{-1})	1.7×10^{-9}
Leachate heigt, H_f /m	1.0
D_e/(m^2 • s^{-1})	4×10^{-10}
C_0 of Cd^{2+} /(mg • L^{-1})	60
C_0 of Pb^{2+} /(mg • L^{-1})	20
Thickness /m	5.0*, 3.0**
Width of landfill, W/m	100
Length of landfill, L /m	100
$v_{b/}$ (m • yr^{-1})	25

*Ariake clay liner **aquifer

The calculated concentrations of Cd^{2+} and Pb^{2+} in the aquifer are shown in Fig. 6. The dashed line in Fig. 6 represents the maximum permissible concentration $C_{\max}$ ($C_{\max} = 0.01$ mg • L^{-1}) for Cd^{2+} and Pb^{2+} regulated by the Japanese Environmental Quality Standards for Groundwater Pollution. It can be seen from Fig. 6 that after the first 350 years, the concentration of Cd^{2+} in the aquifer will exceed $C_{\max}$, indicating an undesirable condition. However, for Pb^{2+}, its concentration is much lower than $C_{\max}$, indicating that the impact of Pb^{2+} is negligible.

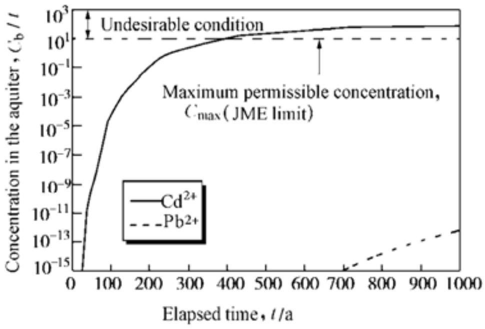

Fig. 6 Calculated concentrations of Cd^{2+} and Pb^{2+} in the aquifer versus time

5 Conclusions

This study presents the results of a laboratory diffusion test for assessment of potential use of Ariake clay as a barrier material in industrial landfills. The effective diffusion coefficient and partition coefficient of two key chemical species, Cd^{2+} and Pb^{2+}, were back-calculated from the test results. The results of the test were applied to a field scenario in which the Ariake clay is used as the landfill barrier. The impacts of Cd^{2+} and Pb^{2+} in the landfill on the water quality of the underlying aquifer were evaluated using the 1D diffusion transport theory. The following conclusions can be drawn:

(1) The diffusion coefficients of Cd^{2+} and Pb^{2+} derived from the lab test are consistent well with the previously published value, indicating that the testing method presented in this study is reasonable.

(2) The highly retained percents of Cd^{2+} and Pb^{2+} on the Ariake clay indicates that Ariake clay may be used as a good barrier material to attenuate contamination of heavv metals in landfills.

(3) In the diffusion test, it is found that the sorption ability of the Ariake clay for Cd^{2+} increased, whereas for Pb^{2+} no similar phenomenon took place. The main reason may be due to the geochemistry change in the test.

(4) With the effective diffusion coefficients and partition coefficients derived from the laboratory diffusion test, the impacts of the assumed landfill on the water quality in the aquifer within 1000-year post-closure were calculated.

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博士简介扩大刊登范围启事

本刊开辟的博士简介栏目在广大读者支持下，自 1995 年以来，已连续 10 年报导我国岩土工程界博士学位获得者简况。今后仍将在每年第 12 期刊登博士简介，凡在海内外取得岩土工程博士学位的我国学者，均属报导范围。简介文稿请在当年 10 月 30 日（截稿）前寄来南京本部。

致本刊编委及各位审稿人

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