

MICROSCOPIC RANGE OF IMMOBILIZATION BETWEEN HEAVY METALS AND AMENDMENT IN SOIL THROUGH WATER MIGRATION

Shouhei Ogawa¹, Masahiko Katoh² and Takeshi Sato³

¹ Graduate School of Engineering, Gifu University, Japan; ^{2,3} Faculty of Engineering, Gifu University, Japan

ABSTRACT: In order to identify the microscopic range of immobilization in soil, the pot test was conducted to evaluate the distance that lead (Pb) and antimony (Sb) transport through water migration and the transport phases during immobilization. The amount of amendment required to immobilize Pb and Sb was investigated on the basis of the microscopic range of immobilization. The results clearly showed that Pb and Sb were transported a maximum of 5 mm and 6 mm, respectively, through water migration which corresponded to precipitation for one month. Both metals were accumulated 1 mm from contaminated soil. The primary Pb transport phase was sorbed on Fe/Mn oxide, while that of Sb was water-soluble. The amount of amendment required for Pb and Sb immobilization was estimated to be 4.62% (w/w) from the microscopic range of immobilization (1 mm).

Keywords: Amendment ratio, Antimony, Immobilization, Lead, Water migration

1. INTRODUCTION

Removing heavy metals, which are highly persistent in soil, is difficult compared to other contaminants, such as oil and volatile organic compounds that readily decompose in soil [1]. Therefore, soil remediation techniques, including excavation and transportation to landfill sites, that can easily reduce risk are often applied to heavy-metal-contaminated soil. However, in addition to construction costs, excavation and transportation to landfills incur conveyance and management costs, which in all probability would lead to an increase in total cost [2].

Chemical immobilization is one of the most economical in situ techniques because the soil can be reused at the site after treatment [3]. The addition of an immobilization amendment to the soil can transform a heavy metal into chemically stable phases and thus reduce solubility, which prevents leaching of heavy metal into groundwater. In this context, immobilization has been investigated for lead (Pb-) and antimony (Sb-) contaminated soil from a shooting range, where contamination is more extensive and asset value is very low [4], [5]. Pb can be immobilized by an apatite, precipitating Pb phosphate minerals, such as pyromorphite [6], and Sb can be immobilized by an iron compound, particularly iron hydroxide, forming surface complexation [7]. However, if the amount of the amendment is not sufficient to immobilize Pb and Sb completely, the treatment can potentially lead to Pb and Sb redissolving in the soil [7].

The required amount of immobilization amendment in the soil is calculated on the basis of

the amendment's maximum sorption capacity estimated by a sorption experiment in solution [8]. However, Pb and Sb would be unstable in the soil unless stable phases are formed, even though the amount of amendment is sufficient to immobilize them. This is because heavy metal transport through water migration, and subsequent contact and reaction with the immobilization amendment, would be essential before the stable phases are formed. Thus, to calculate the required amount of immobilization amendment, it is necessary to identify the microscopic range of Pb and Sb immobilization, i.e., the transport distance of Pb and Sb through water migration during the period of immobilization.

Heavy metals in soil particles would be transported through water migration, repeating various processes, such as ion exchange, precipitation and dissolution, sorption and desorption to soil minerals and organic matters, and colloidal transportation [9], [10]. In addition, heavy metal phases would be altered during transportation. Therefore, to identify the microscopic range of Pb and Sb immobilization, Pb and Sb phases during transport should be taken into consideration.

As stated above, elucidation of the microscopic range of Pb and Sb immobilization during the immobilization period is also necessary to calculate the amount of amendment required for reliable immobilization. However, the required amount of immobilization amendment has not been evaluated in relation to the microscopic range of Pb and Sb immobilization, although heavy metal transport in the soil has been well investigated [10] – [13].

The present study represented Pb and Sb

microscopic transport through water migration in soil from a shooting range by using a compartmentalized pot to represent water flow in unsaturated soil. The objective of this study was to evaluate the distance of Pb and Sb transport and transport phases during the immobilization period, and to identify the microscopic range of their immobilization in the soil. In addition, the required amount of immobilization amendment was investigated on the basis of the microscopic range of immobilization.

2. MATERIALS AND METHODS

2.1 Study Area

This study was conducted using soil from a shooting range located at 35° 28' 6" N and 137° 29' 2" E in Tajimi, Gifu Prefecture, Japan. The mean annual precipitation and temperature at the shooting range are 2200 mm and 14 °C, respectively.

2.2 Soil Preparation and Characterization

The Pb- and Sb-contaminated soil was collected from depths of 5–15 cm. The contaminated soil sample was air-dried, passed through a 2 mm sieve, and used for chemical analysis and the water migration experiment. Table 1 shows the physicochemical properties of the soil samples. Total Pb and Sb content of the

contaminated soil was 19700 and 261 mg/kg, respectively. The water-soluble Pb of the contaminated soil was 95.7 mg/kg, which is 1000 times greater than the standard (0.1 mg/kg) for soil in Japan. The physicochemical properties of non-contaminated soil collected outside the contaminated site were similar to those of the contaminated soil, except for Pb and Sb. The non-contaminated soil was passed through a 1 mm sieve prior to the water migration experiment. Commercial Andosol was used as cultivation soil.

2.3 Materials Preparation and Characterization

Apatite and iron hydroxide were used as Pb and Sb immobilization amendments, respectively. The apatite was synthesized from gypsum and diammonium hydrogen phosphate [14]. Apatite can immobilize Pb in soil through the precipitation of Pb phosphate minerals such as pyromorphite. The iron hydroxide was synthesized from iron nitrate hydrate [15]. Iron hydroxide can immobilize Sb in soil through the formation of an inner-sphere complex. Both amendments were passed through a 0.425 mm sieve, and equal amounts were well mixed prior to use. Table 1 shows the physicochemical properties of the amendments.

2.4 Water Migration Experiment

Figure 1 is a schematic of the experiment. The

Table 1 Physicochemical properties of the soil and immobilization amendment

Soils and Immobilization amendment	pH _(H₂O)	Particles density [g/cm ³]	Total						Water-soluble	
			C	Fe	Ca	P	Pb	Sb	Pb	Sb
			-----			[mg/kg]			-----	
Contaminated soil	7.18	2.62	51000	33700	7570	386	19700	261	95.7	7.6
Non-contaminated soil	6.06	N.A.*	14300	25500	1390	39.3	11.3	1.7	0.1 >	0.1 >
Cultivation soil	5.56	N.A.	71200	40100	5650	1710	58.3	5.8	0.1 >	0.1 >
Immobilization amendment	7.70	3.29	N.A.	254000	181000	45500	14.8	3.5	0.1 >	0.1 >

* : Not Analyzed

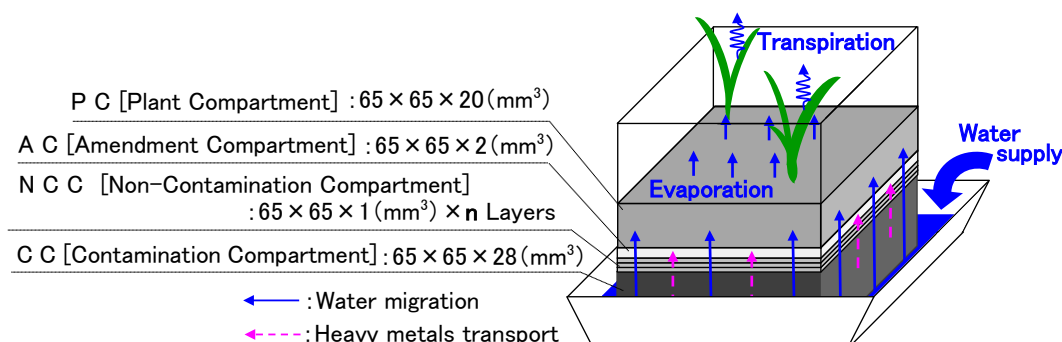


Fig. 1 Water migration schematic

experimental pot was comprised of four layered compartments: contamination, non-contamination, amendment, and plant compartments (CC, NCC, AC, and PC, respectively). NCC was composed of five or ten 1-mm-thick layers (5 mm NC-Pot and 10 mm NC-Pot, respectively). Then, 107.50, 4.68, 8.60, and 100.00 g of contaminated soil, non-contaminated soil, amendment, and cultivation soil, respectively, were added to each layer. The layers were separated using a 25- μ m nylon mesh. Ultrapure water was added to maintain 50% of the water-holding capacity of each soil or amendment. After adding 5 mL of solution containing nitrogen, phosphorus, and potassium as fertilizer at 10 mg/kg to the PC, two Guinea grass (*Panicum maximum* J.) plants were grown in the compartment. Guinea grass can tolerate Pb toxicity. The plants were grown at room temperature (25 °C). This experimental design represented water flow in unsaturated soil. The bottom of the pot had 25 holes with a diameter of 5 mm. The plants were watered the amount corresponding to the lost weight of pot from the last water supply from the bottom at 2 or 3 times a week (Fig. 1). The total water supply was 170 mm, which corresponded to precipitation for one month at the shooting range site. The period of immobilization was defined as one month. After the experiment, the NCC soil samples were collected, air-dried, and passed through a 2 or 0.425 mm sieve.

2.5 Methodology

The pH value of the soil sample was measured in ultrapure water using a pH meter (MM-60R, DKK-TOA Co., Japan). The total carbon (C) content of the soil sample was determined using an NC analyzer (MT-6, Yanaco New Science Inc., Japan). The water-soluble Pb and Sb in the soil was extracted using ultra-pure water (1:10 soil:solution ratio). The total Pb, Sb, calcium (Ca), iron (Fe), and phosphorus (P) content of the soil was determined by acid digestion with 14.5 M HNO₃ and 12 M HCl using a microwave. Sequential extraction was performed on the soil sample following the procedure described in [16]. Then, 1.0 g of soil that had been passed through a 0.425 mm sieve was extracted with 25 mL of a 1 M MgCl₂ solution (exchangeable). The soil remaining after the first extraction procedure was extracted with 25 mL of a 1 M sodium acetate solution with pH 5 (carbonate). The soil remaining after the second extraction was further extracted with 20 mL of 0.04 M NH₂OH-HCl in 25% (v/v) HOAc in a 95 °C water bath with occasional agitation (bound to Fe/Mn oxide). The soil remaining after the third extraction was extracted with 3 mL 0.02 M HNO₃ and 5 mL 30% H₂O₂ solution in an 85 °C water bath with occasional

agitation. After extraction for 3 h, 5 mL 3.2 M NH₄OAc in 20% (v/v) HNO₃ was added and the soil was shaken for 20 min (bound to organic matter). The remaining soil was digested with 5 mL HNO₃ and 2 mL HCl using a microwave oven (residual) and diluted in a 50 mL measuring flask. All the extracted or digested solutions were passed through a 0.45 μ m filter, and analyzed to determine the elemental concentrations by inductively coupled plasma optical emission spectrometry (ICP-OES; ULTIMA2, HORIBA Ltd., Japan) or inductively coupled plasma mass spectrometry (ICP-MS; 7500cx, Agilent Technologies Inc., USA).

3. RESULTS AND DISCUSSION

3.1 Distance of Pb and Sb Transport through Water Migration

Figure 2 shows the levels of total Pb and Sb in each NCC layer in the 5 mm NC-Pot. The levels of total Pb in all the layers were more than twice as high as those in the original non-contaminated soil, indicating that Pb was transported 5 mm from CC in the NCC through water migration during the immobilization period. The highest levels of total Pb were found in the 1 and 4 mm layers of NCC, and the lowest was in the 5 mm layer. The levels of total Sb in all the layers were also more than twice as high as those in the original non-contaminated soil. The highest level of total Sb was found in the 1 mm layer, and the lowest was in the 5 mm layer. This result indicated that Sb was also transported 5 mm. Moreover, it is reasonable to conclude that the levels of transported Pb and Sb would decrease with the increase in transport distance because the lowest level of total Pb and Sb was found in the 5 mm

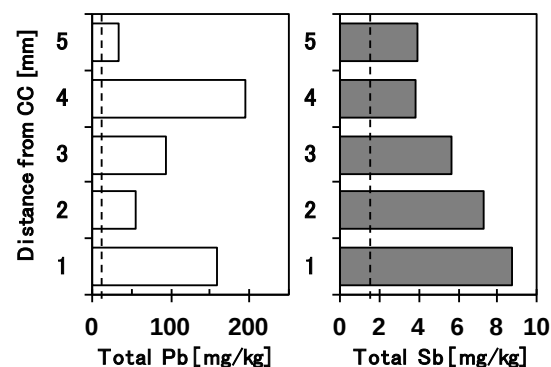


Fig. 2 Total heavy metal levels in NCC (5mm NC-Pot). Broken lines indicate the background levels in the original non-contaminated soil.

layer of NCC. The Pb and Sb transport distance through water migration in unsaturated soil may depend on the distance between CC and PC. Therefore, another water migration experiment was conducted using the 10 mm NC-Pot to compare with the results using the 5 mm NC-Pot.

Figure 3 shows the level of total Pb and Sb in each NCC layer in the 10 mm NC-Pot. The level of total Pb in the 1 mm layer of NCC, which was nearest to CC, was the highest; it was 14 times higher than that in the original non-contaminated soil. In addition, the level of total Pb in the 3 mm layer was approximately two times higher than that in the original non-contaminated soil. However, the level of total Pb in the layers 6 mm or more, which was not provided in the 5 mm NC-Pot, was the same as that in the original non-contaminated soil. These results suggested that Pb was transported 3 mm from CC in the NCC through water migration during the immobilization period in the 10 mm NC-Pot. The level of total Sb was the highest in the 1 mm layer, reduced with the distance, and was the same as the original non-contaminated soil in layers 7 mm or more. Thus, in contrast to Pb, these results demonstrated that Sb was transported 6 mm through water migration during the immobilization period in the 10 mm NC-Pot.

The Pb and Sb transport distances did not differ significantly in the water migration experiments using a 5 mm NC-Pot and a 10 mm NC-Pot. Accordingly, these results demonstrated that Pb and Sb can be transported 3-5 mm and 6 mm, respectively, through water migration, which corresponds to precipitation for one month. Tlustoš *et al.* [17] and Fitz *et al.* [18] conducted a plant growth experiment using a rhizobox and reported that the plant absorbed Pb and arsenic through water uptake from rhizosphere soil, which is a few

millimeters away from the root. These results are consistent with those obtained in this study. Thus, the Pb and Sb transport distances in this study would be valid.

Steely *et al.* [12] and Cao *et al.* [19] have observed that the levels of Pb and Sb in the shooting range soil decreased relative to soil depth. However, in this study, Sb was transported farther than Pb in NCC (Fig. 3). This observation suggested that, compared to Pb, Sb was transported easily through water migration. Sb would exist as an anion ($\text{Sb}(\text{OH})_6^-$) of Sb (V) in the shooting range soil under oxic conditions, and Sb would be transported as $\text{Sb}(\text{OH})_6^-$ through water migration. In contrast, Pb would exist primarily as a Pb compound, such as PbCO_3 and PbO . The Pb compounds were dissolved; dissolved Pb would be transported and sorbed onto the colloid containing iron oxide and organic matter [10], [19]. Therefore, Sb was probably transported easily because transport of a colloid is more difficult than that of an anion. These results suggest that Sb is effectively immobilized in soil because easy transport in soil results in frequent contact and reaction with immobilization amendment.

3.2 Transporting Phases of Pb and Sb

The ratios of water-soluble Pb and Sb to total Pb and Sb in NCC in the 5 mm NC-Pot are shown in Fig. 4. The ratios of water-soluble Pb to total Pb in all layers were very low. The ratio in the 1 mm layer, in which the highest level of total Pb was found, was less than 2%. In contrast to Pb, the ratios of water-soluble Sb in all layers were high, and 30% of the total Sb in the 1 mm layer was water-soluble.

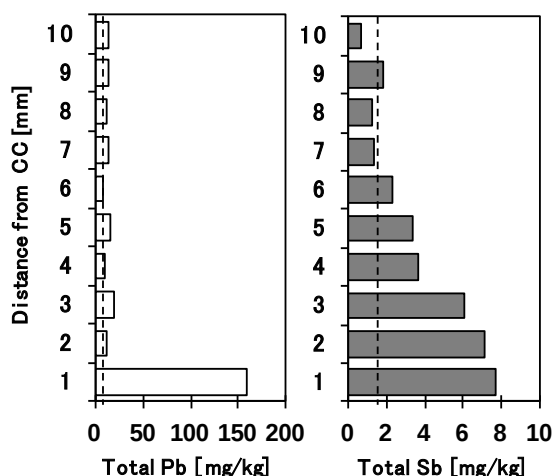


Fig. 3 Total heavy metal levels in NCC of 10 mm NC-Pot. Broken lines indicate the background levels in the original non-contaminated soil.

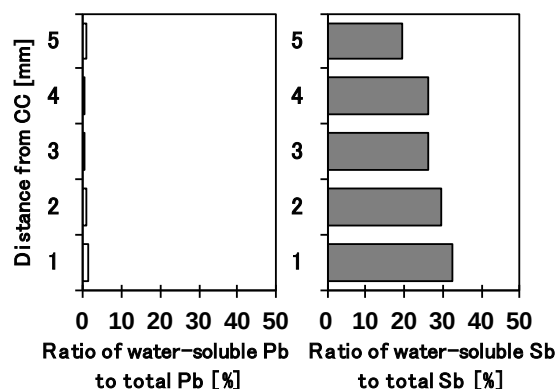


Fig. 4 Ratios of water soluble to total in NCC of 5 mm NC-Pot

Figure 5 shows the ratios of water-soluble Pb and Sb to total Pb and Sb in the 10 mm NC-Pot. As was observed with the 5 mm NC-Pot, the ratios of water-soluble Sb were higher than those of water-soluble Pb in all layers. The ratio of water-soluble Pb in the 1 mm layer was 2%, while that of water-soluble Sb was 42%. Furthermore, 30% to 40% of the total Sb in the 1 to 6 mm layers, in which the level of total Sb exceeded that of the original non-contaminated soil, was water-soluble. These ratios are consistent with the results (Pb: 2%–10%, Sb: 4%–72%) obtained by Sanderson *et al.* [20]. These results demonstrated that Sb in the shooting range soil would be more water-soluble, and thus, Sb would have higher mobility through water migration than Pb.

The Pb and Sb that were passed through the 0.45 μm filters were defined as water-soluble in this study. However, Pb underwent colloid transport in the soil, and Pb transport would strongly relate with 0.45–0.8 μm colloid particles [10]. Thus, approximately Pb transported into the layers of NCC likely existed in the sorbing phase on the colloid; Pb would be transported as non-water-soluble phases. Sb was also sorbed on iron oxides. However, the level of Sb sorption was reduced under neutral to alkali soil pH [20]. Therefore, in the soil used, Sb was less likely to be sorbed, which probably induced Sb to be more water-soluble.

Sequential extraction was performed to evaluate Pb transporting phases because Pb was probably transported in non-water-soluble phases. The levels of each Pb fraction in the 1 and 5 mm layers of NCC in the 5 mm NC-Pot are shown in Fig. 6. The level of Pb fraction in the 1 mm layer was highest in the fraction bound to Fe/Mn oxide at 27 mg/kg. The level of Pb fraction in the 5 mm layer, where the level of total Pb was the lowest, was also highest in the fraction bound to Fe/Mn oxide at 6 mg/kg. The levels of the carbonate and Pb fractions bound to Fe/Mn oxide in the 1 mm layer were significantly higher than those in the 5 mm layer. The level of the residual fraction, which was the most stable, was the same for the 1 and 5

mm layers. Therefore, Pb phases transported through the layers appeared to be Pb compounds, such as carbonate. In addition, the Pb phase bound to Fe/Mn oxide; the Pb phase transported with the colloid would be phase sorbed on the iron oxide. In acid soil, Pb carbonate and Pb sorbed on iron oxide are likely to be dissolved and desorbed, respectively [20]. However, in the soil used, the dissolution and desorption was suppressed because of neutral soil pH.

These results indicate that the distance and phases during transport in the soil differed between Pb and Sb. Therefore, to enhance the effectiveness of immobilization, the type of heavy metal should be taken into account. In this study, only one type soil was used. Further investigation should consider more soil types because soil particle size and chemical composition strongly affect Pb and Sb sorption on the soil surface.

3.3 Required Amount of Immobilization Amendment by Microscopic Range

It was anticipated that, in the case of immobilization in contaminated soil, Pb and Sb would be transported through water migration a

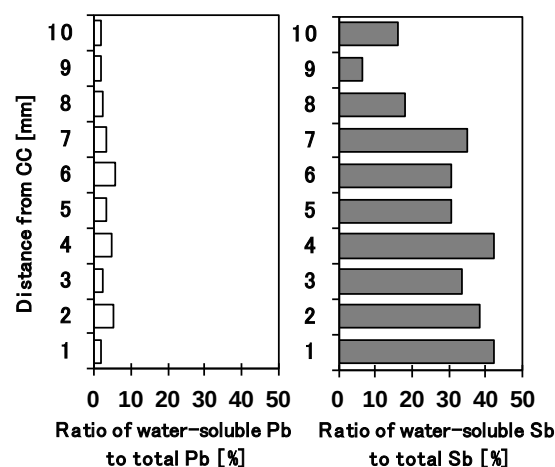


Fig. 5 Ratios of water soluble to total in NCC of 10 mm NC-Pot

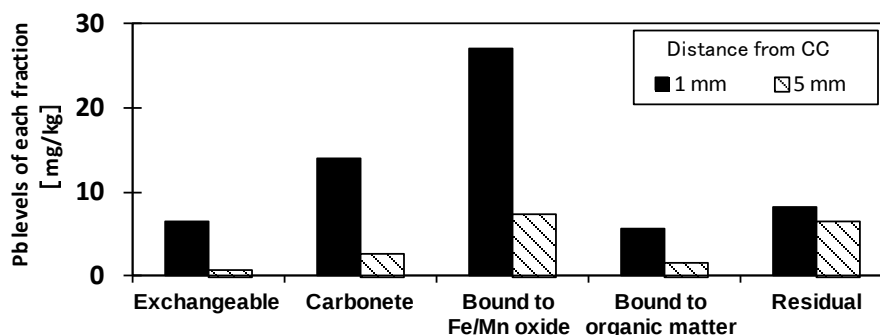


Fig. 6 Pb fractions of sequential extraction in NCC of 5 mm NC-Pot

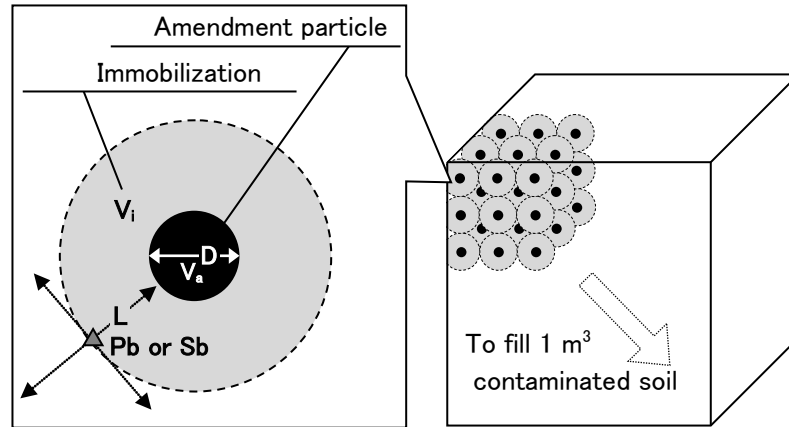


Fig. 7 Overview of microscopic range for immobilization

maximum of 5 to 6 mm, which corresponds to precipitation for one month. However, the transported Pb and Sb were particularly accumulated in the 1 mm layer. Therefore, assuming that the transporting distance to immobilize Pb and Sb is 1 mm, the required amount of amendment was calculated from the following equations on the basis of the microscopic range of immobilization. The overview of the microscopic range is shown in Fig. 7. The microscopic range of Pb and Sb immobilization was calculated according to the following equation:

$$V_i = \frac{4\pi \cdot (L + D/2)^3}{3} \quad (1)$$

where V_i is the microscopic range of Pb and Sb immobilization (mm^3), L is the transporting distance during the immobilization period (1 mm), and D is the diameter of the amendment particle. In case that $D = 1$ mm, the value of V_i was obtained as 14.1 mm^3 using Eq. (1). The required number of immobilization amendment particles to fill 1 m^3 of contaminated soil for the established range of Pb and Sb immobilization (N) can be expressed as

$$N = \frac{1 \times 10^9}{V_i} \quad (2)$$

The value of N was calculated to be 7.09×10^7 .

The volume of one immobilization amendment particle (V_a) can be calculated as

$$V_a = \frac{4\pi \cdot (D/2)^3}{3} \quad (3)$$

Moreover, using V_a and N , the required volume of immobilization amendment (A_v) in 1 m^3 of contaminated soil can be expressed as

$$A_v = \frac{V_a \cdot N}{1 \times 10^9} \times 100 \quad (4)$$

The value of A_v was calculated to be 3.72% using Eq. (4). By converting the volume ratio to weight ratio using the density of the amendment and the contaminated soil (Table 1), the weight ratio of amendment was calculated to be 4.62%.

It would be effective for chemical immobilization to add the phosphorus amendment with 0.6 molar ratio of Pb in the soil and P in the amendment because Pb is immobilized in soil with the precipitation of Pb phosphate minerals such as pyromorphite ($\text{Pb}_5(\text{PO}_4)_3\text{OH}$); its P/Pb molar ratio is 0.6 [6]. In the case that the amendment has been added to the soil at 5% (w/w), the molar ratio of Pb in the soil and P in the amendment would be 0.771. Thus, addition of amendment with 5% (w/w) is probably sufficient to immobilize Pb. Moreover, Álvarez-Ayuso *et al.* [7] added 5% (w/w) iron oxide to the Sb-contaminated soil in which the level of total Sb was 25 times higher than that in the soil used in this study. The results of that study indicated that Sb was completely immobilized. Therefore, in the soil used in the current study, the addition of amendment with 5% (w/w) would be sufficient for Pb and Sb immobilization. Further investigation is necessary because this study simplified estimation conditions to calculate the required amount of immobilization amendment.

4. CONCLUSION

The present study evaluated the distance of Pb and Sb transport through water migration and their transporting phases during the immobilization period to identify the microscopic range of immobilization in the soil. In addition, the required amount of immobilization amendment was investigated on the basis of the microscopic range of immobilization. The results obtained in this study are as follows:

- 1) Pb and Sb were transported 5 mm and 6 mm at most, respectively, and were particularly accumulated 1 mm from contamination.
- 2) Thirty to forty percent of transported Sb was water-soluble, which suggests that Sb was easily transported in soil to contact and react with immobilization amendment. Pb phases transported through water migration sorbed on Fe/Mn oxide and carbonate. Thus, Pb probably has lower mobility than Sb.
- 3) The distance and phases during transport in the soil differed between Pb and Sb. To enhance the effectiveness of immobilization, the type of heavy metal should be considered.
- 4) The required amount of immobilization amendment was estimated from the range of immobilization (1 mm) to be 4.62% (w/w).

5. ACKNOWLEDGEMENTS

The ICP-OES and CHO analyzer used for chemical analysis in this study was made available by the Division of Instrumental Analysis at Gifu University. The authors are grateful to Prof. F. Li and Prof. T. Yamada (Gifu University) for allowing the use of the ICP-MS. This study was partly supported by Japan Society for the Promotion of Science (JSPS) KAKENHI Grant Number 23710089 and the Maeda Engineering Foundation.

6. REFERENCES

- [1] Dickinson NM, "Strategies for sustainable woodland on contaminated soils", *Chemosphere*, Vol. 41, 2000, pp. 259–263.
- [2] Allen A, "Containment landfills: the myth of sustainability", *Eng. Geol.*, Vol. 60, 2001, pp. 3–19.
- [3] Mulligan CN, Yong RN, Gibbs BF, "An evaluation of technologies for the heavy metal remediation of dredged sediments", *J. Hazard. Mater.*, Vol. 85, 2001, pp. 145–163.
- [4] Sorvari J, Antikainen R, Pyy O, "Environmental contamination at Finnish shooting ranges—the scope of the problem and management options", *Sci. Total Environ.*, Vol. 366, 2006, pp. 21–31.
- [5] Siebielec G, Chaney RL, "Testing amendments for remediation of military range contaminated soil", *J. Environ. Manage.*, Vol. 108, 2012, pp. 8–13.
- [6] Chrysochoou M, Dermatas D, Grubb DG, "Phosphate application to firing range soils for Pb immobilization: The unclear role of phosphate", *J. Hazard. Mater.*, Vol. 144, 2007, pp. 1–14.
- [7] Álvarez-Ayuso E, Otones V, Murciego A, García-Sánchez A, "Evaluation of different amendments to stabilize antimony in mining polluted soils", *Chemosphere*, Vol. 90, 2013, pp. 2233–2239.
- [8] Chiang YW, Santos RM, Ghyselbrecht K, Cappuyns V, Martens JA, Swennen R, Gerven TV, Meesschaert B, "Strategic selection of an optimal sorbent mixture for in-situ remediation of heavy metal contaminated sediments: Framework and case study", *J. Environ. Manage.*, Vol. 105, 2012, pp. 1–11.
- [9] Zhang W, Tsang DCW, "Conceptual framework and mathematical model for the transport of metal-chelant complexes during in situ soil remediation", *Chemosphere*, Vol. 91, 2013, pp. 1281–1288.
- [10] Yin X, Gao B, Ma LQ, Saha UK, Sun H, Wang G, "Colloid-facilitated Pb transport in two shooting-range soils in Florida", *J. Hazard. Mater.*, Vol. 177, 2010, pp. 620–625.
- [11] Garrido F, Helmhart M, "Lead and soil properties distributions in a roadside soil: Effect of preferential flow paths", *Geoderma*, Vol. 170, 2012, pp. 305–313.
- [12] Steely S, Amarasingiwardena D, Xing B, "An investigation of inorganic antimony species and antimony associated with soil humic acid molar mass fractions in contaminated soils", *Environ. Pollut.*, Vol. 148, 2007, pp. 590–598.
- [13] Wang S, Choi JH, "Simulating fate and transport of chromium in saturated sediments", *Appl. Math. Model.*, Vol. 37, 2013, pp. 102–111.
- [14] Furuta S, Katsuki H, Komarneni S, "Removal of lead ion using porous hydroxyapatite monoliths synthesized from gypsum waste", *J. Ceram. Soc. Jap.*, Vol. 108, 2000, pp. 315–317.
- [15] Mitsunobu S, Sakai Y, Takahashi Y, "Characterization of Fe(III) (hydr)oxides in arsenic contaminated soil under various redox conditions by XAFS and Mössbauer spectroscopies", *Appl. Geochem.*, Vol. 3, 2008, pp. 3236–3243.
- [16] Tessier A, Campbell PGC, Bisson M, "Sequential extraction procedure for the speciation of particulate trace metals", *Anal. Chem.*, Vol. 51, 1979, pp. 844–851.

- [17] Tlustoš P, Száková J, Kořínek K, Pavlíková D, Hanč A, Balík J, "The effect of liming on cadmium, lead, and zinc uptake reduction by spring wheat grown in contaminated soil", *Plant. Soil Environ.*, Vol. 52, 2006, pp. 16–24.
- [18] Fitz WJ, Wenzel WW, Zhang H, Nurmi J, Štípek K, Fischerová Z, Schweiger P, Köllensperger G, Ma LQ, Stingeder G, "Rhizosphere characteristics of the arsenic hyperaccumulator *Pteris vittata* L. and monitoring of phytoremoval efficiency", *Environ. Sci. Technol.*, Vol. 37, 2003, pp. 5008–5014.
- [19] Cao X, Ma LQ, Chen M, Hardison Jr DW, Harris WG, "Lead transformation and distribution in the soils of shooting ranges in Florida, USA", *Sci. Total Environ.*, Vol. 307, 2003, pp. 179–189.
- [20] Sanderson P, Naidu R, Bolan N, Bowmanc M, Mclure S, "Effect of soil type on distribution and bioaccessibility of metal contaminants in shooting range soils", *Sci. Total Environ.*, Vol. 438, 2012, pp. 452–462.

Int. J. of GEOMATE, June, 2014, Vol. 6, No. 2 (Sl. No. 12), pp. 870-877.

MS No. 3196 received on June 12, 2013 and reviewed under GEOMATE publication policies.

Copyright © 2014, International Journal of GEOMATE. All rights reserved, including the making of copies unless permission is obtained from the copyright proprietors. Pertinent discussion including authors' closure, if any, will be published in the June. 2015 if the discussion is received by Dec. 2014.

Corresponding Author: Shouhei Ogawa
