Weathering Effects on Fractionation Change of Cesium in Soil

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1. Introduction

After nuclear accidents in Chernobyl and Fukushima, concerns about the radionuclides, strontium(Sr), iodine(I), and cesium(Cs), have increased. In particular, Cs is one of the most important dose-forming long lived radionuclides because of relatively long half-life (30 years) and its emission of gamma radiation [1]. When it is exposed to soil, Cs is strongly adsorbed on clay minerals due to the specific adsorption sites in clay minerals [2]. In general, Cs on soil exists as a residual fraction among four fractions: exchangeable, reducible, oxidizable, and residual fraction [3]. The fractionation of Cs could be changed by exposure time or repeated wet/dry conditions, that is, an ageing effect. In this study, ageing effects were investigated due to exposure time and repeated wet/dry conditions using soil samples and clay minerals.

2. Materials and Methods

2.1 Materials

In this study, 5 type soil samples, 3 type soils and 2 type clay minerals, were used. For the representative clay minerals in Korea, illite (K_{0.8}- $_{0.9}$ (Al,Fe,Mg)₂(Si,Al)₄O₁₀(OH)₂, Yong-koong illite company, Korea) and kaolinite (Al₂O₃·2SiO₂·2H₂O, Sigma-aldrich, USA), were used. CsCl (Sigmaaldrich, USA), a nonradioactive Cs (¹³³Cs), was used to artificially contaminate the soil. Potassium chloride (\geq 99.0% KCl, Sigma-aldrich, USA) was used as background solutions in wet/dry repetition.

2.2 Methods

In the experiments for the exposure time of Cs, the soil was artificially contaminated by Cs during a day, week, month, and 3 months. A mass of 3 g of soil was mixed with 30 mL Cs solution (10 mg/L) in 50 mL centrifuge tube at room temperature. In the

experiments for the repetitions of wet/dry condition, the soil was artificially contaminated by Cs during 24 hours. After artificial contamination, the soil and solution were separated by a filter paper (< 0.45 μ m). After then, the soil was dried at 50°C for 24 hours (dry condition) and the dried soil was mixed with 30 mL of 5 mM KCl solution for 24 hours (wet condition). The dry and wet according to the experimental condition were repeated to 5, 10, and 15 times, respectively. The fractionation analysis of Cs was based on the sequential extraction method called as SM&T (Standards, measurement and testing programme) method (Table 1).

Table 1. Sequential extraction condition by SM&T method

Fraction	Extractable phase	Reagents
F1	Exchangeable	0.11 M CH ₃ COOH
F2	Reducible	0.5 M NH ₂ OH·HCl at pH 5
F3	Oxidizable	8.8 M H ₂ O ₂ , 1 M CH ₃ COONH ₄
F4	Residual	Aqua regia

3. Results and discussion

In the result of exposure time test, the fractionation change was confirmed in sample 1 and 3. F1 in sample 1, 2, and 3 decreased while F4 increased. In illite and kaolinite, fractionation change was not shown (Fig. 3). However, F4 in sample 1, 2, and 3 increased according to wet/dry repetitions. It seems that a section of F1 or 2 is changed to F4 by wet/dry repetitions. In other words, Cs could be moved to the soil particle part where is difficult to be desorbed. But F4 in clay minerals, illite and kaoilinte, was not increased (Fig. 4). Actually, most radiocesium in Chernobyl exists as residual fraction. However, as compared to the results of this study, it is estimated that the Cs fractionation would be changed according to wet/dry conditions.

4. Conclusions

In this study, we investigated the fractionation change of Cs in soil environments according to the aging effect, including the exposure time and wet/dry condition. According to the exposure time of Cs, F4 was slowly increased in sample 1, 2, and 3, but F4 in clay minerals were not changed. However, wet/dry repetitions influenced the fractionation change of Cs in sample 1, 2, and 3, but F4 in clay minerals were not changed. It is estimated that F1 or F2 could be changed to F4 by wet/dry repetitions, that is, it means that Cs could be moved to the soil particle part where is difficult to desorb Cs. Therefore, we expected that the fractionation of Cs would be transformed to stable fraction from weakly bound fraction by aging.

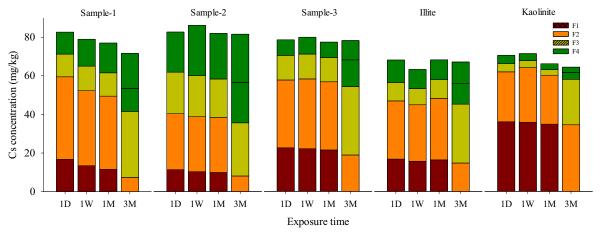


Fig. 3. Fractionation change of Cs according to the exposure time. The exposure time are 1 day, 1 week, 1 month, and 3 months which are 1D, 1W, 1M, and 3M, respectively.

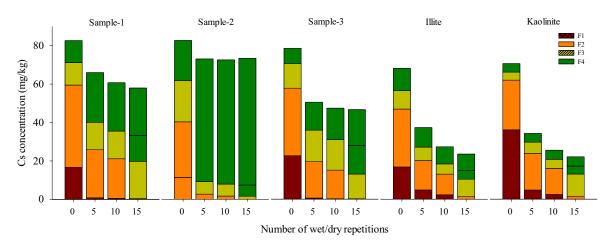


Fig. 4. Fractionation change of Cs according to the wet/dry conditions. Wet condition is 5 mM KCl solution for 24h and dry condition is 50°C for 24h.

REFERENCES

- T. Saiot, H. Makino, and S. Tanaka, "Geochemical and grain-size distribution of radioactive and stable cesium in Fukushima soils: implications for their long-term behavior", Journal of Environmental Radioactivity, 138, 11-18 (2014).
- [2] K. Murota, T. Saito, and S. Tanaka, "Desorption

kinetics of cesium from Fukushima soils", Journal of Environmental Radioactivity, 153, 134-140 (2016).

[3] X.L. Hou, C.L. Fogh, J. Kucera, K.G. Andersson, H. Dahlgaard, and S.P. Nielsen, "Idoine-129 and Cesium-137 in Chernobyl contaminated soil and their chemical fractionation", Science of Total Environments, 308, 97-109 (2003).