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## Behaviour and Phytoavailability of Radiocaesium in Surface Soil

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### *Abstract*

Large amounts of several radionuclides were released into the atmosphere from the Fukushima Daiichi Nuclear Power Plant (F-1NPP) in March 2011 and the ensuing months, and deposited onto the surface soil and vegetation in wide areas of eastern Japan. Among the released radionuclides,  $^{137}\text{Cs}$  (half-life, 30.1 y) is the most important nuclide for assessment of the long-term radiation exposure for the public. The distribution of fallout  $^{137}\text{Cs}$ , derived from past atmospheric nuclear weapons tests, in agricultural fields in Japan was well investigated and these studies are briefly reviewed here. The long-term monitoring data of  $^{137}\text{Cs}$  in soils and crops can provide useful information to predict the behavior of radiocaesium released from the F-1NPP. Since Cs is irreversibly sorbed on frayed-edge sites (FESs) of illitic minerals, its mobility is quite low after its deposition onto surface soil. Behaviour and phytoavailability of Cs in soil is controlled by several factors including clay mineralogy, elapsed time since deposition and ion components in soil solution, especially  $\text{NH}_4^+$  and  $\text{K}^+$ . Then, this paper also reviews studies on the behavior of Cs in soil-plant systems.

### *Introduction*

Damage to reactor buildings of the Fukushima Daiichi Nuclear Power Plant (F-1NPP) due to the Great Eastern Japan Earthquake and the massive tsunami on March 11, 2011, coupled with the loss of electrical power that prevented cooling of the reactors and spent fuel pools and led to hydrogen explosions, resulted in the release of large amounts of radionuclides from the plant. Noble gases and volatile radionuclides predominated in releases to the terrestrial environ-

ment (Tagami et al., 2011) and they were deposited in wide areas of the Tohoku and Kanto districts. The amounts of released  $^{131}\text{I}$  and  $^{137}\text{Cs}$  have been estimated as  $1.5 \times 10^{17}$  Bq and  $1.3 \times 10^{16}$  Bq, respectively (Chino et al., 2011). Approximately 13 % of  $^{131}\text{I}$  and 22% of  $^{137}\text{Cs}$  of the total amounts released were estimated to be deposited over Japanese land areas (Morino et al., 2011). The degree of soil pollution has been gradually clarified by air borne monitoring and soil surveys. Iodine-131 has a short physical half-life (8 d), and it decayed rapidly after the accidental release. From a long-term viewpoint,  $^{134}\text{Cs}$  (half-life, 2.1 y) and  $^{137}\text{Cs}$  (half-life, 30.1 y) are important nuclides to assess radiation exposure of the public.

Shiozawa et al. (2011) reported the vertical distribution of radiocaesium ( $^{134}\text{Cs}$  and  $^{137}\text{Cs}$ ) in a paddy field in Fukushima Prefecture after the F-1NPP accident. They found 96% of the total radiocaesium was in a surface soil layer of 0-5 cm depth, and the radiocaesium was mixed within a depth of 0-15 cm after plowing. It is known that Cs is strongly retained by soil clay minerals. Therefore, the radiocaesium will remain in the surface soil for a long time.

The distribution of global fallout  $^{137}\text{Cs}$ , derived from the atmospheric nuclear weapons testing, in agricultural soils and crops has been well investigated in Japan. Since the Chernobyl Nuclear Power Plant accident in 1986, the fate of  $^{137}\text{Cs}$  in the terrestrial environment and the mechanism of its retention by clay minerals have been extensively studied, mainly in European countries. In this paper, scientific knowledge about the behaviour of Cs in soil-plant systems is reviewed.

### ***Fate of global fallout $^{137}\text{Cs}$ in soils in Japan***

Huge amounts of  $^{137}\text{Cs}$  and some other radionuclides (e.g.  $^{90}\text{Sr}$  and  $^{239+240}\text{Pu}$ ) were released from atmospheric nuclear weapons tests in the late 1950s and early 1960s, leading to global fallout, mainly in the northern hemisphere (Aoyama et al., 2006). The deposition of fallout  $^{137}\text{Cs}$  has been monitored since the 1950s in Japan. The maximum annual deposition was observed in 1963, after the large-scale atmospheric nuclear weapons tests of 1961–1962 (Hirose et al., 2008).

It is known that Cs is strongly retained on surface soil after deposition, and downward migration is very slow in soil (Rosén et al., 1999). Most of the fallout  $^{137}\text{Cs}$  is within a depth of 30 cm from the surface several decades after the main deposition in undisturbed forest in Japan (Fukuyama and Takenaka, 2004). In agricultural fields, on the other hand, fallout  $^{137}\text{Cs}$  is homogeneously mixed within the surface soil layer because soils in agricultural fields are generally mixed by plowing every year prior to planting of crops (Tsukada and Nakamura, 1999; Tsukada et al., 2002).

Long-term monitoring of radioactivity in 17 agricultural fields was carried out from 1959 by the National Institute for Agro-Environmental Sciences (Komamura et al., 2005). Maximum concentrations of  $^{137}\text{Cs}$  in the plowed layer of paddy and upland field soils were observed in 1963 - 1966, which was an effect of the maximum annual fallout in 1963. Komamura et al. (2006) reported that the concentration of  $^{137}\text{Cs}$  in the plowed layer was decreased by the half-times of 9 – 24 y in paddy fields and 8 – 26 y in upland fields. These monitoring data provide useful information to predict long-term behaviour of radiocaesium derived from the F-1NPP accident.

### ***Retention of Cs by soil clay***

It is known that radiocaesium is strongly sorbed by illitic minerals. The weathering fronts in the interlayers of illitic minerals expand into a wedge shape, known as frayed-edge sites (FESs) (Cremers et al., 1988). The contribution of FESs to the overall cation exchange capacity in soil is quite small, e.g. 0.001 – 6 % (Delvaux et al., 2000); however, they selectively sorb poorly hydrated cations including  $\text{K}^+$ ,  $\text{NH}_4^+$  and  $\text{Cs}^+$ . Radiocaesium deposited on surface soil is specifically sorbed by FESs, and the number of FESs affects Cs mobility in the soil.

The radiocaesium interception potential (RIP) was established as a quantitative indicator of FESs (Cremers et al., 1988). The RIP value is defined as the mathematical product of the trace Cs to K selectivity coefficient in the FESs and the FES capacity (Wauters et al. 1996). Vandebroek et al. (2009) compared the RIP among various soils collected worldwide and found that Andosols had one of the lowest RIP values among the soil groups. Nakamaru et al. (2007) also reported that the soil-soil solution distribution coefficient (Kd) of  $^{137}\text{Cs}$  tracer in Andosols is relatively lower than that in other Japanese agricultural soil groups, and they suggested that soil organic matter could increase the mobility of Cs in soils. Ishikawa et al. (2007) investigated the relation between relative illite content and the strongly fixed proportion of  $^{137}\text{Cs}$  tracer in Japanese paddy soils from which they suggested that the mobility of Cs was relatively higher in soils with lower content of illitic minerals.

Ammonium ion is more competitive than K for Cs adsorption on FESs. Therefore, a high concentration of  $\text{NH}_4^+$  in soil solution can inhibit the Cs retention and increase Cs mobility in soil (Comans et al., 1989; Takeda et al. 2008). Ammonium salt solution can extract a relatively high amount of Cs from soil. For example, the mean extraction yield of stable Cs in 16 agricultural soils was 4.1 % with 1 M  $\text{NH}_4\text{NO}_3$  solution, whereas it was 1.5 % with 1 M  $\text{HNO}_3$  solution under the same extraction conditions (Takeda et al., 2006).

Mobility of radiocaesium decreases with time after its deposition in the soil due to an aging effect (Sanzharova et al., 1994; Absalom et al., 1995; Fesenko et al., 1997; Rigol et al., 1999; Roig et al., 2007; Takeda et al., 2009). A collapsing process of the interlayer space of phyllosilicates induces Cs fixation onto soil (Hird et al., 1996).

### ***Soil-to-plant transfer of Cs***

Concentration of radiocaesium in a crop plant (edible part) is estimated by using the soil-to-plant transfer factor (TF). The TF is generally defined as the concentration ratio of Cs in the edible part of crop plant ( $\text{Bq kg}^{-1}$  dry or wet plant) to that in the soil ( $\text{Bq kg}^{-1}$  dry soil), which is obtained by analysis of samples collected from agricultural fields or grown in pot experiments with radioisotope tracer. The TFs of many crops have been summarized by the IAEA (International Atomic Energy Agency, 2010). The TFs

for crops in Japan were reported by measuring the fallout  $^{137}\text{Cs}$  derived from atmospheric nuclear tests (Tsukada and Nakamura, 1999; Tsukada and Hasegawa, 2002; Tsukada et al. 2002; Komamura et al., 2005; Uchida et al., 2007; Uchida and Tagami, 2007; Kamei-Ishikawa et al., 2008). The reported TFs cover wide ranges with about two orders of magnitude in the same crop plant, because the availability of Cs in soil depends on many factors, including soil type, elapsed time after deposition, and coexisting ions.

Delvaux et al. (2000) conducted a cultivation experiment of ryegrass using a wide variety of soils containing  $^{137}\text{Cs}$  tracer. The soil-to-grass transfer of Cs was negatively correlated with RIP values in soil, and larger TF values were observed in lower RIP soils. This result suggested that the number of FESs largely controls the availability of Cs in soil.

Soil-to-plant transfer of Cs generally decreased with time after deposition to the soil due to the aging effect as mentioned above. In a laboratory experiment for an allophanic Andosol, phytoavailability of Cs was determined to decline exponentially up to about 100 d after its addition to the soil, and was almost constant thereafter (Takeda et al., 2009).

The uptake of Cs by plant roots from soil solution is enhanced under the low K condition in soil solution (Shaw 1993; Smolders et al., 1997). Therefore application of K fertilizer reduces Cs uptake effectively in soils with a low amount of available K (Lembrechts, 1993; Nisbet et al., 1993; International Atomic Energy Agency, 1994)

### ***Behaviour of Cs in paddy soil***

Rice is the most important crop for estimation of internal exposure to the public in Japan. As noted above, long-term monitoring of fallout  $^{137}\text{Cs}$  derived from nuclear weapons tests has been carried out for paddy soil and rice grain samples, providing useful information for prediction of radiocaesium concentration in rice. Although direct absorption of fallout  $^{137}\text{Cs}$  from the atmosphere is the major pathway in rice before the 1980s, it became negligible after 1990 (Komamura et al., 2006). The concentration of  $^{137}\text{Cs}$  from root uptake of rice plants varies by one order of magnitude among components (polished rice, rice bran, hull, straw and root). The highest and the lowest concentrations have been found in rice bran and in polished rice, respectively (Tsukada et al., 2002). Therefore, the concentration of  $^{137}\text{Cs}$  in polished rice

is lower than in brown rice (Komamura et al., 2006; Uchida et al., 2009). As well, 10% of the total  $^{137}\text{Cs}$  in an entire rice plant was found in polished rice, and the remaining 90% was in non-edible parts. Only 0.003 % of  $^{137}\text{Cs}$  in the plowed soil was transferred to the above ground biomass of rice plants (Tsukada et al. 2008).

Although behaviour of radiocaesium in contaminated agricultural fields in European countries has been well documented, particularly since the Chernobyl accident in 1986, the behaviour in paddy fields has not been studied much. Tensho et al. (1959) carried out a pot cultivation experiment using paddy and upland soils containing radioactive Cs tracer. Their results showed that about 20 times larger amount of Cs was absorbed by paddy rice than upland rice. Under the anoxic condition, the major form of N in paddy soils is  $\text{NH}_4^+$ , which can increase Cs mobility in soil. Higher availability of Cs in paddy soils would relate to higher concentrations of  $\text{NH}_4^+$  in soil solution (Tensho et al. 1961). These studies indicate that availability of Cs is affected by the nitrogen cycle in paddy soil. In addition, Cs in irrigation water would affect its content in rice plant. Uptake of Cs by rice plant is much higher from solution than from soil when contaminated irrigation water is supplied to the paddy soil (Myttenaere et al., 1969).

Previously reported TFs of fallout  $^{137}\text{Cs}$  for Japanese crops showed that the TFs of rice grain are relatively smaller than other crops including leafy and root vegetables (Tsukada and Nakamura, 1999; Tsukada et al., 2002; Tsukada and Hasegawa, 2002; Komamura et al., 2005; Uchida et al., 2007; Uchida and Tagami, 2007; Kamei-Ishikawa et al., 2008). However, the behaviour of Cs in paddy fields is complicated and knowledge is still insufficient for prediction of Cs transfer to rice plant and for setting countermeasures to reduce Cs uptake. Analysis of risk factors enhancing Cs uptake by rice is important for contaminated regions.

### ***Conclusions***

Behaviour and phytoavailability of radiocaesium in surface soil are affected by many factors including clay mineralogy, elapsed time since deposition, and major ions present in the soil solution. Radiocaesium is specifically and irreversibly sorbed by the FESs of illitic minerals. Therefore, its mobility as dissolved ions is quite low after deposition onto surface soil.

Phytoavailability of Cs partly depends on soil types. Caesium would be more labile in soils with a low number of FESs. Elapsed time after deposition is also an important factor controlling Cs partitioning between the soil solid and solution phase. Concentrations of major ions, especially  $K^+$  and  $NH_4^+$  in soil solution greatly affect Cs behaviour in soil. Since increasing the concentration of  $NH_4^+$  in soil solution generally enhances Cs mobility in soil, the concentration of Cs in paddy soil can be elevated under the anoxic condition. Potassium depletion increases Cs root uptake from soil solution, therefore, fertilizer use should be checked carefully for management of contaminated agricultural soil.

Knowledge of the behaviour of radiocaesium in paddy fields and volcanic ash soils is still not sufficient. In addition, geomorphological effects on Cs behaviour in mountainous and catchment areas, such as northeastern Japan, are more complicated than in past contaminated areas such as around the Chernobyl nuclear power plant. Further investigations are needed for better management of contaminated soils for a long-term.

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