

# Emission Control of Mercury from Stabilized and Solidified Products under Monofill Conditions

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## Abstract

A column leaching test was conducted for 258 days to demonstrate Hg emissions in an engineered landfill where stabilized and solidified mercury by using sulfur and a binder such as cement was disposed of under monofill conditions with a granite soil layer. Thermodynamical analysis considering leachate quality showed that the mercury in the leachate could be chlorides, indicating that the mercury in metacinnabar in stabilized/solidified products (below, s/s products) dissolved in percolation water. However, the gaseous and liquid emissions of soluble mercury were extremely low because of adsorption in the granite soil. Nearly all mercury released from the stabilized/solidified product into the soil stayed within 1.5 cm of the product. Mercury contents were greater in deeper soil than in upper soil layers. It seems that migration of Hg out of the columns was delayed due to absorption of mercury in the soil and moisture. Sulfur polymer effectively immobilized mercury than low-alkali cement. However, both s/s products could achieve effective containment of mercury-containing waste under monofill conditions with a mercury-adsorbing soil layer. It is important not only to achieve stabilization/solidification of mercury but also to install a mercury-adsorption layer surrounding the treated waste to minimize mercury emissions from landfills.

**Key words :** column leaching test, landfill, mercury emissions, solidified/stabilized mercury

## 1. Introduction

The Minamata Convention on Mercury was agreed on by nations in 2013. After the convention enters into force, the utilization of metallic mercury will be restricted and appropriate and safe storage and disposal will be required to minimize mercury emissions into the environment. Fossil fuel (primarily coal) combustion and cement production processes are estimated to be the main sources of global anthropogenic mercury emissions to the atmosphere (Kim & Zoh, 2012; Zhao *et al.*, 2019). In Japan, mercury has mainly been emitted from waste incineration facilities; however, the amount of emissions was 5.44 tons/year in 2014 (Takiguchi & Tamura, 2018), much less than those of other countries around the world. According to a review by Tao *et al.* (2017), emissions from working faces in 10 landfills located in China, Korea and the USA are on the order of one to a thousand g/year, which may depend on differences in Hg content of received waste (0.0328–46.2 mg/kg). Immediately covering them with soil has been described as a strategy for emission control. Hg discharge with leachate from sites for landfilling municipal solid waste was also

investigated by Chai *et al.* (2011) and Ilgen *et al.* (2008). Yanase *et al.* (2009) described long-term Hg behavior in a lysimeter filled with Hg-containing waste, i.e., used-batteries and municipal solid waste. On the other hand, waste consisting of elemental mercury is never landfilled because elemental mercury can be recovered for resource use from the waste and its landfilling is discouraged. Hence, the long-term behaviour of elemental mercury and its compounds when wastes with high concentrations of immobilized mercury are disposed of in landfills remains unknown.

International organizations and countries are developing regulations or guidelines for environmentally sound management of mercury waste. With regard to final disposal, elemental mercury waste in the EU can be permanently stored in geological repositories such as abandoned salt mines or hard rock (European Council, 2008). The U.S. Department of Energy has already constructed an above-ground facility for storage of elemental mercury containers (Lee *et al.*, 2012). In Japan, there has been discussion of the safe disposal of immobilized metal mercury in an engineered landfill if it satisfies certain criteria, e.g., regulation of leachability,

and is separated from other wastes.

It is essential to control the mercury emissions from landfill bodies to the surroundings almost eternally, and appropriate treatment techniques are needed to immobilize mercury in a landfill. There are several mercury waste treatments which include mercury sulfide formation and a solidification method (UNEP, 2012; Donatello *et al.*, 2012; López *et al.*, 2015). As for stabilization/solidification processes, elemental mercury, which has high solubility and volatility, is reacted with sulfur powder to produce metacinnabar to reduce the mobility of the mercury by changing its chemical properties (UNEP, 2012). In addition, the stabilized mercury is mixed with a binder such as sulfur polymer to encapsulate and physically isolate it from the surrounding environment (UNEP, 2012; Rodríguez *et al.*, 2012). A treatment technique for elemental mercury has been developed, with which leached and vaporized Hg concentrations from sulfur-polymer-treated waste containing mercury have been revealed to be at allowable levels and extremely low (Fuhrmann *et al.*, 2002). However, these results were obtained under experimental conditions for assessing treatability. It is important to understand the behaviour of mercury in real landfilling situations in which both liquid and gas come in contact with the treated waste, i.e. under unsaturated percolation and landfill gas flow, from the point of view of identifying long-term mercury emissions, especially in post-aftercare.

In this study, a column leaching test under simulated landfill conditions was conducted to clarify the release of mercury into the atmosphere and leachate, and to suggest appropriate landfilling of treated metallic mercury for safe management of its final storage and disposal.

## 2. Materials and Methods

### 2.1. Stabilized/Solidified Products and Soil for the Column Leaching Test

As a pre-treatment of elemental mercury, metacinnabar was formed by reacting mercury (purity 99.9%) and sulfur following the method shown by Fukuda *et al.* (2014). The product material was ground

and mixed with a binder for solidification. Four types of stabilized/ solidified product (hereinafter called s/s products), were produced in the shape of cubes of about 7.8 cm and named as SP, CA, CB and CB+ according to type of binder and amounts of components as shown in Table 1. Sulfur polymer was produced by mixing the components under decreased pressure in N<sub>2</sub> gas at 130°C for 1 hour. Low-alkali cement cubes were produced with a curing period of 28 days. The mercury content in the s/s products was calculated by the amount of elemental mercury component during the sulfurization (shown in Table 1). The sulfur polymer was a binder which contained ~95% elemental sulfur (Rodríguez *et al.*, 2012). The low-alkali cement was composed of Portland cement, silica fume and fly ash (Codina *et al.*, 2008; Mihara *et al.*, 2008). The pH of the cement paste was lower than the equilibrium pH of portlandite. The two types of low-alkali cements (A and B) were commercial products and produced from different raw materials.

The mercury content of the SP (shown in Table 1) was slightly higher than that of the others because of the high metacinnabar content. The compounded amount of the components differed for each s/s product to facilitate comparison of the ability to bind mercury at the same ratio by weight of the binder and metacinnabar.

Decomposed granite soil was selected for the mercury-absorbing material due to its high permeability ( $2.3 \times 10^{-5}$  m/s). The weight percentages of soil with particle sizes of less than 0.005, 0.005–0.075, 0.075–2 and 2–75 mm were 3.8, 11.8, 51.2, and 33.2%, respectively. The maximum size of the soil particles was 19 mm. The optimum moisture content was 13.4%.

### 2.2. Column Leaching Test

Considering safer landfilling of s/s products at an existing sanitary engineered landfill in Japan, we assumed the following landfilling conditions: The s/s product was filled above an old-waste layer that included organic waste, and was not mixed with other wastes; rainwater infiltrated under unsaturated conditions; the soil-adsorption layer was placed around the s/s product; landfill gas migrated to the mercury-storage layer from the existing waste layer so that the conditions in the latter

**Table 1** Types of binders and components of the s/s products.

s/s product	Type of binder	Component ratio by weight (%)				Wp* (g)	MC** % w/w
		Binder	Water	Me <sup>a</sup>	Agant <sup>b</sup>		
SP	Sulfur polymer	50	0	50	0	1509	43
CA	Low-alkali cement “A”	40	20	40	0	1202	34
CB	Low-alkali cement “B”	40	20	40	0	1145	34
CB+	Low-alkali cement “B”	40.8	18.3	40.8	0.2	1147	35

a: Metacinnabar (ground samples), b: Water-reducing agent for cement

\* Weight of s/s product, \*\* Mercury content in the s/s product

layer became anaerobic. Figure 1 shows a schematic of one experimental column. The columns consisted of three main parts: a sprinkling device, column space for filling, and drainage. Each part was tightly sealed with a flange connection. Drops of water fell from the needles of the sprinkling device. A gas bag was connected to the vent hole of the sprinkling device to allow the gas pressure in the column to adjust to that of atmosphere. A Hg-adsorption tube containing a gold trap and air pump was connected to the vent hole to constantly circulate the gases and trap gaseous mercury in the top space of the column. The columns were set in a room with ambient temperature kept around 25 °C.

Decomposed granite soil was filled at a density of 1,692 kg-dry/m<sup>3</sup> above glass beads on the punched stainless-steel plate. The s/s product was placed at the center of the soil layer with a maximum thickness of 15 cm. The top of the filling layer was covered with glass beads. The moisture content of the soil was adjusted to 13.4% (OMC) before filling.

Approximately 320 mL of degassed water was added once a day from the top of the column, so that the amount of infiltration per cross-sectional area of the column was about 4.5 mm/d, approximately corresponding to the average annual precipitation in Japan. Precipitation in a landfill cannot all percolate into the waste layer because of evaporation and run-off from the cover (Nakajima *et al.*, 1991; Baccini *et al.*, 1987). This experiment, however, assumed more Hg-leachable conditions. The cumulative amount of infiltration water per cross-sectional area was 1,027–1,074 mm, depending on the column, at the end of the column leaching test (258 days). Aiming to create anaerobic circumstances in the columns

and adjust the pH of the supplied water, L-cysteine and sodium hydroxide were added to the degassed water. Hence, the average quality of the supplied water was as follows: pH 7.5; Cl<sup>-</sup> concentration of 10.4 mg/L; and oxidation-reduction potential (Eh) of 28.4 mV. The leachate was kept constantly at a particular water level in the drainage system and smoothly collected, then stored in polypropylene bottles without exposure to air. The leachate weights were measured and their qualities were analyzed; the pH and Eh were measured by dedicated electrodes; concentrations of Ca<sup>2+</sup>, Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup> were analyzed by ion chromatography and inductively coupled plasma atomic emission spectroscopy; and total mercury amounts in the leachate were measured by a direct thermal decomposition mercury analyzer (MA-3000, Nippon Instruments Corp., Tokyo, Japan).

As for simulating landfill gas exposure to the s/s products, 20 L of standard gas mixture was sent into the columns through the down vent hole. This ventilation was conducted 21 times during days 22–50 and 155–258 of the column leaching test. The gas mixture was composed of 40% carbon dioxide, approx. 20 ppm hydrogen sulfide, and nitrogen (balance gas), chosen with reference to landfill gas concentration in anaerobic conditions (Nagamori *et al.*, 2016; Kim *et al.*, 2005; Gebert *et al.*, 2011), except for CH<sub>4</sub> gas which was considered not to affect the behavior of Hg. Gas inside the soil layer was sampled via the sampling pipe and the composition of CO<sub>2</sub>, N<sub>2</sub> and O<sub>2</sub> by GC-TCD and H<sub>2</sub>S was measured by GC-FID. The gas in the upper part of the columns was constantly circulated by air pump so that gaseous mercury could be collected by the Hg adsorption tube (gold trap) installed in the upper vent hole of each column. The release rate was calculated by dividing the released Hg via leachate or gas by the initial amount of metallic Hg in each s/s product and the interval of sampling leachate or cumulative gaseous Hg.

After 258 days of the column leaching test, different zones of the granite soil were sampled from the columns at different depths. The zone surrounding the s/s product was less than 2 cm in thickness with an area of about 100 cm<sup>2</sup>. The upper zone was at a depth of 0 to 2.5 cm, the lower zone was below the surrounding zone, at depth of 11.5 to 15.0 cm, and the horizontal zone was other bodies of soil at depths of 2.5 to 11.5 cm. The portions of the soil sampled from the columns, approx. 10 g each, were extracted using a solution containing nitric acid, sulfuric acid and potassium permanganate (30 g/L). The amount of Hg in the extract was analyzed using the mercury analyzer. Distributions of Hg in the granite soil were calculated by dividing amount of Hg in each part of the soil by the total Hg in all of the granite soil.

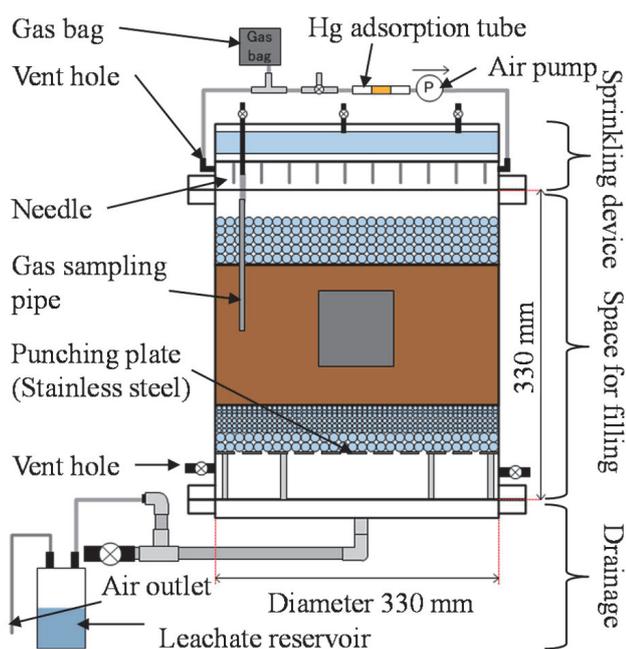


Fig. 1 Outline of the experimental column.

### 3. Results and Discussion

#### 3.1. Gas Composition in the Columns and Leachate Quality

Table 2 shows the range of gas compositions in the soil layer during gas ventilation. The CO<sub>2</sub> concentrations were higher than the atmospheric composition, and O<sub>2</sub> gas also existed in the soil layer. After the gas replacement, the CO<sub>2</sub> concentration in columns gradually declined. In contrast, the O<sub>2</sub> concentration increased. No H<sub>2</sub>S gas was detected even just after the replacement. The solubility of CO<sub>2</sub> and H<sub>2</sub>S gas in water are higher than

that of O<sub>2</sub> gas. It was thought that the CO<sub>2</sub> and H<sub>2</sub>S gases were dissolving in the pore water. During times with no ventilation, the gas composition in the soil layer was the same as the atmospheric condition.

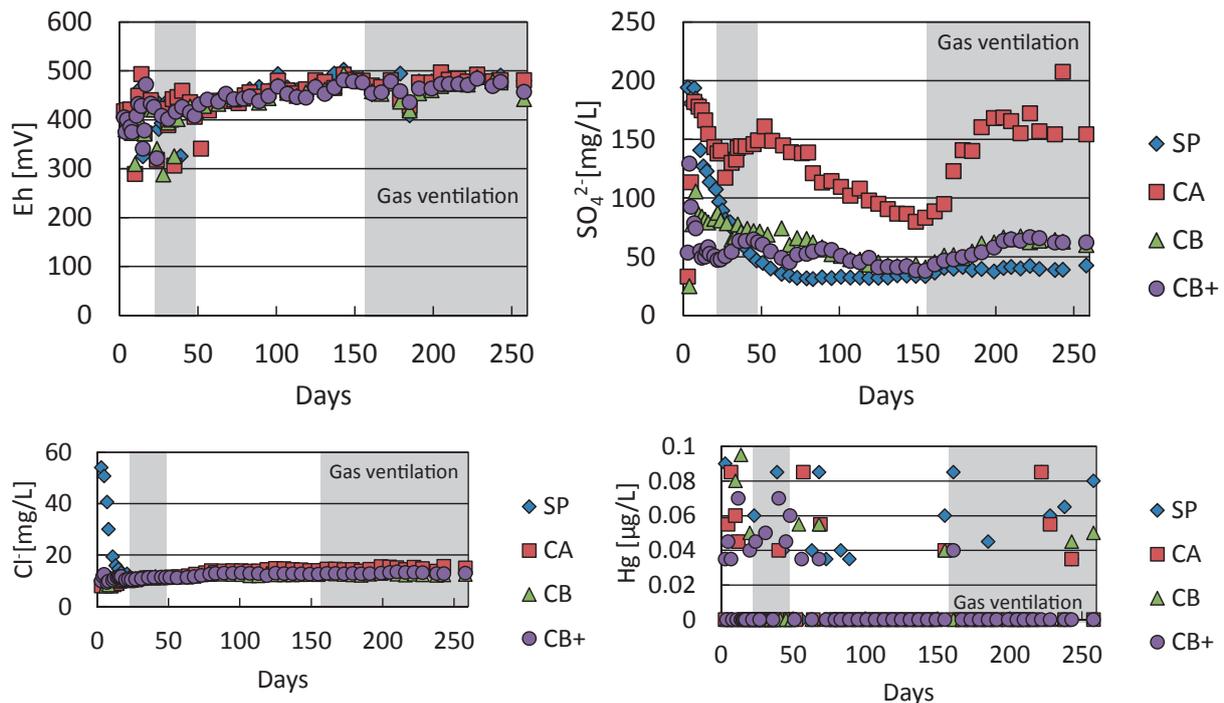
The ranges of leachate quality in each column are shown in Table 3. The pH of the leachates was almost stable and neutral. Figure 2 shows the Eh and concentrations of SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, and Hg in the leachates. The Eh did not greatly change and ranged mostly from 400 to 500 mV in each column because of the high concentration of O<sub>2</sub> gas and insufficiency of reducing agents (L-Cysteine hydrochloride monohydrate and sodium

**Table 2** Gas composition in the soil layer during gas ventilation.

	CO <sub>2</sub> composition [%]				O <sub>2</sub> composition [%]			
	SP	CA	CB	CB+	SP	CA	CB	CB+
Minimum	5	2	4	2	4	5	3	6
Maximum	26	24	27	21	16	19	17	15
Average	14	9	13	11	10	12	10	10

**Table 3** Leachate quality in the column leaching test (“Ave” is average value during the test).

	Cl <sup>-</sup> [mg/L]			SO <sub>4</sub> <sup>2-</sup> [mg/L]			Ca <sup>2+</sup> [mg/L]			pH		
	Min	Max	Ave	Min	Max	Ave	Min	Max	Ave	Min	Max	Ave
SP	11	54	15	31	194	62	11	71	20	5.6	6.5	6.1
CA	7.8	15	13	33	182	133	30	66	53	5.6	6.7	6.2
CB	7.8	13	12	25	106	63	14	28	26	5.6	6.7	6.2
CB+	9.3	13	12	41	129	56	13	35	23	5.5	7.3	6.2



**Fig. 2** Eh and SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, Hg concentrations in the leachate during the leaching test.

hydroxide) in the supplied water.  $\text{SO}_4^{2-}$  concentrations in the leachate from each column, except the SP column, increased during the periods of gas ventilation and gradually decreased in the other periods. High concentrations of  $\text{SO}_4^{2-}$  in the leachate were detected in the CA column. The changes in  $\text{Ca}^{2+}$  concentrations were similar to those of  $\text{SO}_4^{2-}$ . The concentration of  $\text{Cl}^-$  in the leachate from the SP column initially rose to a maximum and then declined to the range of 12–15 mg/L. In other columns, the  $\text{Cl}^-$  concentration increased from a minimum to a maximum as time went by.

The change in  $\text{Ca}^{2+}$  and  $\text{SO}_4^{2-}$  concentrations in the leachates of the CA, CB, and CB+ columns indicated dissolution of cement hydrates such as ettringite, including calcium and sulfate. Hydration of the low-alkali cement “A” was said the main cause of ettringite formation. The low-alkali cement “B” contained calcium silicate hydrates and a little ettringite (Hoshino *et al.*, 2014). Hence, the different  $\text{SO}_4^{2-}$  concentrations in the leachates resulted from leaching of ettringite that seemed to decompose by carbonation during gas ventilation (Nishikawa *et al.*, 1992).

Hg concentrations in the leachate were higher than the limit for detection (0.03  $\mu\text{g/L}$ ) of Hg several times, never exceeding that for quantification (0.1  $\mu\text{g/L}$ ) in the analytical method used in this experiment. It is widely known that the leaching behavior of metacinnabar is affected by pH (Ravichandran *et al.*, 1999). In addition, according to the pH–Eh diagram (shown by Gavis & Ferguson, 1972; Hem, 1970), the chemical form of Hg and its solubility in water depend on the presence of Cl and S and on the pH and Eh values. A thermodynamic analysis using software (FactSage) indicated that Hg in

leachate could form chlorides ( $\text{HgCl}_2$ ,  $[\text{HgCl}_4]^{2-}$ ) under these leaching test conditions ( $[\text{Cl}^-] = 10^{-3.53}$  mol/L,  $[\text{SO}_4^{2-}] = 10^{-2.82}$  mol/L,  $[\text{Ca}^{2+}] = 10^{-2.7}$  mol/L). Hence, Hg leached from the s/s products and changed to chloride, which has higher solubility than metacinnabar.

The cumulative amounts of Hg in the leachates and gases are shown in Fig. 3. The amount of Hg released in the leachate increased prominently in the initial quarter term in which cumulative percolation was about 300 mm, except for the SP column. Hg was constantly released via gases throughout the days of the experiment.

Table 4 shows the cumulative amount of Hg released in leachate or gas and adsorbed in the soil at the end of the column leaching test per the initial amount of metallic mercury in the s/s products. Emissions from the columns were extremely low, at a few ng/g, and almost the same via gas and leachate in any of the columns. On the other hand, the amount of Hg adsorbed by the soil was higher than that of the emissions and differed between the HgS-binding sulfur polymer and low-alkali cement. Metacinnabar has low solubility (Drott *et al.*, 2013), but Hg in leachate may have formed chloride, which has higher solubility, under the conditions in this column test, as mentioned above. It has been well observed that not only dissolution of soluble matter on the surface of a solid, especially a porous one, but also diffusion from inside to the surface of it contribute to leaching behavior. Gavis and Ferguson (1972) suggested that Hg forms hydroxide, which also has a higher solubility than metacinnabar, under the high pH and Eh of a system with S, Cl and Hg coexistence. The pH of the low-alkali cement is maintained at over 10, as Codina *et al.* (2008) showed, so more Hg may be diffused and dissolved from

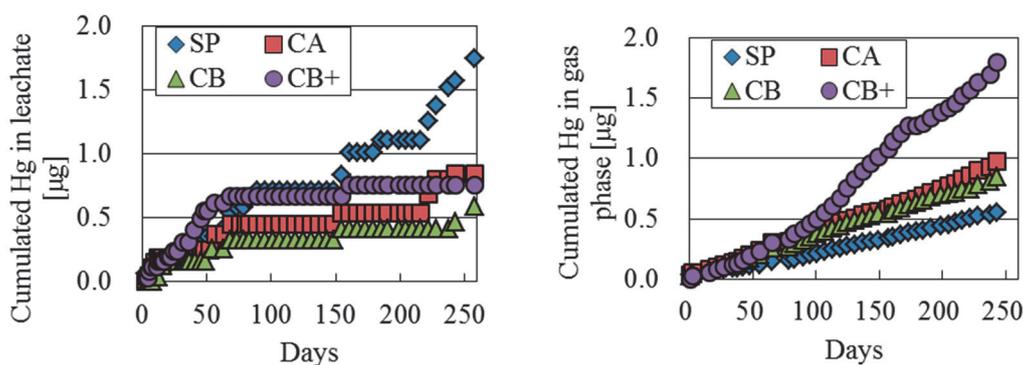


Fig. 3 Cumulative amounts of Hg released from the column b via leachate (left) and evaporation (right).

Table 4 Mercury release (ng/g) from s/s products under landfill conditions.

		SP	CA	CB	CB+
Emission from column	Gas	0.85	2.4	2.2	4.5
	Soluble	2.7	2.1	1.5	1.9
Adsorption in granite soils		5,660	122,000	39,200	55,900
Release from s/s products		5,664	122,004	39,204	55,906

the products of low-alkali cement than those of the sulfur polymer. Although the amounts of dissolved Hg from metacinnabar differed between the *s/s* products, the Hg emissions from the columns under the simulated landfill conditions were at the same level. This could be considered the result of adsorption in the soil.

Regarding the distribution of Hg in the granite soil at the end of the column leaching test (Table 5), 64% of mercury released from *s/s* product CB to the granite soil existed in the surrounding part within approx. 1.5 cm of the product, suggesting delayed migration of Hg by adsorption into the soil. The other 25.7% of the Hg was mobilized to the horizontal bulk parts. This indicates migration by diffusion. The upper and lower zones of *s/s* product CB over 1.5 cm from the products contained 0.4 % and 9.9 % of Hg released to the soil from the *s/s* products, respectively. Similar results were also obtained from the column leaching test for *s/s* products SP, CA, and CB+. In terms of Hg content in each part of the soil (Table 5), the Hg content values were maximized in the surrounding soil within 1.5 cm of the *s/s* products. In addition, the Hg content tended to increase in the deeper zone of the columns. These results indicate that the soil at least 1.5 cm away from the *s/s* products was non-saturated with Hg, although the strictly local soil might not be. Therefore, Hg leached from the *s/s* products seems to be slowly emitted from the columns due to repeated adsorption/desorption of soluble and gaseous Hg to/from the soils, and dissolution/evaporation of gaseous Hg to/from moist soils under conditions of unsaturated water flow. On the other hand, this Hg distribution among the three phases might change spatially and temporally, and the distributional variation more likely affects the temporal variation of Hg concentration than variations in pH and ion species in the leachates, as shown in Fig. 2. Moreover this variation results in increased release via leachates in the SP column, as shown in Fig. 3, but the Hg content of the soil was lower yet than in the other columns, as shown in Table 5: Thus soil has the potential for Hg adsorption. Therefore, it was thought that the release of Hg via the leachates could not constantly increase unless the metacinnabar in the SP leached dramatically.

Although discussion is needed on how much the amount of anthropogenic emissions affects the environment, selection of a stabilization/solidification method for waste mercury should also consider treatment costs and the containable amount per solidified volume. A higher Hg content per volume could lead to less space required and easier waste management in landfills.

For greater minimization of long-term mercury emissions from landfilling of *s/s* products, control of amounts of penetration should be discussed in terms of whether or not it would be effective at delaying migration of Hg via both leachates and gases.

#### 4. Conclusions

In this study, a column leaching test for the solidified product of metacinnabar, which was obtained by stabilizing metallic mercury, was conducted in monofill conditions with a granite soil layer. With unsaturated-water flow of 4.5 mm/day, the column entered conditions of oxidation with a natural pH. The results of thermodynamical analysis considering leachate quality show that the Hg in the leachate could be chlorides, indicating that the metacinnabar was soluble in pore water. However, the emissions of soluble Hg via leachates and gaseous Hg were extremely low because of Hg adsorption in the granite soil. Almost all Hg released from the *s/s* products existed in the soil within 1.5 cm away from it. Moreover, the deeper layers of soil contained more Hg than the upper ones. It seems that migration of Hg out of the columns was delayed due to adsorption/desorption of soluble Hg and dissolution/evaporation of gaseous Hg between the soil, moisture, and pores in unsaturated water flow. The sulfur polymer immobilized Hg in the *s/s* product. However, both the sulfur polymer and low-alkali cement *s/s* products could achieve effective containment of mercury-containing waste under monofill conditions with a Hg-adsorbing soil layer. It is important not only to stabilize/solidify mercury but also to install a Hg-adsorption layer surrounding the treated waste to minimize Hg emissions from landfills.

**Table 5** Distribution and content of Hg in soil in each zone at the end of the column leaching test.

Zone in column	Distribution of Hg (%)				Hg Content ( $\mu\text{g/g-wet}$ )			
	SP	CA	CB	CB+	SP	CA	CB	CB+
Surrounding part <sup>*1</sup>	58	70	64	57	1.7	19.9	5.7	7.8
Upper part <sup>*2</sup>	0.5	0.1	0.4	0.6	0.008-0.011	0.018-0.033	0.016-0.054	0.023-0.232
Horizontal part <sup>*3</sup>	17	19	26	31	0.059, 0.066	0.53, 1.1	0.32, 0.36	0.49, 0.81
Lower part <sup>*4</sup>	24	10	9.3	9.3	0.12-0.31	0.97-1.8	0.57-0.58	0.51-1.9

\*1 within 1.5 cm of the *s/s* product, \*2 at depths of 0-2.5 cm, \*3 over 1.5 cm horizontally from the *s/s* product at depths of 2.5-11.5 cm, \*4 depths of 11.5-15.0 cm

## Acknowledgement

This research was partially supported by the Environment Research and Technology Development Fund (3K153004 and 3-1802) of the Ministry of the Environment, Japan.

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(Received 10 December 2019, Accepted 16 May 2020)