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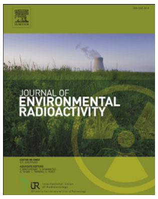

Variability of the soil-to-plant radiocaesium transfer factor for Japanese soils predicted with soil and plant properties

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ABSTRACT

Food chain contamination with radiocaesium (RCs) in the aftermath of the Fukushima accident calls for an analysis of the specific factors that control the RCs transfer. Here, soil-to-plant transfer factors (TF) of RCs for grass were predicted from the potassium concentration in soil solution (m_K) and the Radiocaesium Interception Potential (RIP) of the soil using existing mechanistic models. The m_K and RIP were (a) either measured for 37 topsoils collected from the Fukushima accident affected area or (b) predicted from the soil clay content and the soil exchangeable potassium content using the models that had been calibrated for European soils. An average ammonium concentration was used throughout in the prediction. The measured RIP ranged 14-fold and measured m_K varied 37-fold among the soils. The measured RIP was lower than the RIP predicted from the soil clay content likely due to the lower content of weathered micas in the clay fraction of Japanese soils. Also the measured m_K was lower than that predicted. As a result, the predicted TFs relying on the measured RIP and m_K were, on average, about 22-fold larger than the TFs predicted using the European calibrated models. The geometric mean of the measured TFs for grass in the affected area ($N = 82$) was in the middle of both. The TFs were poorly related to soil classification classes, likely because soil fertility (m_K) was obscuring the effects of the soil classification related to the soil mineralogy (RIP). This study suggests that, on average, Japanese soils are more vulnerable than European soils at equal soil clay and exchangeable K content. The affected regions will be targeted for refined model validation.

1. Introduction

The mechanisms of soil-to-plant transfer of radiocaesium (both ^{137}Cs and ^{134}Cs; hereafter RCs) are crucial for assessing the long-term risks in agricultural ecosystems in the area affected by the accident at Fukushima in March 2011. Caesium has a relatively high solubility in soil solution and Cs present in soil solution is taken up by plants through its analogy with potassium (K) (White and Broadley, 2000). The plant availability of RCs from soils is generally assessed in terms of the soil-to-plant transfer factor (TF), which is defined as (IAEA, 2009);

$$\mathrm{TF} = \frac{\text{RCs concentration in plant } (\text{Bq kg}^{-1} \text{ dry weight})}{\text{RCs concentration in soil } (\text{Bq kg}^{-1} \text{ dry weight})} \left(\text{kg kg}^{-1}\right) \tag{1}$$

The TF is a simple but useful parameter for evaluating the risks of agricultural food chain contamination after a large-scale soil contamination in the Fukushima affected area and for the estimation of the resulting radiation dose to the public.

The TFs of RCs have been collected for a wide range of soil-plant combinations (IAEA, 2009), revealing a high variability. Even for each soil-plant type combination, the TF is still highly variable, covering up to three orders of magnitude. Nisbet et al. (1999) reported that simple regressions linking the TF with single soil properties yield poor models for quantitative prediction of the TF. Studies with Belgian soils unravelled that plant availability of RCs

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can be predicted with RCs and K^+ concentrations in soil solution (Smolders et al., 1997). Since then, the TF for RCs has been predicted in a mechanistic way using soil and plant characteristics (Absalom et al., 1999, 2001; Tarsitano et al., 2011). Absalom et al. (1999) reported that the predicted TF for ryegrass explained 68% of variation of the measured TF (in the log TF- log TF relationship) by introducing three input parameters: soil clay content, soil exchangeable K content and time since contamination.

One of the advantages of the existing models is that they successfully used more readily available soil parameters to predict the TF by linking them with the K^+ concentration in soil solution (m_K) and the radiocaesium interception potential (RIP). Absalom et al. (2001) broadened the applicability of the existing models by introducing additional soil input parameters such as soil organic matter, soil pH and ammonium (NH_4^+) concentration in soil solution, to describe RCs transfer observed on highly organic soils (Sanchez et al., 1999). Tarsitano et al. (2011) analysed statistically redundant and noise parameters in Absalom et al. (2001), then removed soil pH from input parameters and re-parametrised the model using a larger dataset.

The relationships between soil input parameters and RIP and m_K have been calibrated for soils of mainly northern Europe (Sanchez et al., 1999; Smolders et al., 1997). Our previous study (Uematsu et al., 2015) concluded that the relationship between soil clay content and its affinity to RCs, expressed as the RIP, derived for soils of northern Europe unlikely holds for Japanese soils. The particular clay mineralogy of the Japanese soils, often volcanic soils, has a lower affinity for RCs (Takeda et al., 2014). Hence, this calls for an analysis of the TF with local soil properties to relate bioavailability of RCs with site specific conditions.

The goal of this study was to evaluate the existing models calibrated to European soils, to predict the TF for Japanese soils. We characterised RIP (51 soils) and m_K (37 soils) for representative soils collected from the Fukushima accident affected area and compared the measured RIP and m_K values with those predicted from the existing models. The effect of the K^+ concentrations in soil solution on plant uptake of RCs is likely consistent between Europe and Japan when considering the same crop. Therefore, we compared (a) the predicted TF using measured values of RIP and m_K (region-specific parameters) and (b) the predicted TF using the RIP and m_K predicted from the soil clay content and exchangeable K with the models calibrated to European soils (generic parameters). Finally, the standard deviation of the predicted TFs was compared with that of the measured TFs for the Fukushima accident affected area (82 soils). It should be mentioned that the predicted TF and the field measured TF are not for identical soils.

The TFs were evaluated for grassland. Grass is widely cultivated for forage production in Japan. For grass, an activity concentration limit of 500 Bq kg^{-1} dry weight (the sum of ^{137}Cs and ^{134}Cs) in forage for cattle and horses was proposed in 2012 by the Japanese government. RCs in forage exceeding the limit has been observed in 2012 (Harada, 2014). Harada (2013, 2014) and the Ministry of Agriculture, Forestry and Fisheries (2013) reported the relationship between the TF to grass and exchangeable K content in soils. Based on this relationship, they proposed to maintain the soil exchangeable K content above 6.4 $mmol_c\ kg^{-1}$, in order to keep the TF for grass lower than approximately 5 kg kg^{-1}.

2. Semi-mechanistic models

2.1. Model descriptions

Accepting plant root cells take up RCs from soil solution in adjacent rhizosphere soils, the soil-to-plant transport of RCs can be described by the simple combination of two fundamental processes (Absalom et al., 1999; Smolders et al., 1997);

$$\mathrm{RCs_{soil}} \overset{K_D}{\leftrightarrow} \mathrm{RCs_{soil\ solution}} \xrightarrow{\mathrm{CF}} \mathrm{RCs_{plant}} \tag{2}$$

where K_D (the solid–liquid distribution coefficient: L kg^{-1}) is the ratio of RCs concentration in the soil (Bq kg^{-1}) to that in the soil solution (Bq L^{-1}) and CF (the concentration factor: L kg^{-1}) the ratio of RCs concentration in the plant (Bq kg^{-1}) to that in the soil solution. In this way, the TF is then calculated as a simple quotient;

$$\mathrm{TF} = \mathrm{CF}/K_D \tag{3}$$

It is a key to distinguish the solid–liquid partitioning and the plant uptake processes of RCs and to predict both the K_D and the CF values in a semi-mechanistic way by taking into account dominant factors affecting each process.

We adopted three existing models (Absalom et al., 1999, 2001; Tarsitano et al., 2011) for predicting the TF of RCs for Japanese soils. The variables in the existing models are defined in Table 1. A series of calculations and parameter values in these existing models are shown in Tables A.1 and A.2 in the supplementary data. The overview of semi-mechanistic processes of the TF prediction is summarised in the following sections.

2.1.1. Solid-liquid distribution of RCs (K_D)

The solid–liquid distribution has been described in terms of a labile distribution coefficient K_{Dl} (L kg^{-1}), which is defined as the ratio of labile sorbed RCs to RCs in soil solution. To illustrate a dynamic change of the solid–liquid distribution with time in soils, it is useful to assume that the K_{Dl} is a constant and to compute the K_D at time t since contamination as $K_D = K_{Dl}/D$, where D (<1) is the dynamic factor (see section 2.1.2).

All the three models considered the solid–liquid distribution of trace amounts of RCs on highly selective sorption sites in the soil clay fraction, called frayed edge sites (FES) (Sawhney, 1972), and the K^+ and NH_4^+ concentrations in soil solution, which can compete with RCs on FES by cation exchange reactions.

Absalom et al. (1999) calibrated the model for Belgian mineral soils (Smolders et al., 1997) where the sorption of RCs on nonselective sites on soil organic matter can be negligible and where the ammonium (NH_4^+) concentration in soil solution was low so that it can be ignored, using the following formula;

$$K_{Dl} = \frac{k_a \cdot \mathrm{RIP}}{(m_K)^{n1}} \tag{4}$$

where RIP (mol kg^{-1}) is the radiocaesium interception potential of soils (Cremers et al., 1988), m_K (M) is the K^+ concentration in soil solution and k_a and n_1 are fitted parameters.

Besides the distribution of RCs on FES in the soil clay (K_D^{clay}, L kg^{-1}), Absalom et al. (2001) and Tarsitano et al. (2011) included the distribution of RCs on the soil organic matter fraction to demonstrate the RCs behaviour in highly organic soils in which significant amounts of RCs may be sorbed on nonselective organic matter sites. Absalom et al. (2001) adopted a variable K_D^{humus} (L kg^{-1}; Eq. [B.5] in Table A.1) as the K_D of RCs on soil organic matter sites, whereas Tarsitano et al. (2011) used a fitted constant K_{Dmin} (L kg^{-1}; Table A.2) as the minimum K_{Dl} value for a soil where the RCs sorption on clay surfaces is negligible (i.e. K_D^{clay} is small). Their models were calibrated for highly organic soils (Sanchez et al., 1999; mean organic matter 65%) as well as for the mineral soils (Smolders et al., 1997);