Correlation between Different Type of Caesium Carrier in
The Radiocaesium Interception Potential Measurement for
Forest Soils

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Abstract. Radiocaesium, especially 137Cs, is one of major heavy fission product radionuclide
that has a relatively long half-life and acts as a dangerous persistent pollutant to the
environment. It could be dispersed widely through the atmosphere during nuclear weapon test
and reactor accident such as Fukushima Dai-Ichi Nuclear Power Plant (FDNPP) disaster in
Japan. High retention of radiocaesium in the surface of forest floor surrounding Fukushima
area is important issues to measure further potential risk of health and environment
contamination. Radiocaesium Interception Potential (RIP) values are related to the sorption
capacity of radioactive Cs in clay minerals, and in this study, we investigated the correlation
between different type of Cs carrier to the RIP and their adsorption character in soil. The RIP
values of all soil sample were varied from 27.27 to 522.66 mmol kg⁻¹. The carrier effect on
RIP was conducted using carrier 134Cs, carrier-free that contains 134Cs, 136Cs and 137Cs and
133Cs stable isotopes. Correlation between different type of carrier suggests a relatively lower
RIP for stable caesium as a carrier. This phenomena could be caused by competitive adsorption
among of caesium ions in the frayed edge sites of soil mineral. Organic carbon content and
CEC also contributed to the RIP value. No significant difference found of C/N ratio and CEC
values between deciduous and coniferous forest soil as they also have similarity of mineral
content in soil.

Keywords: Radiocaesium, RIP, carrier effect, adsorption, frayed edge sites.

1. Introduction

Result of radioactivity measurements in surrounding Fukushima Daiichi Nuclear Power Plant
(FDNPP) disaster revealed that radionuclides spread dominantly affected in the central and eastern
part of Honshu-Island due to movement of radioactive cloud by northeast wind and following rainfall
during March 21-23 [1]. High contamination area spread from the reactor site to about 60 km
northwest region in the Fukushima Prefecture. Among radionuclides that have been discharged to the
environment, 137Cs merits consideration since it has a unique combination of physical properties and
historical notoriety. It is readily produced in vast amount during fission from nuclear weapon-testing
and reactor accident, has an intermediate half-life (longer than the half-life of 134Cs and 131I), high-
energy decay pathway, and is chemically reactive and highly soluble. These properties made 137Cs a
major contribution to the total radiation released during nuclear accidents so that become a dangerous
legacy of major nuclear accidents.

The total released amounts of 137Cs have been estimated as 1.3 x 10¹⁶ Bq [2], with approximately
22% deposited over Japanese land areas [3]. About 66% (428 km²) of the heavily contaminated area
with Fukushima-derived radiocaesium (134, 137Cs ≥ 1 MBq m⁻²) is covered by forests [4]. Litter fall
from the canopy layer is the main radiocaesium transport pathway to the ground surface and around
85% of the 137Cs inventories exhibited on the forest floor [5,6]. Radiocaesium was initially deposited
and absorbed to canopy layers in coniferous forests due to the presence of leaves during the major fall
during early spring [4,7]. In contrast, the canopy interception effect in deciduous forests was less important because at the time of accident were leafless [8]. Result from lysimeter study shows that radioactive $^{137}$Cs was rapidly leached from the litter layer of forest soil, but then immobilized in the topsoil layer (mineral soil layer) [9]. It is well known that radiocaesium is strongly retained in clay soil and hardly migrated downward with time after its deposition onto soil [10–12]. The activity of adsorbed Cs in soil was highest in the clay fraction ($<$2 μm) and tended to decrease as the particle size is increases [13]. High retention of radiocaesium in the clay minerals associate with the frayed-edge sites (FES) which is located at the layer edges of micaceous particles [14].

Micaceous clay minerals such as illite, vermiculite and smectite are 2:1 phyllosilicates having 1.0 nm basal spacing, if negatively charged, can form strong inner-sphere complexes with monovalent cations with low energy hydrations such K$^+$, Rb$^+$ and Cs$^+$ [15,16]. Weathered micaceous minerals forms the wedge zone between non-expanded (1.0 nm) and expanded layers (1.4 nm), which is referred as FES. The release of K$^+$ ions leads this sites to negatively charged and the size is similar to the ionic radius of dehydrated K$^+$ or Cs$^+$, therefore have a great affinity for Cs$^+$ [14,17]. The role of the FES in the specific retention of radiocaesium can be quantified in terms of the radiocaesium interception potential (RIP), defined as the product of the trace Cs$^+$ to K$^+$ selectivity coefficient in the FES and the FES capacity [18–22]:

$$RIP = [FES] \cdot K^FES(Cs^+/K^+).$$

where $[FES]$ is the sorption capacity for radiocaesium by the frayed edge sites (FES) of micaceous clay minerals and $K^FES(Cs^+/K^+)$ the selectivity coefficient of the ion exchange Cs$^+/K^+$. Thus, the RIP is usable as an index of the mobility of the radiocaesium in soil, and known as intrinsic soil parameter. Mapping of regions with RIP values could also useful to determine regions that are potentially enhanced the risk of radiocaesium contamination from nuclear accident or atmospheric fallout.

The RIP value obtained from many research report are mostly using radioactive caesium $^{137}$Cs as a carrier. We know that caesium is the element with most isotopes form, 40 known isotopes with the atomic masses range from 112 to 151 but only $^{133}$Cs is in stable form. In this paper, we investigated correlation of carrier effect on the RIP values including stable Cs. Carrier $^{134}$Cs and carrier free that contains $^{134}$Cs, $^{136}$Cs and $^{137}$Cs were used as subject of study. Soil sample were taken from different type of forest soil, focused on coniferous forest soil and deciduous forest soil, and different pedogenetic conditions (i.e. Japan and Indonesia).

2. Materials and Method

2.1. Site and Soil description

The soil samples used in the experiment were collected from the top-soil (0-10 cm) of four areas in Takizawa Research Forest, Iwate Prefecture, Japan (39°46 N, 141°9 E) and one soil sample for comparison is from Serpong site, Banten Province, Java Island, Indonesia (6°21 S, 106°39 E). Four areas of Iwate soil were two coniferous forest floor and two deciduous forest floor. In coniferous forest floor, the point sampling location were chosen based on their major vegetation type, such as the evergreen Sugi trees (Cryptomeria japonica) and Karamatsu trees (Larix kaempferi). The major vegetation type in around deciduous forest is Japanese Oak trees such as Konara trees (Quercus serrata). This site has an altitude of 250 – 260 m, mean annual temperature of 9.2°C and annual precipitation is about of 1300 – 1800 mm. The soil type is Andosol according to Food Agriculture Organization (FAO). The other site was located in botanical garden of Serpong Science and Technology Research Center (PUSPIPTEK Serpong), Tangerang Selatan, which was planted with various type of plants typical from all region in Indonesia including rainforest’s vegetation. This area has an altitude of 50 m, mean annual temperature of 28°C and annual precipitation is about of 142.60
mm. The surface soil is contain humus, roots of the grass and small latteric clay and classified as latteric clay group [23].

The collected samples were dried at 50°C for 5-6 days and passed through a 2 mm sieve. All of these sample were analysed for major parameters, such as pH, CEC, and total C content. In brief pH water and pH CaCl₂ were measured in suspension using soil:water and soil:CaCl₂ 1 M ratios of 1:2.5. The basic physicochemical properties of soil samples were analysed as shown in Table 1. Cation exchange capacity (CEC) was performed by shaking extraction method using ammonium acetate (pH 7) and were analysed using CKC AT-2000 instrument [24]. Total hydrogen, carbon and nitrogen analysis were performed using CHN analyzer JMA10 Auto sampler coupled with JM10 Micro-corder (J Science co. Ltd, Japan). Antipirine was used as a calibration chemical (total C content is 70 %). The C:N ratio of Iwate soils (sample C1, C2, D1, and D2) ranged from 12.8 – 13.7 and the C:N ratio for Indonesia soil (sample IDN) is 7.7. Water content was carried out by gravimetry after drying at a maximum temperature of 105°C for 4 h, and analysed weight of sample before (air-dried) and after drying at 105°C. To measure loss on ignition (LOI), soil samples were oven-dried for a period of 24 hours at a temperature of 105°C, then crushing and sieving at <2 mm. The soil fines were stored in desiccator at room temperature. To minimize sample variation in LOI measurements, about 20g of soil sample were used and an ignition temperature were set to 550°C for 3 hours to attain complete oxidation [25].

| Sample code | Vegetation type origin | Soil group | Soil pH<sub>1:2.5</sub> | Total H (%) | Total C (%) | Total N (%) | CEC (meq/100g) | Water Content at 105°C (%) | LOI550<sup>b</sup> (%) |
|-------------|------------------------|------------|-------------------------|-------------|-------------|-------------|-----------------|-----------------------------|-----------------|
| C1          | Coniferous             | Andosol    | 4.92                    | 4.79        | 2.72        | 14.18       | 1.07            | 54.68                        | 55.31           |
| C2          | Coniferous             | Andosol    | 5.01                    | 4.82        | 2.59        | 12.03       | 0.94            | 42.27                        | 49.94           |
| D1          | Deciduous              | Andosol    | 5.17                    | 5.30        | 2.80        | 17.56       | 1.29            | 57.05                        | 54.08           |
| D2          | Deciduous              | Andosol    | 4.71                    | 4.55        | 2.72        | 12.03       | 0.92            | 50.18                        | 50.11           |
| IDN         | Tropical               | Lateric clay | 5.76                   | 5.17        | 0.84        | 1.91        | 0.25            | 13.45                        | 17.79           |

<sup>a</sup> Soil pH: soil pH measured at a soil-to-water and soil-to-CaCl₂ ratio of 1:2.5

<sup>b</sup> LOI<sub>550</sub>: loss on ignition at 550°C

### 2.2. Chemicals

A carrier-free tracer that include of $^{134}$Cs, $^{136}$Cs and $^{137}$Cs, $^{134}$Cs carrier and $^{133}$Cs stable in CsCl standard solution (Wako Pure Chemical Industries, Ltd). All the other chemicals used in the experiments were analytical grade and were not subjected to any purification or pre-treatment.

### 2.3. X-ray Diffraction analysis

The composition of clay minerals in the soil was estimated using X-ray diffractometer (Rigaku, Japan) with CuKα radiation at 40 kV and 20 mA. Each scan was taken from 5° to 60°, with step interval of 0.02° at a rate of 5°/min. Soil samples were oriented using Milli-Q water, and after measuring oriented samples, subsequently heated for 2 hours at 550°C to remove organic matter in soil. The XRD patterns were then measured again under the same measurement conditions.

### 2.4. Radiocaesium Interception Potential (RIP)

To evaluate the RIP, soil samples were air-dried at 50°C for 6 days and sieved through <2mm sieve. Each 1.0 g of soil samples was put into a dialysis bag (Visking tube dialysis membrane, size 19.1 mm, As One) along with 5 mL of 100 mM CaCl₂ and 0.5 mM KCl solution. The dialysis bag then transferred to 250 mL plastic bottle containing 200 mL of 100 mM CaCl₂ and 0.5 mM KCl solution as outer solution. Every bottle contained maximum three bags of dialysis bag, and then shaken for 2 h
twice a day during 5 days dialysis. The outer solution was renewed each time before shaking to maintain the desired condition. After first step of dialysis, the bag was put into a new 50 mL plastic bottle containing the outer solution and each bottle was added with same amount of Cs concentration (carrier, carrier-free and stable Caesium), about $2.5 \times 10^{-6}$ g of mass stable Cs concentration equivalent with the radiation concentration of radio-caesium (carrier and carrier-free). In this study, radioactive tracer that we used are 10 kBq of carrier $^{134}$Cs and 1072 Bq of carrier-free that contains $^{134}$Cs, $^{136}$Cs and $^{137}$Cs. In the second step of dialysis also each bottle was shaken continuously during 5 d. After diluted, the outer solution was filtered using a membrane filter with a pore size of 0.45μm (polycarbonate, ADVANTEC) and then analysed using HPGe detector (ORTEC GMX-30190) for radioactive tracer and ICP MS for stable Cs. The RIP value can be obtained from the product of $K_D^R$ and the concentration of K$^+$ in the aqueous phase.

3. Result and Discussion

3.1. Clay mineralogy

The XRD pattern of soil sample presented in Figure 1. It shows that only Indonesia soil (IDN) that have clear peak in the 0.72 nm even though there are no peak observed in the 1.4 nm and 1.0 nm. No clear peaks were observed for illitic clay minerals in Iwate soils (C1, C2, D1 and D2). This result was in agreement with the result of XRD analysis by Takeda et al. (2013) for andosol soil, same type of group soil [26]. Analyses using XRD database of clay minerals revealed that mineral components in soils were quartz, anorthite, and albite as shown in Figure 2.

![Figure 1](image.png)

**Figure 1.** Representative X-ray diffraction spectra patterns of soil profile drying at 50°C.

After heat treatment of 550°C, clearly that no illitic/vermiculite clays were still exist in each type of soils. It means that heat treatment could break the structure of illitic/vermiculite layers of clay along
with organic matter removal. Mineral content in each soil sample are quite similar for coniferous and deciduous soil from Iwate soil as shown in similar XRD pattern both in peak and intensity spectra.

Figure 2. The XRD spectra pattern of soil sample after heat treatment at 550°C for 2 h. 
Vm = Vermicullite, It = Illite, M = Mica, K = Kaolinite, Ch = Chlorite, Q = Quartz, An = Anorthite, Al = Albite, Pl = Plagioclase.

3.2. Relation RIP and Kd
The RIP is defined as the product of the FES capacity and the selectivity coefficient of the ion exchange Cs⁺/K⁺ within the FES. To determine quantitatively the RIP, we calculated the solid/liquid distribution coefficient for ion caesium in an equilibration solution with specific K⁺ and Ca²⁺ ionic strength (KCa) and the K concentration in the equilibration solution (CK). With the assumption of the adsorption reaction follows reversible linear adsorption model, then calculation of Kd value could be expressed as:

\[
\frac{C_\text{a}}{m_{\text{soil}}} - \frac{V C_b}{m_{\text{evol}}} = K_d C_a \#(2)
\]

where:  
Kd: Distribution coefficient (L/g)  
m_{\text{soil}}: Weight of soil (g)  
V: Volume of equilibration solution (L)  
C_a: Concentration of Cs originally adsorbed in soil (ppb)  
C_b: Concentration of Cs in solution after blank test (ppb)
where: \( C_c \): Concentration of Cs in stable Cs added solution before shaking (ppb)
\( C_e \): Concentration of Cs in stable Cs added solution after equilibration (ppb)

From equation (2) and equation (3), \( K_d \) value could be obtained by arranging both equations into a new equation as follows:

\[
K_d = \frac{V(C_0 + C_y - C_e)}{m_{sol}(C_e - C_h)} \tag{4}
\]

RIP (mol L\(^{-1}\)) can be calculated from the following equation:

\[
RIP (mol/kg) = K_d (L/kg) \cdot C_k (mol/L) \tag{5}
\]

In this equation, \( K_d \) is the distribution coefficient of Cs in (L kg\(^{-1}\)) and \( C_k \) is the concentration of K in equilibration solution (mol L\(^{-1}\)).

\( K_d \) values of all soil sample from RIP measurement is presented in the Figure 3. From the bar graphs of \( K_d \) values, it shows that using carrier-free in the RIP measurement give higher \( K_d \) values with the range from around 400 to 900 L kg\(^{-1}\). Spiked with the stable caesium in RIP measurement could give \( K_d \) values relatively lower than the other carriers. This phenomena could be caused by competitive adsorption among of caesium ions in the FES of soil mineral. The selective sorption enhanced mobility of competing ions and impact the bioavailability of trace elements in soils [27]. Carrier-free that used in this study is contains three radio isotopes of caesium, which are have different radioactivity and half-life (\(^{136}\)Cs have shortest half-life, only 13.16 d). We assumes that the FES capacity is a fixed of sorption sites for all competing caesium ions and K\(^+\) ions. This multicomponent competition could be explained by Langmuir type model. However, further research is necessary to fill this uncertainty.

![Figure 3. Comparison \( K_d \) values of each soil samples and their relations on different type of caesium carriers. \( K_d \) value of IDN soil using carrier \(^{134}\)Cs was not measured.](image)

The RIP values of Iwate soils (andosol) were varied from 69.22 to 522.66 mmol kg\(^{-1}\), which was within the range reported for andosol soils collected worldwide (94 – 1320 mmol kg\(^{-1}\)) by Vandebroek et al. (2012) and the RIP values of Indonesia soils also varied from 27.27 to 293.37 mmol kg\(^{-1}\) within the range based on land reported for Indonesia soil as shown in Table 2 [28]. From the equation (5),
we could expect that the RIP values also affected by different type of carrier. Relation between
different type of carrier to the RIP value of Iwate soils are presented in Figure 4. Radioactive caesium
as a carrier in RIP measurement tends to give higher RIP values.

![Figure 4. Scatter plot of correlation between RIP values from different type carrier of caesium
for Iwate soils. The dotted line is a correlation line and the dashed line shows 1:1 relationship.]

Table 2. The RIP values of soil sample

| No.  | Ref. Group | Land         | RIP (mmol kg⁻¹) | Type of Cs carrier | Type of forest/trees  |
|------|------------|--------------|-----------------|--------------------|-----------------------|
| 1    | Lateritic clay | Indonesia  | 27.27           | Stable Cs          | Tropical vegetation    |
| 2    | Lateritic clay | Indonesia  | 49.22           | Stable Cs          | Tropical vegetation    |
| 3    | Lateritic clay | Indonesia  | 53.36           | Stable Cs          | Tropical vegetation    |
| 4    | Andosol     | Japan        | 69.22           | Stable Cs          | Deciduous             |
| 5    | Andosol     | Japan        | 78.42           | Stable Cs          | Deciduous             |
| 6    | Andosol     | Japan        | 81.24           | Stable Cs          | Coniferous            |
| 7    | Andosol     | Japan        | 88.26           | Stable Cs          | Coniferous            |
| 8    | Andosol     | Japan        | 99.02           | Carrier             | Coniferous            |
| 9    | Andosol     | Japan        | 101.79          | Stable Cs          | Coniferous            |
| 10   | Andosol     | Japan        | 119.85          | Carrier             | Coniferous            |
| 11   | Andosol     | Japan        | 143.25          | Stable Cs          | Deciduous             |
| 12   | Andosol     | Japan        | 144.35          | Stable Cs          | Deciduous             |
| 13   | Andosol     | Japan        | 148.31          | Stable Cs          | Deciduous             |
| 14   | Andosol     | Japan        | 150.81          | Carrier             | Deciduous             |
| 15   | Andosol     | Japan        | 163.73          | Stable Cs          | Coniferous            |
| 16   | Andosol     | Japan        | 163.98          | Stable Cs          | Coniferous            |
| 17   | Andosol     | Japan        | 166.06          | Stable Cs          | Deciduous             |
| 18   | Andosol     | Japan        | 167.82          | Carrier             | Coniferous            |
| 19   | Andosol     | Japan        | 168.11          | Carrier             | Deciduous             |
| 20   | Andosol     | Japan        | 172.51          | Carrier             | Deciduous             |
| 21   | Lateritic clay | Indonesia  | 186.84          | Carrier free       | Tropical vegetation    |
| 22   | Andosol     | Japan        | 189.23          | Carrier free       | Deciduous             |
| 23   | Andosol     | Japan        | 191.15          | Stable Cs          | Coniferous            |
The relation of C/N ratio and CEC to the RIP value, in this case is RIP\textsubscript{carrier-free}, is presented in Figure 5. From this picture, we can see that IDN soil have the lowest CEC value as well as of C/N ratio. The low organic carbon content in IDN soil as shown on Table 1, may contributed to the weak of capacity exchange cations. Therefore, RIP for IDN soil also have a low value. There are no significant C/N ratio and CEC differences between coniferous and deciduous soil from Iwate. This is supported by XRD result that mineral content in coniferous and deciduous soil of Iwate soil are similar.
4. Conclusion

RIP and other psychochemical properties were determined for deciduous and coniferous soil from Iwate and soil from Indonesia. The RIP values of all soil sample were varied from 27.27 to 522.66 mmol kg\(^{-1}\). The carrier effect on RIP was conducted using carrier 134Cs, carrier-free that contains 134Cs, 136Cs and 137Cs and 133Cs stable isotopes. Correlation between different type of carrier suggests a relatively lower RIP for stable caesium as a carrier. This phenomena could be caused by competitive adsorption among of caesium ions in the frayed edge sites of soil mineral. Organic carbon content and CEC also contributed to the RIP value. No significant C/N ratio and CEC difference found between deciduous and coniferous forest soil as they also have similarity of mineral content in soil.

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