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Short communication

Relationship between the radiocesium interception potential and the transfer of radiocesium from soil to soybean cultivated in 2011 in Fukushima Prefecture, Japan

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The concentration of radiocesium (^{134}Cs and ^{137}Cs) in agricultural fields around Fukushima Dai-ichi Nuclear Power Plant (FDNPP) was elevated after the accident in March 2011. Evaluation of soil properties that influence phytoavailability of radiocesium is important for optimal soil management to minimize radiocesium transfer to crops. In this study, soybean grain and soil samples (0–15 cm) were collected from 46 locations in Fukushima Prefecture in 2011, and ^{137}Cs concentrations were measured. ^{137}Cs concentration ranges were 11–329 Bq kg^{-1}-dry in soybean grain samples, and 0.29–2.49 kBq kg^{-1}-dry in soil samples. The radiocesium interception potential (RIP) values in the soil samples ranged from 0.30 to 8.61 mol kg^{-1}. RIP negatively correlated with total carbon content and oxalate-extractable Si and Al + 1/2 Fe in the soils, suggesting that soils rich in organic matter and poorly crystalline clays tended to have lower RIP in this region. The soil-to-plant transfer factor for ^{137}Cs, analyzed in relation with various soil characteristics, varied by two orders of magnitude and was significantly negatively correlated with RIP and exchangeable K concentration in soil. The results show that RIP is useful for evaluating the efficiency of radiocesium transfer from soil to plants in this region.

1. Introduction

Large amounts of radiocesium including ^{134}Cs (half-life 2.1 y) and ^{137}Cs (half-life 30.1 y) were accidentally released from the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) following the Great Eastern Japan Earthquake and the massive tsunami on March 11, 2011. The amounts of released ^{137}Cs have been estimated as 1.3×10^{16} Bq (Chino et al., 2011), with approximately 22% deposited over Japanese land areas (Morino et al., 2011).

Radiocesium is strongly retained by clay minerals in soil, and remains in the surface soil for a long time after its deposition. Although the mobility of radiocesium in soil is rather low, it is transferred to the edible parts of crops via root uptake in agricultural fields. The soil-to-plant transfer factor (TF) is an empirical parameter generally defined as the ratio of Cs concentration in the edible parts of a crop plant to that in soil (in a field or in a pot containing a radioisotope tracer). The reported TFs vary up to three orders of magnitude even for the same soil–crop combinations (Ehlken and Kirchner, 2002). TF is largely affected by K^+ concentration in soil solution and the labile radiocesium distribution coefficient, which can be estimated on the basis of soil properties associated with exchangeable K status and the radiocesium retention ability of soil clay (Absalom et al., 1999; Tarsitano et al., 2011). Radiocesium is strongly and irreversibly adsorbed by micaceous clay minerals. The weathering fronts in the mineral interlayers expanding into wedge shapes are known as the frayed-edge sites (FESs). The radiocesium interception potential (RIP) has been established as a quantitative indicator of FES in soil (Cremers et al., 1988). The RIP is defined as the product of the selectivity coefficient of Cs to K in the FES ($K_{c(Cs-K)}^{FES}$) and the FES capacity ([FES]) as shown in the following equation (Wauters et al., 1996):

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$$\mathrm{RIP} = K_{c(\mathrm{Cs}-K)}^{\mathrm{FES}}[\mathrm{FES}] = K_D^{\mathrm{Cs}}\mathrm{mK}\left(\mathrm{mol\ kg}^{-1}\right) \quad (1)$$

The product is calculated from the solid–liquid distribution coefficient for ^{137}Cs (K_D^{Cs}, L kg^{-1}) in a solution with specific K^+ and Ca^{2+} ionic strength, and K concentration (mK, mol L^{-1}) in the solution.

Vandebroek et al. (2012) measured the RIP values for a wide variety of soils collected worldwide, and found the lowest values in Andosols, Podzols and Ferralsols. Andosols, mainly formed on volcanic ejecta, are found in volcanic regions all over the world. Although the total area of Andosols is estimated at less than 1% of the global land surface (IUSS Working Group, 2006), it is an important soil type for upland farming in Japan, occupying almost half of the total upland field area (Saigusa and Matsuyama, 1998). Vandebroek et al. (2012) also showed that RIP is highly variable in each soil type; thus, it is difficult to predict the RIP value from the soil type. Delvaux et al. (2000) conducted a small-scale ryegrass cultivation experiment using a wide variety of soils containing a ^{137}Cs tracer. The soil-to-plant Cs transfer clearly negatively correlated with the soil RIP values, and more efficient transfer was observed in soils with lower RIP. This result suggested that RIP is the major parameter that reflects the availability of radiocesium in soil. Nakao et al. (2014) reported the RIP values of clay fractions isolated from 97 paddy soils in Fukushima Prefecture and found a large contribution of micaceous minerals to the radiocesium retention ability of soil clays in this region. However, the relationship between RIP and TF of radiocesium derived from the FDNPP accident is still not clear.

The objectives of this study were (1) to determine the RIP values of arable soils contaminated with radiocesium from the FDNPP accident, and to evaluate soil properties affecting the radiocesium retention ability; and (2) to clarify the soil factors controlling variance in TF for radiocesium from soil to soybean grains cultivated in the first year after the accident for better TF prediction and effective soil management to reduce radiocesium in the crop.

2. Materials and methods

Soil plowing and soybean sowing were conducted from the end of May to the end of June 2011, and the growing period was approximately 5 months. Forty six soybean grain and surface soil (0–15 cm) sample sets were collected from agricultural fields located 20–83 km away from FDNPP (Fig. 1). Five subsamples of soils and crops were collected and mixed to produce a composite sample for each field. Soil properties for each sample are shown in Table 1. Soil pH (measured at a soil-to-water ratio of 1:2.5) ranged from 5.2 to 7.9. Total carbon content in soil, measured by the dry combustion method (FLASH2000, Thermo Fisher Scientific Inc, Waltham, MA, USA), ranged from 2.5 to 56 g kg^{-1}. The oxalate-extractable Si (Si_o), Al (Al_o), and Fe (Fe_o) were extracted using the methods described by Blakemore et al. (1981) and determined using inductively coupled plasma atomic emission spectrometry (Vista, Varian, Palo Alto, CA, USA).

In the region where the samples were collected, most soils are classified as Fluvisols, Gleysols or Cambisols; five soil samples satisfied the criterion for andic soil properties ($Al_o + 1/2\ Fe_o > 20$ g kg^{-1}) (IUSS Working Group, 2006). Exchangeable K and cation exchange capacity (CEC) were measured after saturation with 1 mol L^{-1} ammonium acetate solution (pH 7). Potassium saturation ranged from 0.5 to 12.5% of CEC.

The soil and soybean grain samples were packed in individual plastic containers. Radioactivity of ^{137}Cs was measured with Ge gamma-ray detectors (GC3020, GC4020, GC3520; Canberra, Meriden, CT, USA) connected to multichannel analyzers. The detectors

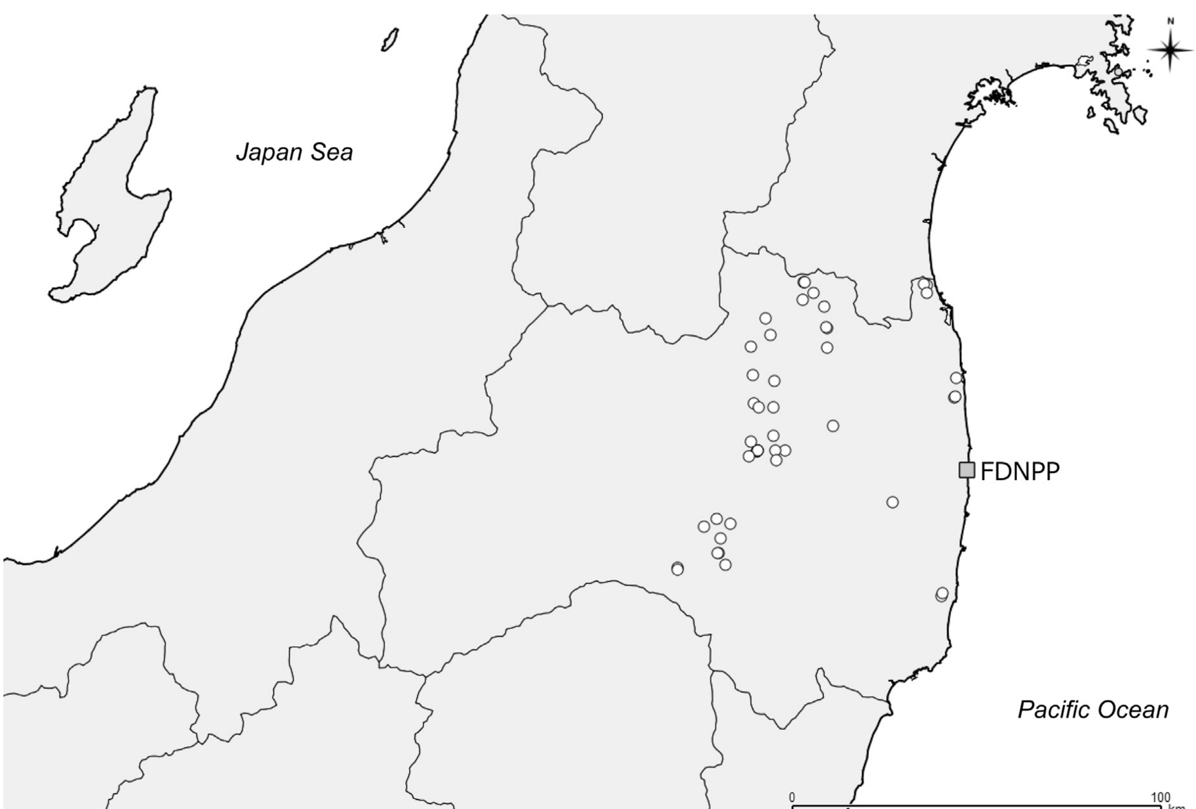

Fig. 1. Soil sampling sites (circles) in Fukushima Prefecture, Japan.