

Leaching of Cs and Sr from Sewage Sludge Ash Buried in a Landfill Site

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Radionuclide contamination from the nuclear accident at the Fukushima Daiichi Nuclear Power Plant has been found in sewage sludge ash produced in eastern Japan. When such contaminated waste contains less than 8,000 Bq/kg radiocesium, it is being disposed in controlled landfill sites. In order to assess the possible spread of the radionuclides by their leaching from the landfill sites, it is important to know the leaching behavior of the radionuclides from the sewage sludge ash and factors influencing the leaching behavior. In this study, leaching experiments using stable Cs and Sr were conducted for sewage sludge ash under several conditions to investigate effects of chemical composition of leachate, pH, and solid/liquid ratio on Cs and Sr leaching behaviors. In the pH range from 6 to 12, the leaching ratio of Cs or Sr was less than 5.2 or 0.21%, respectively. Additionally, the leaching ratio of Sr decreased with increasing pH of the leachate. In contrast, the higher the pH in the leachate was, the higher the leaching ratio of Cs was. Finally, possible radionuclide leaching from contaminated sewage sludge ash and then radionuclide concentrations in an actual landfill leachate were assessed. It could be suggested that ⁹⁰Sr leaching from the landfill site had the least effect on the environment, whereas ¹³⁴+¹³⁷Cs leaching needed to be taken into account for spreading radioactive materials from the landfill site to the environment.

KEYWORDS: cesium, strontium, sewage sludge ash, leaching, landfill site, pH

I. Introduction

Radioactive materials were released into the environment during the Fukushima Daiichi Nuclear Power Plant Accident in 2011, and they migrated via various pathways such as water bodies, soil, and living organisms¹⁻⁴). Therefore, many places are currently suffering contamination problems. It has been reported that radioactive materials flowed into sewers and migrated into sewage sludge which was then converted to sewage sludge ash and condensed⁵), and periodic measurements of radioactive cesium (¹³⁴Cs and ¹³⁷Cs) concentrations in sewage sludge⁶) confirm this occurrence. However, concentrations differ depending on the area and the sewage system involved, and maximum concentrations have been reported as 446,000 Bq/kg in Fukushima Prefecture, 1,430 Bq/kg in Miyagi Prefecture, and

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2,510 Bq/kg in Iwate Prefecture⁷). The government permits the disposal of sewage sludge and sewage sludge ash (after appropriate treatment) with radioactive Cs concentrations under 8,000 Bq/kg in existing controlled landfill sites⁸, and landfill disposal of sewage sludge ash containing under 8,000 Bq/kg is occurring in accordance with this stipulation.

Leachate water is generated from controlled final disposal sites after rainwater infiltrates and passes through the waste (sludge ash, glasses, ceramics and concrete rubbish) and landfill deposited therein. Radioactivity has been reported in rainwater leachate from waste sites⁹. It is known that the leachate from sewage sludge ash contains much less radioactive Cs than that from general refuse incineration ash^{10, 11}. However, the leachate characteristics depend on the type of waste disposed of in the landfill site, and are also related to pH conditions, which generally range from neutral to alkaline, with high concentrations of salts with cations such as Ca¹²⁻¹⁴. To date, there have been no reports on the leachate properties of radioactive materials from sewage sludge ash with respect to alkaline or high salt concentration solutions. This study therefore aims to clarify the pH dependence (in a range of neutral to alkaline) of the solute content of leachate from sewage sludge ash generated in Iwate Prefecture and to determine the influence of the leachate salt type and solid/liquid ratio during leaching.

The radioactive materials of concern are radioactive Cs (¹³⁴Cs and ¹³⁷Cs) and radioactive Sr (⁹⁰Sr). As their concentrations in the sewage sludge ash used for experiments were too low for use in conducting direct measurements, their leaching properties were estimated by determining the leaching properties of stable isotopes ¹³³Cs and ⁸⁸Sr.

II. Experimental Methodology

1. Sampling Collection and Method of Analysis

This study used samples of sewage sludge ash from the Tonan Purification Center in Morioka Iwate Prefecture. The concentrations of Fe, P, and Si in the sewage sludge ash were measured using a sequential X-ray fluorescence spectrometer (Shimadzu, XRF-1800). For other elements, the concentrations of Cs and Rb were measured using inductively coupled plasma mass spectrometry (ICP-MS, Thermo, iCAP Qs), and the concentrations of Al, Ca, K, Mg, Mn, and Sr were measured using inductively coupled plasma atomic emission spectrometry (ICP-AES, Shimadzu, ICPE9000) after decomposing each sample in accordance with the boiling and elution method and using nitric acid and hydrochloric acid in the sewage testing method¹⁵. The concentrations of elements within the sewage sludge ash are shown in **Table 1**, where it is evident that the content of Al, Ca, P, and Si was high.

2. Leaching Experiment

(1) Experiment to determine leaching equilibrium time

The batch leaching experiment involved conducting the following experiments to determine the time until equilibration was reached in the leaching of Cs and Sr from sewage sludge ash.

A mixture of 3 g of sewage sludge ash and 30 mL of leachate was prepared in a 50-mL polyethylene bottle, then shaken at 25 degrees C and 120 rpm. Ultrapure water, 10⁻⁴ mol/L NaOH solution, and 10⁻² mol/L Ca(OH)₂ solution were used as leaching liquids. After shaking, the samples were collected at various intervals ranging from 30 minutes to 96 hours. The samples were then filtered through a 0.45 μm membrane filter, element concentrations in the filtrate

Table 1 Element concentrations leaching from ash

| | [mg/kg] | | [g/kg] |
|----|---------|----|--------|
| Cs | 0.63 | Al | 39.7 |
| Rb | 8.27 | Ca | 96.4 |
| | | Fe | 33.3 |
| | | K | 6.37 |
| | | Mg | 28.4 |
| | | Mn | 1.34 |
| | | P | 89.5 |
| | | Si | 74.0 |
| | | Sr | 0.44 |

were measured using ICP-MS and ICP-AES, and the leaching ratio was calculated using the following equation,

$$\begin{aligned} & \text{Leaching Ratio [\%]} \\ & = \frac{\text{Amount of Element Leaching from Ash [mg/kg]}}{\text{Element Content in Ash used for Leaching Experiment [mg/kg]}} \times 100 \end{aligned} \quad (1)$$

Also, the pH of the filtrate was measured using a pH meter (TOADKK, HM-25R).

(2) Leaching experiment

To clarify the leaching of Cs and Sr from sewage sludge ash, a batch leaching experiment was conducted. Three solution compositions, containing either NaOH, KOH, or Ca(OH)₂ were used as the leaching liquids. The several initial molar concentrations (and pH) of each were as follows. NaOH: 10⁻⁴(6.7), 10⁻³(9.0), 10⁻²(11.5), 5 × 10⁻²(12.3) and 10⁻¹(12.6); KOH: 10⁻⁴(6.6), 10⁻³(8.0), 10⁻²(11.4), 0.5 × 10⁻¹(12.3), and 10⁻¹(12.6); and Ca(OH)₂: 10⁻⁵(7.2), 10⁻⁴(10.5), 10⁻³(11.9), and 10⁻²(12.9). An experiment using ultrapure water as the leaching liquid was also conducted.

A mixture of sewage sludge ash and leaching liquid (in a solid/liquid ratio of 0.1 (3 g: 30 mL)) was prepared in a 50 mL polyethylene bottle and then shaken by a shaker at 120 rpm and 25 degrees C. As leaching equilibration was reached within three days after shaking started (in accordance with information presented in Section III-1), the shaking duration was set at three days. After shaking, suction filtration of the sample was conducted using a 0.45-μm membrane filter, the element concentration in the filtrate was measured using ICP-MS and ICP-AES, and the leaching ratio was acquired.

Also, the pH of the filtrate was measured using a pH meter (TOADKK, HM-25R).

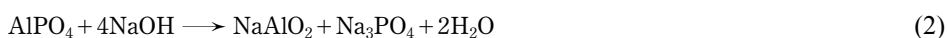
Observations of the influence of the solid/liquid ratio on the leaching ratio were conducted using the NaOH solution and KOH solution at 10⁻² mol/L and five varied solid/liquid ratios of sewage sludge ash and leaching liquids in the range of 0.01 to 0.5 (0.3 g: 30 mL, 1.5 g: 30 mL, 3 g: 30 mL, 9 g: 30 mL, 15 g: 30 mL). A similar experiment was conducted as described above and the leaching ratio was acquired.

III. Results and Discussions

1. Determination of Leaching Equilibration Time

The leaching ratios of Cs and Sr in the sample with respect to the shaking duration are shown in **Figure 1**, where it is evident that a constant leaching ratio was reached in approximately three days. The pH fluctuations in the samples were determined as follows: for the NaOH solution, the initial pH 7.1 stabilized at around 6.9 on the third day, and for the $\text{Ca}(\text{OH})_2$ solution, the initial pH 11.7 stabilized at around 8.6 within three days. From the above, it was considered that equilibration was reached after a shaking duration of three days.

The decrease in pH could be attributed to the dissolution of Al that existed in the sewage sludge ash, which caused the reaction $\text{Al}^{3+} + 4\text{OH}^- \rightarrow [\text{Al}(\text{OH})_4]^-$ ¹⁶⁾, or related to OH^- consumption by the leaching of AlPO_4 existing in the ash by the following reaction¹⁷⁾,



2. Leaching Properties of Cs and Sr

The leaching ratios of Cs at the pH of each leachate are shown in **Figure 2**, where it is evident that the leaching ratio is generally lower than 6% between pH 6 and 12, showing that leaching is restricted. Leaching ratios were approximately 0.4% for all leachates in the neutral pH range, but Cs showed a higher leaching ratio tendency with increasing pH. Furthermore,

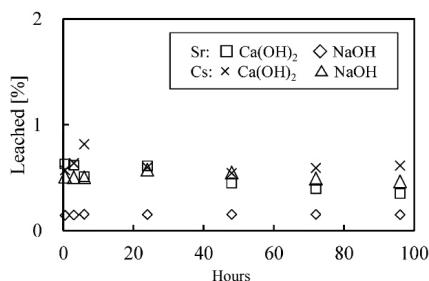


Figure 1 Effect of contact time on the leaching of Cs and Sr

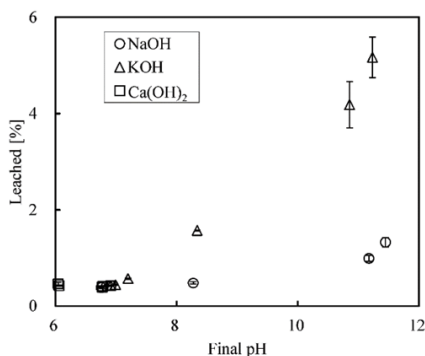


Figure 2 pH dependence of leaching of Cs (solid/liquid ratio = 0.1)

a higher leaching ratio was seen when leached with KOH solution than with NaOH solution, even at the same pH. This is because K has a behavior similar to Cs, and the exchange reaction is occurred between Cs existing in the ash in a comparatively mobile form and K in the leachate, so that the leaching rate of Cs may be high if the K concentration is high. To determine the influence of K concentration on Cs leaching, the leaching ratio between K and Cs with NaOH was determined by subtraction from that with KOH at almost equal final pHs (pH 6.9, 8.3, and 11.2). The value of $L_{\text{KOH-NaOH}}$ represents increments of the leaching ratio due to increases in the K concentration when the influence of pH is eliminated. **Figure 3** shows fluctuations of the $L_{\text{KOH-NaOH}}$ value related to concentrations of K. Linear regression shows that the rate of increase in the Cs leaching ratio relative to the increase in the K concentration can be estimated as 0.82% per K concentration increment of 0.01 mol/L (in the range of K concentrations lower than 0.05 mol/L).

Figure 4 shows the leaching ratio of Sr in leachate with varying pH. The leaching ratio of Sr was lower than 0.3% between pH 6–12, which is one order of magnitude lower than that of Cs. The pH dependence in the leaching of Sr shows a leaching ratio that decreases with increasing pH, unlike the behavior of Cs. The leaching ratio was less than 0.004% (and almost no leaching was observed) for all types of leachate at pH 8 and above. Also differing from Cs, there were no recognizable differences in the leaching ratios between different salt types of leaching liquids.

Figure 5 shows the leaching ratios of Cs and Sr congeners when NaOH is used for leaching. There are increases in the leaching ratios with Cs congeners K and Rb and increases in pH, but decreases in leaching ratios with congeners Mg and Ca and increases in pH, which is similar

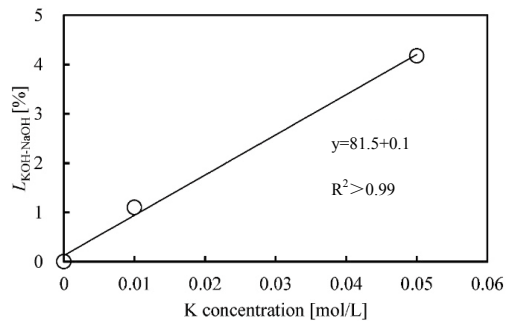


Figure 3 Effect of K concentration on $L_{\text{KOH-NaOH}}$ value

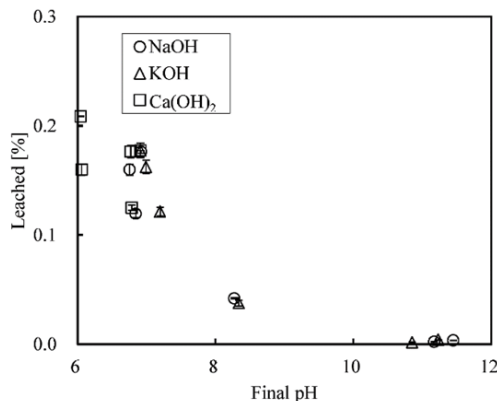


Figure 4 pH dependence of leaching of Sr (solid/liquid ratio = 0.1)

to the behavior of Sr.

Ca is reported to exist in sewage sludge ash as calcium phosphate ($\text{Ca}_3(\text{PO}_4)_2$) or hydroxyapatite ($\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$)^{17, 18}. The solubility products indicate low solubilities of 10^{-26} for $\text{Ca}_3(\text{PO}_4)_2$ and 10^{-108} – 10^{-128} for $\text{Ca}_{10}(\text{PO}_4)_6(\text{OH})_2$ and show higher values of still difficult solubilities with increasing pH, with a peak at around pH 9¹⁹. Sr and Ca show similar behavior, and therefore, Sr is considered to exist in ash as strontium phosphate ($\text{Sr}_3(\text{PO}_4)_2$), solubility product $10^{-27.4}$, and insoluble under alkaline conditions.

The possibility of Cs existing in sewage sludge ash as an aluminosilicate compound ($\text{CsAl-Si}_2\text{O}_6$, etc.) has also been reported¹¹. The leaching ratio of Al was 0.01% at pH 8.3, but increased to 14.7% at pH 11.4 when leached by an NaOH solution. These results indicate that some of the Al or Si within aluminosilicate compounds in the sewage sludge ash was dissolved due to the increased pH, and Cs was then accordingly leached.

3. Change in Leaching Ratio and pH in Relation to Solid/Liquid Ratio

A higher solid/liquid ratio is expected at final disposal sites where precipitation levels are high, as a larger amount of liquid will contact the solid phase over time. To confirm whether the leaching ratio varies with rainfall, we studied the influence of the solid/liquid ratio on the leaching ratio.

The leaching ratios of Cs and Sr with respect to the solid/liquid ratio are shown in **Figure 6**. It is evident that the leaching ratio of Cs decreased with an increase in the solid/liquid ratio, and

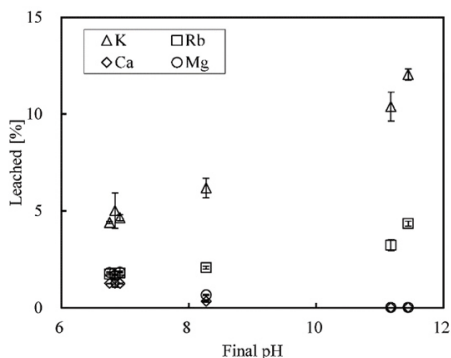


Figure 5 pH dependence of leaching of K, Rb, Ca, and Mg in NaOH solution

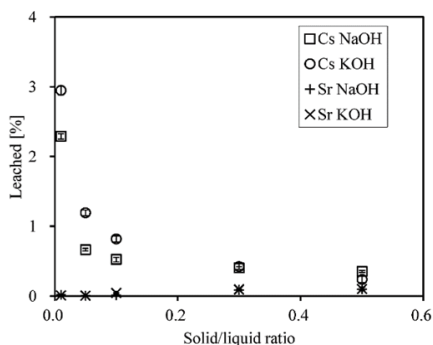


Figure 6 Solid/liquid ratio dependence of leaching of Cs and Sr

the leaching ratio was higher when the solid/liquid ratio was low and when the KOH solution was used compared to when leaching with the NaOH solution. However, no differences were seen between the leaching ratios of the two solutions when the solid/liquid ratio was 0.3 and over. The leaching ratio of Sr differed from that of Cs; although it showed a low leaching rate itself, this amount increased 10-fold within a solid/liquid ratio range of 0.01 to 0.5. Furthermore, no difference was observed in leaching ratios when leaching with either NaOH or KOH solution.

The pH decreased with an increase in the solid/liquid ratio; this is considered to be due to the higher OH^- consumption with a higher solid phase percentage via the reaction of $\text{Al}^{3+} + 4\text{OH}^- \rightarrow [\text{Al}(\text{OH})_4]^-$, which occurs when the ash and solution make contact. Additionally, the variation in leaching ratio with various solid/liquid ratios showed a match with the influence of pH on the leaching ratio for a solid/liquid ratio of 0.1, as shown in Figures 2 and 4, for Cs and Sr (Figures 7 and 8). Therefore, a study investigating leaching from ash was conducted to separately observe the influences of the solid/liquid ratio and the pH, where the leaching quantity per 1 L of liquid (L_1 [$\mu\text{g}/\text{L}$]) was first calculated using the Cs leaching ratio (L [%]) when leached at various NaOH concentrations, and then with a constant solid/liquid ratio of 0.1. Figures 2 and 4 show the relationship between pH and the leaching ratio (L [%]) for Cs and Sr when leached with the NaOH solution, and this can be expressed by the following equation as

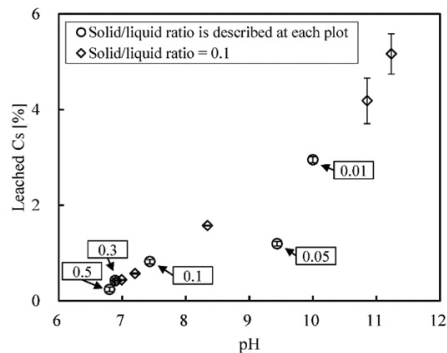


Figure 7 Comparison of leaching of Cs with pH at different experimental conditions using KOH as leaching solution

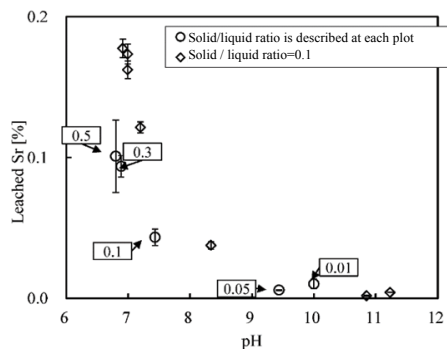


Figure 8 Comparison of leaching of Sr with pH at different experimental conditions using KOH as leaching solution

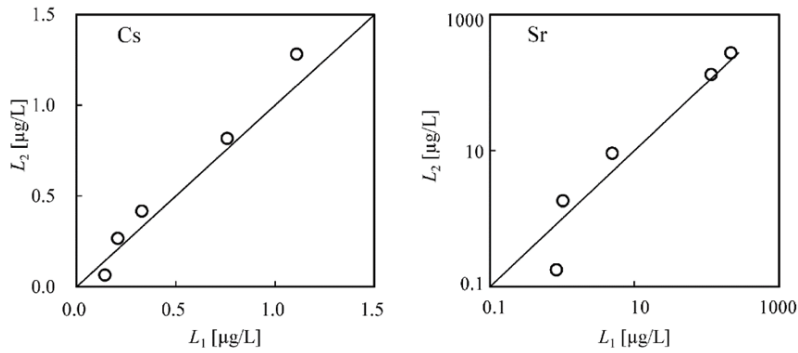


Figure 9 Relationships between L_1 and L_2 values

$$\text{Cs} : L = 0.076e^{0.24\text{pH}} \quad (R^2 = 0.96) \quad (3)$$

$$\text{Sr} : L = 62.47e^{-0.88\text{pH}} \quad (R^2 = 0.98) \quad (4)$$

Using the leaching ratio acquired by substituting the equations above with the final pH at various solid/liquid ratios, the leached quantity (L_2 [$\mu\text{g/L}$]) per liquid quantity was calculated. The relationship of L_1 and L_2 is shown in **Figure 9**. As the ratios of L_1 and L_2 were approximately 1 for both Cs and Sr, the behavior of leaching from sewage sludge ash can only be explained with respect to the influence of pH. It is thus indicated that pH, rather than the solid/liquid ratio, is the dominant factor in the leaching of Cs and Sr from sewage sludge ash.

4. Quantity Leached from Sewage Sludge Ash in Landfill Site

Based on this study, the amounts of radioactive Cs and Sr leached in leachate, and their concentrations in leachate, were evaluated at a landfill site where sewage sludge ash was deposited. First, the leaching ratio was determined, and the leaching experiment indicated that the leaching ratio of Cs and Sr from sewage sludge ash is dependent on pH. In addition to the actual pH of leachate at landfill sites, which differs greatly between sites, from near pH 7²⁰⁾ to approximately pH 6–8¹⁴⁾ or pH 11.5–11.8²¹⁾, inter-annual variability may have an influence. Accordingly, as an evaluation on the safe side, the highest Cs leaching ratio of 5.2% (solid/liquid ratio 0.1, KOH 10^{-1} mol/L) and Sr leaching ratio of 0.18% (solid/liquid ratio 0.1, ultrapure water) were adopted for this calculation.

The upper limit for radioactive Cs in the waste permitted in landfill at existing controlled final disposal sites is 8,000 Bq/kg. For sewage sludge ash containing radioactive Cs producing 8,000 Bq/kg, the highest leaching rate (L_{max}) in this experiment was 5.2%; therefore, the maximum quantity of leaching radioactive Cs was 416 Bq/kg. One study determined that the optimum moisture content ($w = (\text{moisture weight [g]}/\text{soil weight [g]}) \times 100$) for compacting sewage sludge ash is 58%²²⁾; therefore, the maximum radioactive Cs concentration (C_{max}) in the leachate water was estimated as 717 Bq/L using the equation below

$$C_{\text{max}} = \frac{8000 \times L_{\text{max}}}{w/100} \quad (5)$$

The standard values for radioactive material concentration in landfill effluent are 60 Bq/L for ^{134}Cs and 90 Bq/L for ^{137}Cs ⁷⁾; therefore, our estimated concentration exceeds the effluent standard. Consequently, the possibility of leachate containing a high radioactive Cs concentration

that exceeds the effluent standard value is indicated if it contains a high concentration of radioactive Cs, even in sewage sludge ash with a low Cs leaching ratio.

Sewage sludge ash is disposed of as landfill as a three-meter-thick swathe that is covered by a soil layer measuring 50 cm to retain radioactive Cs, or by proper treatment of radioactive Cs and leachate treatment. Using the experimental results and data from existing research, the concentration of radioactive Cs in the leachate generated from a final disposal site where ash is disposed of as landfill was estimated and studied. Various waste in addition to sewage sludge ash is disposed of at final disposal sites, and therefore leaching from these wastes also needs to be considered. However, data regarding the leaching of ^{133}Cs from other waste, such as cement-type material, has not been reported and is thus difficult to consider. Ishikawa et al.²³⁾ made several measurements of ^{133}Cs in leachate at a controlled final disposal site where sewage sludge ash was disposed of as landfill and reported an average of 2.78 $\mu\text{g/L}$, while the concentration of ^{133}Cs leached from sewage sludge ash measured in this research was 3.4 $\mu\text{g/L}$. Furthermore, the concentration of K (a congener of Cs that has an adsorbing behavior similar to soil) has been reported as $423 \pm 172 \mu\text{g/L}$ ²⁰⁾ for leachate, while the maximum leaching concentration of K was 806 mg/L in the sewage sludge ash acquired in this research. It is considered that the rate of reduction in the Cs or K concentration in leachate occurs when the salt concentration in the leachate is high; the adsorption of Cs or K to soil is low when the leachate infiltrates the soil layer because of competition with other elements present in leachate. The result shows the same order for the leaching concentration, and the concentration in the leachate implies a certain level of reasonability. From the evaluation conducted in this research, ^{133}Cs in leachate is assumed to be derived from sewage sludge ash.

The highest reported ^{133}Cs concentration in sewage sludge ash was approximately 0.85 mg/kg²⁴⁾, and based on this result, the ratio of ^{133}Cs /radioactive Cs can be determined to be 0.106 mg/Bq when sewage sludge ash of 8,000 Bq/kg is disposed of as landfill. The maximum concentration of radioactive Cs (C_{max}') in leachate can then be calculated using the formula below,

$$C_{\text{max}}' = \frac{2.78}{0.106} = 26.2 \quad (6)$$

C_{max}' was estimated as 26.2 Bq/L, which does not exceed the effluent standard, and this result indicates that the current prevailing method for retaining radioactive Cs using a soil layer is working.

An increase in the pH of leachate has been observed in the dry season when there is less rainfall at final disposal sites²⁵⁾. The pH of leachate lowers during heavy rain, and the leaching ratio from ash is thus accordingly lower; therefore, it is not possible for the concentration in the leachate to exceed the level evaluated above.

The effluent standard for radioactive Sr is not stipulated, but the concentration limit for ^{90}Sr in effluent has been defined as 30 Bq/L in an announcement that specifies the dose limits, etc. in accordance with stipulations relating to the installation or operation of nuclear reactors used for experiments and research. With respect to the Fukushima Daiichi Nuclear Power Plant Accident, the $^{90}\text{Sr}/^{137}\text{Cs}$ ratio in contaminated material is estimated as 3×10^{-3} ²⁶⁾, and if it is assumed that all of the 8,000 Bq/kg of radioactive Cs consists of ^{137}Cs , the concentration of ^{90}Sr is thus assumed to be 24 Bq/kg. The highest leaching ratio of Sr in this experiment was 0.18%, therefore the leaching rate of radioactive Sr estimated by this research is 43.2 mBq/kg, and the concentration in the leachate is 74 mBq/L if the optimum moisture content above is considered. This value is thus lower than the limit concentration in effluent, as explained above.

This shows that the contribution of ^{90}Sr to the leaching of radioactive materials from the landfill site to the leachate is extremely small (for sewage sludge ash containing radioactive materials), and results indicate that it is only necessary to consider the effects of ^{137}Cs .

IV. Summary

In this research, a series of batch leaching experiments was conducted to determine the leaching properties of Cs and Sr from sewage sludge ash. The leaching ratio of Sr was found to be 0.21–0.004% with pH 6–12. As with Ca, a higher pH inhibited sewage sludge ash leaching, and almost no leaching occurred with a pH of 8 and above. In contrast to Sr, Cs showed an increased leaching ratio with higher pH: the maximum leaching ratio was 5.16% with pH 11.2. These results show that it is mainly necessary to consider the leaching of radioactive Cs to prevent excessive radioactive material from escaping in the leachate when sewage sludge ash is disposed of as landfill.

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