Science of the Total Environment 524–525 (2015) 148–156

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Predicting radiocaesium sorption characteristics with soil chemical properties for Japanese soils

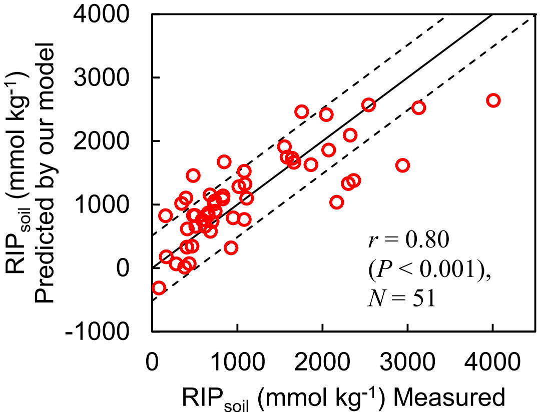
Shinichiro Uematsu [a,b,*], Erik Smolders [b], Lieve Sweeck [a], Jean Wannijn [a], May Van Hees [a], Hildegarde Vandenhove [a]

[a] Biosphere Impact Studies, SCK•CEN, Belgian Nuclear Research Centre, Boeretang 200, B-2400 Mol, Belgium
[b] Division of Soil and Water Management, Katholieke Universiteit Leuven, Kasteelpark Arenberg 20, B-3001 Leuven, Belgium

HIGHLIGHTS

- A model to predict the RIP from soil properties was developed for Japanese soils.
- The RIP decreased with increasing organic matter, a main factor affecting the RIP.
- Soil organic matter and CEC explained 64% of the variation in RIP.
- Andosols exhibited significantly lower RIP than other Japanese soils.
- This model can be applied for soil types typical in the Fukushima affected area.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 28 November 2014
Received in revised form 7 April 2015
Accepted 7 April 2015
Available online 16 April 2015

Editor: Mae Sexauer Gustin

Keywords:
^{137}Cs sorption in soil
Radiocaesium interception potential (RIP)
Fukushima contaminated area
Andosols
Soil organic matter
Soil clay content

ABSTRACT

The high variability of the soil-to-plant transfer factor of radiocaesium (RCs) compels a detailed analysis of the radiocaesium interception potential (RIP) of soil, which is one of the specific factors ruling the RCs transfer. The range of the RIP values for agricultural soils in the Fukushima accident affected area has not yet been fully surveyed. Here, the RIP and other major soil chemical properties were characterised for 51 representative topsoils collected in the vicinity of the Fukushima contaminated area. The RIP ranged a factor of 50 among the soils and RIP values were lower for Andosols compared to other soils, suggesting a role of soil mineralogy. Correlation analysis revealed that the RIP was most strongly and negatively correlated to soil organic matter content and oxalate extractable aluminium. The RIP correlated weakly but positively to soil clay content. The slope of the correlation between RIP and clay content showed that the RIP per unit clay was only 4.8 mmol g^{-1} clay, about threefold lower than that for clays of European soils, suggesting more amorphous minerals and less micaceous minerals in the clay fraction of Japanese soils. The negative correlation between RIP and soil organic matter may indicate that organic matter can mask highly selective sorption sites to RCs. Multiple regression analysis with soil organic matter and cation exchange capacity explained the soil RIP ($R^2 = 0.64$), allowing us to map soil RIP based on existing soil map information.

---

[*] Corresponding author at: Biosphere Impact Studies, SCK•CEN, Belgian Nuclear Research Centre, Boeretang 200, B-2400 Mol, Belgium.
E-mail addresses: suematsu@sckcen.be, shin-uematsu@frontier.hokudai.ac.jp (S. Uematsu), erik.smolders@ees.kuleuven.be (E. Smolders), lsweeck@sckcen.be (L. Sweeck), jwannijg@sckcen.be (J. Wannijn), mvhees@sckcen.be (M. Van Hees), hvandenh@sckcen.be (H. Vandenhove).

http://dx.doi.org/10.1016/j.scitotenv.2015.04.028
0048-9697/

1. Introduction

The uptake of radiocaesium (both ^{137}Cs and ^{134}Cs; hereafter RCs) in the food chain via soil-to-plant transfer is of great concern following the accidental releases from the Fukushima Daiichi Nuclear Power Plant, Japan, in March 2011. The total amount of ^{137}Cs released by the accident was estimated to be 1.2×10^{16} Bq (Chino et al., 2011). ^{137}Cs has a relatively long half-life of 30 years and thus, can lead to long-term risks of human radiation exposure in agricultural ecosystems. Although the area of agricultural fields contaminated at activity concentration higher than 5000 Bq kg^{-1} (the sum of both of the RCs) decreased from ~89 km^2 in November 2011 to ~75 km^2 in December 2012 (Ministry of Agriculture, Forestry and Fisheries, 2012, 2013), there is still interest in the RCs transfer mechanisms to plants in the contaminated area. The contaminated agricultural fields cover various soil types, mainly Andosols, Gleysols and Fluvisols. This land is cropped by paddy rice, wheat, barley, soybeans, vegetables, fruit trees and pasture.

In contaminated soils, the transfer of RCs to plants is often evaluated using the soil-to-plant transfer factor (TF) of RCs, which is defined by the ratio of concentration of RCs in plants to that in soils (IAEA, 2009). It is well known that TFs are highly variable depending on soil and plant types (IAEA, 2009). For the Fukushima affected area, Takeda et al. (2014) reported that TFs varied two orders of magnitude for soybean grain between 46 different fields in the Fukushima Prefecture. For agricultural production in the affected area, it is therefore of prime interest to identify the vulnerable soils, where the RCs concentration in the agricultural produce may exceed the regulatory limit for foodstuffs, even when soil contamination has become relatively low.

The general consensus is that the fate of trace amounts of RCs in soils is ruled by particularly highly selective sorption (Brouwer et al., 1983; Cremers et al., 1988). This selective sorption occurs at a wedge shape area between collapsed and weathered clay layers, called frayed edge sites (FES), of 2:1 phyllosilicates such as micaceous minerals (Sawhney, 1972). Illite is a partially weathered micaceous mineral and has highly selective sorption sites with the trace Cs-to-K selectivity coefficient (K_c(Cs–K)) of the order of 1000 (Cremers et al., 1988). Highly selective sorption, similar to illite, was also observed in montmorillonite by specific treatments, such as a repeated wetting drying cycle of homoionic K-montmorillonite and the reduction of layer charge (Maes et al., 1985). They attributed the selective sorption to the formation of the FES through a collapse of expanded interlayers of montmorillonite.

The sorption characteristics of RCs on FES can be quantified in terms of the radiocaesium interception potential (RIP), which is defined as the product of the FES capacity ([FES]) and the Cs-to-K selectivity coefficient (K_c^{FES}(Cs–K)) on FES (Cremers et al., 1988; Wauters et al., 1996a). The RIP is a measurable soil parameter and experimental techniques are used to mask non-selective sorption sites (regular exchange sites) to restrict RCs sorption only on FES.

In the previous studies, the TF of ^{137}Cs was directly correlated with the RIP of soil for some regional soil groups (Delvaux et al., 2000; Takeda et al., 2014). The data of Delvaux et al. (2000) confirmed a negative correlation between log (TF) and log (RIP) ($r^2 = 0.77$) obtained from laboratory transfer experiments with a number of European soils in K deficient condition. A similar relationship was recently applied for the Fukushima ^{137}Cs deposition. Takeda et al. (2014) reported that the logarithm of TF for soybeans observed for 46 different sites in the Fukushima affected area in 2011 was negatively correlated to the logarithm of soil RIP ($r = -0.52$, $P < 0.001$). They also showed a negative correlation between log (TF) and the logarithm of exchangeable K in soils ($r = -0.54$, $P < 0.001$; Takeda et al., 2014).

Indeed, several studies revealed that other factors than RIP also affect the TF of RCs. First, potassium (K^+), ammonium (NH_4^+) and sodium (Na^+) concentration in soil solution can compete with RCs on FES by ion exchange. In addition, lower K^+ concentration in soil solution strongly increases the root uptake of RCs by plants due to the competition effect on plant uptake. It was demonstrated that the TF is a function of the RIP and soil solution K concentration (e.g. Sanchez et al., 1999; Smolders et al., 1997; Vandenhove et al., 2003).

Semi-mechanistic models to predict the soil-to-plant transfer factor (TF) for RCs were developed (Absalom et al., 1999, 2001). In these models, the clay content and exchangeable K are among the independent soil parameters to predict the TF via their correlations with RIP and soil solution K concentration. One of the biggest advantages of the Absalom models is that the TF can be predicted for different types of soils which have different soil chemical properties (i.e. different K fertilisation condition and clay content). These types of models may be applied for the prediction of TF at a large-scale in the Fukushima affected area. However, above-mentioned Absalom models were developed based on European soil data and might need to be recalibrated for typical soils in the Fukushima area because it is still unknown if the affinity of the soil minerals in this area for RCs is different from that in European soils.

It can be expected that the affinity of clay minerals to RCs for Japanese soils is, on average, lower than that in European soils. Andosols are common in Japan. Vandebroek et al. (2012) compared the soil RIP among soils in a worldwide scale soil collection and revealed that the RIP of Andosols (ranged from 94 to 1630 mmol kg^{-1}) was one of the lowest among all soil groups (from 1.8 to 13300 mmol kg^{-1}). In previous studies on Fukushima soils (Nakao et al., 2014; Takeda et al., 2014), only small numbers of Japanese Andosols were included for investigating the relationship between RIP and soil chemical properties, and RCs sorption characteristics of Japanese Andosols have never been compared with those of other Japanese soils.

Andosols are generally classified by the high amount of reactive Al. The clay mineralogy is quite different between allophanic Andosols and non-allophanic Andosols (Saigusa and Matsuyama, 1998). The major clay types in allophanic Andosols are amorphous minerals, whereas 2:1 phyllosilicate clays and Al- and Fe-humus complexes are dominated in non-allophanic Andosols (Matus et al., 2014). The selectivity of RCs on amorphous minerals is likely lower than that on FES in 2:1 phyllosilicates, because they lack layered structures like FES. Some studies, however, observed that 2:1 phyllosilicates coexisted in allophanic Andosols (e.g. Masui et al., 1966). Inoue and Naruse (1987) determined the mineralogy of eolian dust deposition in Japan, transported from loess in China, and illite was one of the dominant minerals. This may be a possible source of selective sorption sites of RCs in Andosols. The dust deposition rate likely depends on the precipitation rate, as suggested by Inoue and Mizota (1988). Joussein et al. (2004) proposed that halloysite–smectite mixed layered clays contributed to the formation of RCs selective sorption sites in Andosols under K fertilisation and wetting–drying cycles in field condition. Halloysite is a common clay in Andosols as halloysite can be formed as a weathering product of allophane. However, Saigusa et al. (1978) did not observe detectable amounts of halloysite in the surface layer of Andosols in Japan.

For paddy soils in the Fukushima Prefecture, micaceous minerals are seldom the dominating clay fraction, rather several types of clay minerals coexisted or smectite was predominant (Nakao et al., 2014; Sano et al., 2010). Sano et al. (2010) reported that smectite was a common clay in the western part of the Fukushima Prefecture. Smectite in this region was developed from sedimentary rocks including tertiary green tuff distributed as a mother rock in the Ou Mountains (Sano et al., 2010). Micaceous minerals dominated in the eastern part where soils are developed from granite in the mother rock of the Abukuma Mountains.

In the Tochigi Prefecture, most paddy soils are categorised as Andosols (wet characteristic) and Gleysols, while soils for other crops and grasslands are categorised as Andosols, Fluvisols and Cambisols. Gleysols and Fluvisols in the Tochigi Prefecture were developed from clastic sedimentary rocks (Tochigi Prefecture, 2006). Most paddy soils in the Ibaraki Prefecture are Gleysols and Andosols (Ibaraki Prefecture, 2008).