Chemical equilibrium modelling of radiocesium elution from contaminated litter and its humus into water phase

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Elution of radiocesium (137 Cs) from litter and its humus in 137 Cs-contaminated forest can be a key process determining a discharge of 137 Cs from forests into rivers around the Fukushima Dai-ichi Nuclear Power Station (FDNPS). Quantitative understanding of this process is then necessary for predicting the contamination situation in the future. However, chemical equilibrium modelling of 137 Cs in the elution has not been addressed. The influence of solution conditions such as pH, coexisting cations, and dissolved organic matter (DOM) on the elution is still unclear. In this study, degree of the elution of 137 Cs from litter (*i.e.*, fallen leaves) or its humus in the contaminated forest near the FDNPS was evaluated by a batch method and was then compared with those from the calculation with NICA-Donnan model, assuming a ternary system of Cs, DOM, and solid organic matter (SOM).

The degrees of the elution of ¹³⁷Cs from the litter and the humus were 22% and 6-12%, respectively. The pH values in the eluates were 4.4 for the litter and 5.8-6.4 for the humus. The DOM concentration were 2306 mg-C L^{-1} for the litter and 690–750 mg-C L^{-1} for the humus. Coelution of cations such as ¹³³Cs, Rb, K, Na, Ca, Mg, Fe, Al were also observed. The experimentally obtained logarithmic distribution coefficients of ¹³⁷Cs for the litter and the humus (log $K_{d exp}$) were evaluated to be 1.6 L kg-C⁻¹ and 2.0–2.2 L kg-C⁻¹, respectively. These values corresponded with those of ¹³³Cs in both the experiments and the calculations (*i.e.*, $\log K_{d \text{ cal.}} = 1.5 \text{ L kg-C}^{-1}$ for the litter and 2.1 L $kg-C^{-1}$ for the humus). The concentrations of the coeluted cations, i.e., Rb, K, Na, Ca, Mg, Al, were also predicted by the calculations. Besides, the calculations showed that the degree of the elution can be dominated by the competition in the distribution of ¹³⁷Cs between DOM and SOM. Thus, these suggest that the ternary model using NICA-Donnan model can be useful for predicting and understanding the elution of ¹³⁷Cs from the contaminated litter and its humus.