汚染リター中から分離した有機質コロイド画分に含まれる放射性 Cs
Distribution of radioactive cesium in organic colloid fractionated from polluted litter

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1.　　　Introduction

　　　After 3 years from the nuclear power plant (NPP) accident in March 11th, 2011, radioactive cesium concentration is still high in surface soil and litter layer at Iitate village in Fukushima prefecture. Mobility of cesium was mainly related to its sorption to mobile inorganic and organic colloids in soil. The sorption of cesium to inorganic colloid is almost irreversible but the sorption to organic colloid is reversible via ion exchange and decomposition or transformation of organic matter, therefore, cesium that bound to mobile organic colloids may be the main source of transferable cesium to surface, subsurface water, and plants. Based on our survey, there is a little knowledge on the bounding of radioactive cesium to organic colloid in polluted soil and litter, thus this paper aims to clarify the distribution of radioactive cesium on organic colloid from polluted litter at Fukushima prefecture.

2.　　　Materials and methods

　　　Cesium-polluted litter was collected from a cleared forest at Fukaya, Iitate village which is located 40 kilometers northwest of the Fukushima Daiichi NPP. The litter sample was then sieved through 2.0-mm mesh. Thirty gram of litter was mixed with 400 ml deionized water and adjusted to pH 8 using 0.1M NaOH solution. The mixture was then shaken for 24 hour and passed through 53-μm mesh. The residue was washed with100 ml of pH 8 solution.

　　　The filtrate solution was adjusted to pH 8 and transferred to 1000-ml cylinder for gravitational fractionation. A surface to 4-cm deep portion of the suspension in the cylinder was decanted every 4 hours and defined as an organic colloid solution. The gravitational fractionation was repeated several times after adding deionized water and adjustment to pH 8. Each 40-ml organic colloid solution was then centrifuged for 5 and 10 minutes at the speed of 500, 1000, 2500, 5000 and 10000 rpm, respectively. The litter, litter residue, sediment, colloid solutions were analyzed for total carbon, total nitrogen, Cesium-137 and Cesium-134. Particle size distribution and colloid concentration of all colloid solution were also determined using nanoparticle size analyzer (SALD-7500 nano, Shimadzu Co., Ltd), and oven dry method, respectively.

3.　　　Results and discussion

3.1　　　Distribution of radioactive cesium in polluted litter
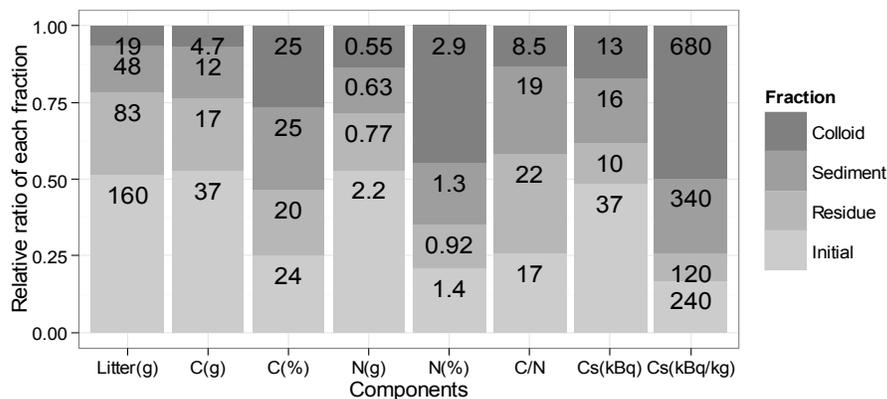

Figure 1 Distribution of cesium, carbon and nitrogen before (initial litter) and after fractionation at pH8 (dry colloid, residue and sediment). The values inside the bar indicate the mass and concentration of each components in x-axis. Sum of all fractions are normalized to unity and their relative fractions are presented in y-axis.

The distribution of each components after fractionation at pH8 as well as their relative mass and concentration as compared to the initial litter are shown in Figure 1. Mass of organic colloid in the colloid solution was 19 gram which accounted for 8 % of the initial litter mass (160 gram), however it contained 30% of total cesium in the initial litter. The cesium concentration in organic colloid was 2.8 times higher than that in original litter. The differences in total carbon content among all fractions such as colloid and initial litter and sediment were not obvious except for the low organic carbon content of residue (20%), however the concentration of total radioactive cesium were largely different among them. It suggested that the carbon in these fractions could be originated in different humus type or formulation in the litter. Humus acid in organic colloid are from the fraction that weakly bound to litter. Low C/N ratio in colloid indicated these humus acids are more degraded type.

3.2   Particle size distribution and particle density of organic colloid

The average diameters of organic colloid ranged from 6.4 – 12 μm and 0.5 – 10.4 μm for gravitational and centrifugal fractionation conditions, respectively in this study. After the fractionations, the colloid particle densities were estimated based on Stokes' law (Eq.1), and Eq.2, respectively for both gravitational and centrifugal methods.

$$\rho_s = \rho_l + \frac{18\eta h}{gtd^2} \quad \text{(Eq.1)}$$

$$\rho_s = \frac{18\eta \ln\frac{R}{S}}{\left(\frac{2\pi}{60}rpm\right)^2 d^2 t} + \rho_l \quad \text{(Eq.2)}$$

where

ρ_s is colloid particle density. ρ_l is water density. d is colloid diameter. t is gravitational or centrifugal time. h is travel depth of colloid particle. rpm is centrifugal speed. η is water viscosity. g is gravitational acceleration. R is the distance from the axis of rotation to the level from where the supernant is decanted. S is the distance from the axis of rotation to the surface of the suspension in the centrifugal tube.
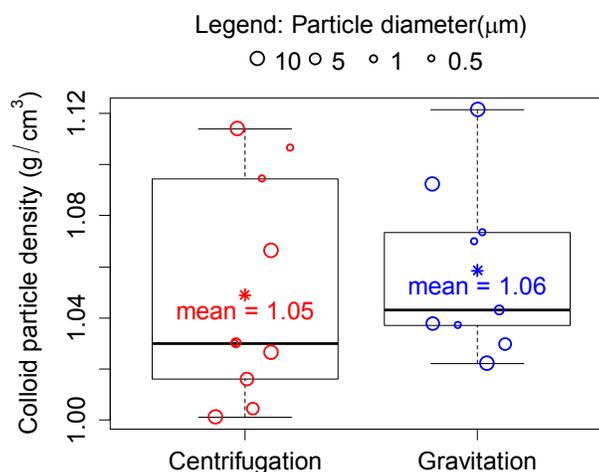

Figure 2 Boxplot of estimated organic colloid particle density using centrifugal and gravitational fractionation methods.

The estimated colloid particle density was 1.06 g/cm^3 which was lower than the conventional particle density of organic matter in soil 1.4 g/cm^3 as reported by Mayer et al. (2004) and Wagai et al. (2009). The result suggested that the humus acid in organic colloid could be originated from low-density fraction of organic matter in litter.

4.   Conclusions

Organic colloid accounted for 8% of total litter mass and 30 % of total radioactive cesium in polluted litter. Cesium concentration in organic colloid was 2.8 times higher than that in original polluted litter. Organic colloid was originated from low-density organic matter in litter and more likely from degraded fraction. Its particle density was about 1.06 g/cm^3.

References

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Wagai R. et al., Soil Sci. Plant Nutr. **55**(1), 13-25 (2009).

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