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Japan Atomic Energy Agency

Bioaccessibility of Fukushima accident-derived Cs in soils and the contribution of soil ingestion to radiation doses in children

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ABSTRACT

Ingestion of contaminated soil is one potential internal exposure pathway in areas contaminated by the Fukushima Daiichi Nuclear Power Plant accident. Doses from this pathway can be overestimated if the availability of radioactive nuclides in soils for the gastrointestinal tract is not considered. The concept of bioaccessibility has been adopted to evaluate this availability based on *in vitro* tests. This study evaluated the bioaccessibility of radioactive cesium from soils via the physiologically based extraction test (PBET) and the extractability of those via an extraction test with 1 mol L^{-1} of hydrochloric acid (HCl). The bioaccessibility obtained in the PBET was 5.3% ± 1%, and the extractability in the tests with HCl was 16% ± 3%. The bioaccessibility was strongly correlated with the extractability. This result indicates the possibility that the extractability in HCl can be used as a good predictor of the bioaccessibility with PBET. In addition, we assessed the doses to children from the ingestion of soil via hand-to-mouth activity based on our PBET results using a probabilistic approach considering the spatial distribution of radioactive cesium in Date City in Fukushima Prefecture and the inter-individual differences in the surveyed amounts of soil ingestion in Japan. The results of this assessment indicate that even if children were to routinely ingest a large amount of soil with relatively high contamination, the radiation doses from this pathway are negligible compared with doses from external exposure owing to deposited radionuclides in Fukushima Prefecture.

Keywords: Fukushima Daiichi Nuclear Power Plant Accident, soil ingestion, bioaccessibility, radiation dose, probabilistic approach

1. INTRODUCTION

The Fukushima Daiichi Nuclear Power Plant accident (the Fukushima accident) caused large-scale contamination owing to radioactive materials, and people living in contaminated areas are exposed to radiation in their daily lives. Therefore, determining the radiation doses received by people to manage radiation exposure in areas contaminated by the Fukushima accident is necessary. Radiation exposure occurs through internal and external pathways, and the ingestion of soil contaminated by radioactive Cs is one potential internal exposure pathway[1,2]. In particular, after the Fukushima accident, this pathway has been a concern for people with children in terms of their exposure when they play in sandboxes and playgrounds[3–5]. Therefore, to address their anxiety and improve the child-rearing environment in areas contaminated by the accident, understanding and explaining the effect of this pathway on children is necessary.

To assess radiation doses from the soil ingestion pathway, several factors related to the transfer of radioactive Cs from the ground into the human body need to be considered as along with the mechanisms of absorption in the human body. In accordance with the recommended models[6,7], at least three components need to be assessed. The first is the amount of soil ingested. To parameterize this factor, it is necessary to model a lifestyle habit related to contact with the contaminant considering inter-population variability owing to individual attributes such as age and occupation. The second is the concentration of radioactive Cs and its enrichment following the transfer. When people ingest soils contaminated by radioactive Cs through hand-to-mouth activities, the transfer of soil from the ground to the hands results in the selection of fine particles[8–10]. Consequently, the size classification of the transfer results in the enrichment of the concentration of radioactive Cs. Therefore, the particles that contribute to exposure from ingestion may have higher or enriched concentrations of contaminants relative to the original soils. This factor is called the enrichment factor[6,9]. Third, the fraction of metals in an administered soil that reaches the body fluids compartment from the gastrointestinal tract needs to be considered. This factor is known as "bioavailability"[6,7,11–16]. It is well known that metals in soils are not absorbed in the gastrointestinal tract

with 100% bioavailability. Therefore, exposure to radiation from ingestion pathways can be overestimated if bioavailability is not considered. Even though data on bioavailability can be obtained from *in vivo* tests on humans or animals, *in vivo* tests involve several issues such as difficult procedures and ethical concerns. In the light of these issues, the concept of "bioaccessibility" has been adopted for health risk assessments of soil ingestion, which is defined as the fraction of soluble substances in the gastrointestinal environment and those available for absorption[14]. Therefore, on the basis of the results of *in vitro* tests, the evaluation of bioaccessibility for radioactive Cs can be calculated as the ratio of radioactive Cs dissolved from the soil sample in an *in vitro* test to that present in the soil sample before the *in vitro* test[6, 7, 11–15].

To assess radiation doses from the ingestion pathway, the International Commission on Radiological Protection (ICRP) has formulated a biokinetic model[17] (**Fig. 1).** This biokinetic model considers the transit of ingested radioactive materials through four regions of the alimentary tract, i.e., the stomach, small intestine, upper large intestine, and lower large intestine. In this model, bioaccessibility is equal to the absorption fraction from the small intestine to the body fluids, which is called f_1 in the ICRP model[17–21], because the ICRP model was developed on the assumption that the absorption of radionuclides to the body fluids occurs entirely in the small intestine. The transfer coefficient from the small intestine to the body fluids, λ_B, can be represented in the following form using the f_1 value [17]:

$$\lambda_B = \frac{f_1 \cdot \lambda_{SI}}{1 - f_1}. \quad (1)$$

Here, λ_i is the transfer coefficient from compartment *i* to the next compartment. Even though the value of 1 has been used for the f_1 value in ICRP publications[17–21], Cs that is incorporated into insoluble particles such as soils is less available for absorption. Therefore, if we assess doses from the ingestion of soils contaminated by radioactive Cs, it is necessary to analyze the doses based on the actual bioaccessibility of Cs adsorbed on soils.

To simulate the effects of human digestion, several *in vitro* techniques have been developed[6,7,11–15]. Even though some studies have been based on these techniques, it is well known that the bioaccessibility of

metals in soils depends on the types of contaminants and soils[11]. Therefore, contaminant- and site-specific bioaccessibility is necessary to assess doses from soil ingestion in areas contaminated by the Fukushima accident. One aim of this study is to evaluate the bioaccessibility of radioactive Cs in order to assess doses from the ingestion of soil contaminated by the Fukushima accident. To this end, we performed the physiologically based extraction test (PBET)[14,15] using soil samples from Fukushima Prefecture.

PBET was recommended as a practical methodology for *in vitro* measurement to determine the bioaccessibility of metals in soils. However, the PBET procedure is more complex than procedures of single-extraction tests such as HNO_3 or HCl extraction. If we applied the PBET to assess the bioaccessibility in areas contaminated by a nuclear accident, enormous amount of testing would be needed to evaluate the bioaccessibility with great accuracy when considering the differences in soil types and contaminants, and thus generate much radioactive waste. Because a vast area is contaminated by a nuclear accident and there are various soil types corresponding to the difference of areas. As pointed out by Beresford et al.[22], the ability to predict radioactive Cs bioaccessibility rapidly without complex procedures is desirable, especially in the event of accidental contamination. Here, we focus on extraction tests with 1N HCl, which has been adopted under Japanese national law to evaluate the fractional absorption of heavy metals in the gastrointestinal system[16]. In this study, we define “extractability” as the ratio of radioactive Cs dissolved from a soil sample to that present in the soil sample before the extraction test with 1N HCl. A previous study [23] explored the relationship between bioaccessibility with PBET and extractability with 1N HCl. However, there is currently no insight into the relationship between them with regards to Cs. Therefore, we aim to explore the relationship between the bioaccessibility of the Fukushima accident-derived Cs from soils with PBET and the extractability with 1N HCl extraction.

Finally, we assess the potential doses to children from hand-to-mouth activities after the Fukushima accident on the basis of PBET results for soils in Fukushima Prefecture considering hand-to-mouth activities actually observed in a Japanese kindergarten. Accordingly, we performed calculations using the biokinetic

model developed by the ICRP along with the bioaccessibility obtained from our experiments. In addition, to describe an assessment procedure of soil ingestion pathway via hand-to-mouth using a probabilistic approach is also an aim of this study.

2. MATERIALS AND METHODS

2.1. Soil Samples

Soil samples were collected in Date City, Fukushima Prefecture, in September 2014. Date City is located to the northwest of the Fukushima Daiichi Nuclear Power station at a distance of approximately 60 km. The samples were collected from bare surfaces in a park that was not decontaminated after the Fukushima accident prior to our sampling. The latitude and longitude of the sampling point is 37° 75' 85" N and 140° 68' 81" E. We collected six soil samples from the ground surface to a depth of 1 cm. The sampled soils were air-dried in a desiccator at 45°C for 24 h or until they were visually dried. After air drying, the soils were placed in clean sieves with 0.1-mm meshes and shaken on a mechanical shaker for 6 h. A mesh size of 0.1 mm was employed based on a previous study reporting that the soil particles ingested through the hand-to-mouth pathway are mostly smaller than 0.1 mm[10]. Soil pH was measured using a pH meter (DDK-TOA, HM-31P) in a sample with 1:5 ratio of soil to distilled water. The fraction of the organic matter in the soil samples was characterized using an ignition loss test in accordance with Japanese Industrial Standards (JIS)[24]. The characteristics of the soil samples are summarized in **Table I**.

2.2. Extraction Test

2.2.1. Extraction test with 1N HCl

The extraction test with 1N HCl was performed in accordance with Notification No. 19 from the Ministry of the Environment[16]. The notification stipulates that the soil-to-fluid ratio should be 3% (in grams per milliliter). Because radioactivity measurements will be used to determine the extracted quantity, the

samples need to contain greater amounts of radioactive nuclides to ensure measurement accuracy. In these tests, we used a 10% sample in addition to a 3% sample to explore the dependence of the extractability on the soil-to-fluid ratio. Soil samples weighing 5 g and 1.5 g were chosen for extraction with 50 mL of 1N HCl. The soil–liquid mixed samples were shaken with a mechanical shaker at 200 rpm for 2 h at 25°C. The shaken samples were centrifuged at 3,000 rpm for 10 min and then filtered through 0.45-μm filters.

2.2.2. The Physiologically based extraction test (PBET)

The PBETs were performed on the basis of the procedure suggested by Ruby et al.[14]. Soil-to-fluid ratios of 1%, used by Ruby et al.[14], and 10% were adopted for the same reason mentioned above. Soil samples weighing 5 g and 0.5 g were tested with 50 mL of artificial gastric juice. The gastric juice was prepared as follows: 1 L of distillated water was mixed with 1.25 g of pepsin (Wako Pure Chemical Industries, Ltd.), 0.5 g of citrate (Wako Pure Chemical Industries, Ltd.), 0.5 g of malate (Wako Pure Chemical Industries, Ltd.), 420 μL of lactic acid (Wako Pure Chemical Industries, Ltd.), and 500 μL of acetic acid (Wako Pure Chemical Industries, Ltd.). Ruby et al.[14] showed the dependence of lead and arsenic bioaccessibility on the pH value of the gastric juice. Apparently, bioaccessibility in the stomach and small intestine increases with decreasing pH value. In our tests, the pH of the gastric juice was set to 1.3, representing the fasting condition, for a conservative assessment of the doses. Even though the mixing of the soil with the gastric juice was achieved by passing argon gas through the reaction vessels by Ruby et al.[14], several studies[11,15] have shown that it is not necessary to maintain anaerobic conditions with argon gas and that the extraction results of mixing using a mechanical shaker within a temperature-controlled water bath appeared to correlate well with bioavailability values from swine studies[11]. Therefore, we used a mechanical shaker with a temperature-controlled water bath to mix the samples. The samples were shaken at 100 rpm in a water bath maintained at a constant temperature of 37°C for 1 h. After the gastric phase, hydrogen-ion concentrations of the sample solution were titrated to a pH of 7 by adding a cellulose tube containing a

saturated $NaHCO_3$ solution. Then, 1.75 mg mL^{-1} of bile salts (Wako Pure Chemical Industries, Ltd.) and 0.5 mg mL^{-1} of pancreatin were added to the solution. After an intestinal incubation time of 4 h, the samples were centrifuged at 3,000 rpm for 10 min and then filtered through 0.45-μm filters.

3. RESULTS AND DISCUSSION

3.1 Radioactivity of Soils and Enrichment

The results of the radioactivity measurement are summarized in **Table II**. The activities shown in this table were decay-corrected to September 11, 2014. The measurements were performed with an error of several percent using a high-purity germanium detector (ORTEC, GMX-30190). The concentration of ^{137}Cs in the dried soil sample from Date City ranged from 3.25×10^4 Bq kg^{-1} dry wt. to 7.16×10^4 Bq kg^{-1} dry wt. Note that the soil samples were collected within a depth of 1 cm from the ground surface in non-decontaminated areas. Therefore, the data does not represent the other areas in Date City that have been decontaminated. The ^{137}Cs concentration measured after size classification using a 0.1-mm sieve ranged from 6.67×10^4 Bq kg^{-1} dry wt. to 1.81×10^5 Bq kg^{-1} dry wt. Therefore, an enrichment factor, defined as the ratio of the ^{137}Cs concentrations measured before and after sieving, of 2.6 ± 0.3 was obtained.

It is well known that contaminant concentrations are higher in smaller soil particles than in larger soil particles, and Cs the same tendency has been reported in previous studies of soils contaminated by radioactive Cs[25–29]. While a generalized model to describe the relationship between the particle size and the concentration has not yet been developed, experiments conducted thus far have determined that the concentration of radioactive Cs depends on the particle size distribution of the soil and other parameters such as the organic matter content[25–29]. For example, Livens and Baxter[25] observed that ^{137}Cs was concentrated in clay-sized (<2 μm) particles with enrichment factors of 3–35 times. Spenzzano[27] reported that the concentration of ^{137}Cs in soil samples is essentially associated with finer size fractions. For clay-sized

particles, the enrichment factors for the concentrations of ^{137}Cs were 3–5 times higher than for the bulk samples[27].

In addition, Sheppard[6,9] suggested an *a priori* model to determine the enrichment factor owing to the adhesion of soil to the skin. Based on this model and the observation results of several metals (not-included Cs data), Sheppard[6] observed that the value of the enrichment factor for sand soils such as in playgrounds and sandboxes is 5–10. For other soils, the value is lower than 2–3. The experimental results summarized in **Table II** are consistent with the values obtained in previous studies, and we used an average enrichment factor of 2.6 in our assessments.

3.2 Extractability and Bioaccessibility of Radioactive Cs

Table III summarizes the results of radioactivity measurements using the liquid samples extracted with 1N HCl and those performing the PBET as well as the evaluated values of the extractability and bioaccessibility. When the soil-to-fluid ratio was set to 3% and 10%, the extractability was 0.16 ± 0.03 and 0.065 ± 0.01, respectively. The bioaccessibility values for soil-to-fluid ratios of 1% and 10% were 0.053 ± 0.01 and 0.0093 ± 0.003, respectively. As indicated by Cave et al.[15], the differences in the extractability and the bioaccessibility between individual samples are derived from a combination of a number of sources such as (i) the repeatability of the precision in the laboratory, (ii) the systematic bias of the PBET method, and (iii) the uncertainty that is induced during the sampling of the soils in the field. In our results, at least, there is a possibility that the differences in the bioaccessibility and the extractability shown in Table III were introduced owing to the differences of the character of the soils (e.g., the organic matter content and the particle size distribution).

The relation between the extractability determined using the extraction test with 1N HCl and the bioaccessibility determined using the PBET is shown in **Fig. 2**. These relation were experimentally determined on the basis of the experimental results from the soil-to-fluid ratios of 3% and 1% for the

extraction test with 1N HCl and the PBET and the soil-to-fluid ratios of 10% and 10%. The soil-to-fluid ratios of 3% and 1% are designated values as the extraction test with 1N HCl and the PBET in Japanese law and previous studies, respectively. The bioaccessibility was observed to be strongly correlated with the extractability in both cases. However, as shown in **Fig. 2**, the correlation determined on the basis of the experimental results from the soil-to-fluid ratios of 3% and 1% and that determined from the soil-to-fluid ratios of 10% and 10% have the difference in the slope of the regression line. This difference is attributable to the dependence of bioaccessibility and extractability on the soil-to-fluid ratios. **Fig. 3** shows the bioaccessibility and the extractability as a function of the soil-to-fluid ratio. For bioaccessibitility, Hamel et al.[30] found an increase in bioaccessibility with decreasing soil-to-fluid ratio due to solution saturation. Although we need further experiments will be needed to get the information on the dependence of the bioaccessibitlity and the extractability on the soil-to-fluid ratio, our results shown in **Fig. 3** are not inconsistent with Hamel's results.

In addition, based on our results, if the soil-to-fluid ratios is more than 17%, the extractability is less than the bioaccessibility. This means that we cannot use the extractability via the extraction test with 1N HCl to conservatively determine the bioaccessibitilty under the experimental condition that the soil-to-fluid ratio is exceeding 17%. Therefore, it is reiterated that the soil-to-fluid ratio needs to be maintained at the recommended values, which are 3% and 1%, for the extraction test with 1N HCl and the PBET. However, these insights were obtained from limited data described in this study. For the bioaccessibility from the PBET and the extractability via the extraction test with 1N HCl, to explore the dependence of those on the soil-to-fluid ratio, and to clarify the relationship between them, further experiments and analysis will be needed.

Previous studies[22,31–40] have addressed the bioavailability and bioaccessibility of radioactive Cs in soils and foodstuff (**Table IV**). It is generally accepted that Cs ingested as soluble Cs is well absorbed in the gastrointestinal tracts of humans and animals. Results from controlled studies on human subjects indicate that

orally administered soluble ^{137}Cs is rapidly and almost completely absorbed[31,32,35,41]. Furthermore, radioactive Cs in foodstuffs is equally available for absorption in humans and animals[33,35,39]. By contrast, previous studies clearly indicate that insoluble radioactive Cs such as that adsorbed on soils tends to be unavailable for absorption in the gastrointestinal tract compared with soluble Cs and other foodstuffs such as meats and grass[22,32,33,36–40].

Our results, which were obtained by performing extraction tests using only soil samples (**Table III**), agree well with the literature. However, the bioavailability of radioactive Cs in soil depends on organic content[42,43], the origin of the Cs (worldwide fallout or the Chernobyl accident)[44], and soil physicochemical characteristics[38]. Therefore, further investigations are required to clarify the uncertainty in terms of different soil types and physical and chemical forms of Cs adsorbed on soil.

3.3 Doses from Soil Ingestion to the Public Living in Areas Affected by the Fukushima Accident

The annual committed effective doses after March 15, 2011 were assessed for soil ingestion via hand-to-mouth activities for the contributions of ^{134}Cs and ^{137}Cs. The assessments were performed in terms of doses to children because children are more vulnerable to radiation exposure and have more frequent contact with soil than adults. Age categories of "1 y" and "5 y" were adopted for our assessments, as defined by ICRP recommendations[18,20,21]. The categories of 1 y and 5 y include the age ranges of 1–2 y and 2–7 y, respectively.

The radiation dose from ingested soils, D_{soil}, during the period between t_1 and t_2 is expressed as follows:

$$D_{soil,j} = \int_{t_1}^{t_2} SI_j \cdot A_{Cs137}(t) \cdot EF \cdot \left(C_{Cs134} \cdot k_{Cs134,j} + k_{Cs137,j}\right) \cdot dt, \quad (2)$$

where the index j represents the age categories. SI is the amount of soil ingested per day (kg d^{-1}), $A_{Cs137}(t)$ is the concentration of ^{137}Cs in the soil at an elapsed time t after the contamination (Bq kg^{-1}), EF is the

concentration enrichment factor, C_{Cs134} is the ratio of the concentration of ^{134}Cs to that of ^{137}Cs, and $k_{Cs134,j}$ and $k_{Cs137,j}$ are the committed effective dose coefficients of ^{134}Cs and ^{137}Cs, respectively (Sv Bq^{-1}).

The committed effective dose coefficients are evaluated as the sum of the committed equivalent doses to the organs or tissues per unit radionuclide ingested. We calculated the coefficients using the ICRP's biokinetic model shown in **Fig. 1**. In these calculations, we used the DCAL system, which was developed by Oak Ridge National Laboratory[45–47]. In the calculation for the coefficients, the standard assumption adopted in the ICRP model is that the absorption of the radionuclides to the body fluids occurs entirely in the small intestine. This fractional transfer from the small intestine to the body fluids is called f_1 in ICRP publications[17–21] and is equal to the bioaccessibility. Even though a value of 1 has been used for this parameter in ICRP publications[17–21], in this study, we made calculations with various values of f_1 to explore the contribution of insoluble particles adsorbing ^{134}Cs and ^{137}Cs.

Figure 4 shows the committed effective dose coefficient as a function of the bioaccessibility (i.e., f_1). When a value of 1 was adopted for the bioaccessibility, the committed effective dose coefficients of ^{134}Cs and ^{137}Cs for the age categories 1 y and 5 y have the same value as in the ICRP publications[21]. Here, if complete absorption is indicated from the small intestine into the body fluids, in the same manner of the ICRP recommendation[19], the calculations of Eq. (2) were performed using a value of 0.99 for the bioaccissibility for technical reasons. In our assessments, we adopted committed effective dose coefficients, which were calculated using a bioaccessibility of 0.053 based on the experimental results. The coefficient values of ^{134}Cs and ^{137}Cs for the age category 1 y are 9.1×10^{-9} Sv Bq^{-1} and 9.0×10^{-9} Sv Bq^{-1}, respectively, and those for the age category 5 y are 5.2×10^{-9} Sv Bq^{-1} and 4.8×10^{-9} Sv Bq^{-1}, respectively. Note that the ICRP is currently revising the biokinetic model[48–50] and this revision is not yet complete. Therefore, once the models and coefficients are prepared, it may be necessary to assess the doses from the ingestion pathway based on the new model.

The hand-to-mouth activities of children were assumed to take place in Date City in Fukushima Prefecture. A probabilistic approach was adopted to consider the spatial and inter-individual variability of the contamination and the behavioral patterns related to soil ingestion. This approach has been widely used for health risk assessments[7,51,52]. The technical basis and the data to use in this approach have also been developed in the field of risk assessment for the ingestion of contaminated soil[53–56]. However, to reflect the characteristics of the physical attributes and lifestyle habits of Japanese children, we used data based on surveys performed in Japan. In our assessments, we used the distributions of the amounts of soil ingested per day, which were determined from actual surveys performed in a Japanese kindergarten[57]. Part of the soil that adheres to the hands can be a potential exposure source for hand-to-mouth activities. Therefore, to avoid underestimates in the amounts ingested by the children, the doses were assessed based on the assumption that all the adhered soils were ingested. The distributions of soils adhered to hands are characterized by a lognormal form. The geometric mean (GM) and geometric standard deviation (GSD) values of each age category are summarized in **Table V**. We used these statistics as the input for *SI* in Eq. (2) for our probabilistic assessments.

The spatial distribution of the surface density of ^{137}Cs (in Bq m^{-2}) in Date City was measured by the Ministry of Education, Culture, Sports, Science, and Technology (MEXT)[58,59] after the Fukushima accident. From these measurements, the GM and GSD of the concentration of ^{137}Cs in the soil in Date City were 8.66 × 10^{2} Bq kg^{-1} dry wt. and 1.60, respectively, as of November 2013. Conversions from surface density (Bq m^{-2}) to bulk concentration (Bq kg^{-1}) were made assuming that the soil density was 1.6 g cm^{-3}. The concentration of ^{134}Cs on the ground was assumed to be equal to that of ^{137}Cs as of March 15, 2011 based on UNSCEAR's report[60]. In our probabilistic approach, the set of ^{137}Cs concentration, $A_{Cs137}(t)$, and the amount of soils ingested, *SI*, was generated on the basis of the statistical characteristics described above using a Monte Carlo analysis code, i.e., GSALab[61], which was developed by the Japan Atomic Energy Agency. The

dose calculations were performed for 10,000 sets of sample values. Relative errors in the assessments were less than 0.05. The parameters used in our assessment are summarized in **Table V**.

Figure 5 shows the distribution of the annual committed effective doses to children for the two age categories in Date City during the first year after the contamination (i.e., March 15, 2011–March 15,, 2012). These results only consider the contributions of ^{134}Cs and ^{137}Cs. Therefore, the radiation exposure owing to other short-lived radionuclides such as ^{131}I and ^{132}Te were not considered. The GM of the annual committed effective doses to children for the age categories 1 y and 5 y are 0.19 μSv and 0.07 μSv, respectively. Even if the children were to ingest a large amount of soil with relatively high contamination, the 95th percentile of the doses from this pathway is only 0.73 μSv and 0.39 μSv per year for the age categories 1 y and 5 y, respectively. **Figure 6** shows the time-dependence of the annual committed effective doses to children for the age category 1 y as a function of time after the contamination. The doses were calculated on the condition that $t_2 - t_1$ in Eq. (2) is 1 year. The dependence was calculated from the radioactive decay without considering the transfer of Cs in the environment owing to weathering or migration. The annual committed effective doses from the ingestion of contaminated soils were less than 1 μSv for a long time after contamination.

As compared to the doses from other exposures pathways, for example, the doses from external exposures measured during the period between July 2012 and June 2013 in Date City was 0.89 mSv y^{-1} on a population-wide average[62]. Therefore, the doses from the ingestion of contaminated soils are ten or more times less than those from external exposure in Date City. In addition, we performed sensitivity analysis to clarify when we would actually need to worry that soil ingestion pathway could pose a significant threat in children. In our calculations, the 95th percentile of the doses were obtained on the condition that a child ingests about 50 mg of soils contaminated with a concentration of about 1,000 Bq kg^{-1} per day. Based on our sensitivity analysis, if a child ingest 1000 mg of soils contaminated with a concentration of 10,000 Bq kg^{-1} per day, the annual committed effective dose from this pathway is 0.16 mSv. This level of doses is nearly

equal to the doses from external exposures and it would be a concern in the contaminated area. However, in fact, such level of amount of soil ingestion is far from reality, and the concentration of ^{137}Cs is less than 5,000 Bq kq^{-1} in Date City[58,59]. As consequently, we concluded that the contribution of ingestion of contaminated soils to the doses in children is negligible compared with those from external exposures.

After the Fukushima accident, a few investigations of the internal radiation contamination by radioactive Cs were performed on children in Fukushima Prefecture[63–66]. Tsubokura[65,66] reported the results of one such measurement program using a whole body counter (WBC) in the city of Minamisoma during the period between April 2012 and March 2013 and between May and July 2013. The detection limits of the WBCs were 220 Bq body^{-1} for ^{134}Cs and 250 Bq body^{-1} for ^{137}Cs. More than 4,000 elementary school (6–12 y) and junior high school students (12–15 y) were measured in the two programs, and most of the students were observed to have non-detectable levels of radioactive Cs. Tsubokura et al.[66] also assessed the maximum annual effective dose if the children daily ingested an equivalent amount of soil within the detection limit of the WBC measurements. The maximum level of annual committed effective dose owing to ^{134}Cs and ^{137}Cs was estimated to be 0.066 mSv y^{-1} for children aged 6–7 years. This result is consistent with our results, which showed that the contribution of contaminated soil ingestion to internal doses is negligibly small.

These assessments were performed on the basis of some important limitations and assumptions. First, bioaccessibility was evaluated in the fasting state and gastric pH values were selected to represent that state. When the pH values were varied in the range of 1.3–4, corresponding to various gastric states, the bioaccessibility decreased by several times[14]. Second, the extraction tests were performed using soil samples taken from a single site. Therefore, the bioaccessibility obtained from this study is site-specific, which limits its general use. Finally, the doses were assessed on the assumption that children ingest the same amounts of soil per day over the assessment period, i.e., one year, to avoid underestimation. In general, it is known that the distribution of the estimated average daily soil ingestion over a short period is more widely spread than

the actual soil ingestion over a longer period[67,68]. Therefore, the 95th percentiles of the doses for the observed 1-day soil ingestion estimates will be higher than those obtained using the true 1-day soil ingestion. To reduce the variabilities and uncertainties owing to the limitations mentioned above, further studies are needed to examine various types of soils and long-term observations of lifestyle habits. In addition, doses owing to internal exposure could occur from pathways other than soil ingestion (e.g., ingestion of contaminated foodstuffs or the resuspension of depositions)[2]. Therefore, to accurately assess the doses from internal exposure, we need to make further efforts to consider the contributions from these pathways.

4. CONCLUSIONS

The bioaccessibility of radioactive Cs in soils was evaluated by performing extraction tests with 1N HCl and PBET. The bioaccessibility value obtained in the PBET was 0.053 ± 0.01, and the extractability in the extraction tests with 1N HCl was 0.16 ± 0.03. The bioaccessibility was strongly correlated with the extractability. This result indicates the possibility that the extractability obtained in extraction tests with 1N HCl can be used as a good predictor of the bioaccessibility obtained by performing the PBET. Furthermore, we assessed the doses to children from ingestion of soils via hand-to-mouth activities using a probabilistic approach based on the spatial distribution of radioactive Cs in Date City in Fukushima Prefecture and the inter-individual differences in the amounts of soil ingestion surveyed in a Japanese kindergarten. The GMs of the annual committed effective doses to children in the age categories of 1 y and 5 y were 0.19 μSv and 0.07 μSv, respectively. Even if the children were to ingest large amounts of soils with relatively high contaminations, the 95th percentile of the doses from this pathway is below one μSv per year. Compared to the actual measurements of the doses from external exposures, the doses from the ingestion of contaminated soils are ten or more times less than those from external exposure. As consequently, we concluded that the contribution of ingestion of contaminated soils to the doses in children is negligible compared with those from external exposures. However, these assessments were performed under important limitations and

assumptions. To reduce the variabilities and uncertainties owing to these limitations, further studies are necessary using various types of soils and long-term observations of lifestyle habits.

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REFERENCES

1. Calabrese E. Improving the scientific foundations for estimating health risks from the Fukushima incident, *Proc Natl Acad Sci USA*, 2011; 108(49): 19447–19448.
2. IAEA. Actions to Protect the Public in an Emergency due to Severe Conditions at a Light Water Reactor; 2013. EPR-NPP-PPA.
3. Fukushima City Assembly. Minutes of regular meeting of Fukushima City Assembly, 2011; No. 4 9-December. (in Japanese) Available at: http://www.city.fukushima.fukushima.jp/voices/ (accessed 6 April 2016)
4. Fukushima Prefectural Assembly. Minutes of regular meeting of Fukushima Prefectural Assembly, 2012; No. 4, 27-Feburuary. (in Japanese) Available at: http://www.kaigiroku.net/kensaku/fukushima/fukushima.html (accessed 6 April 2016)
5. Kawahara K. and Kasama H. Sandbox Project in the areas affected by the Fukushima Daiichi Nuclear Power Plant Accident, *Health Care*, 2013; 55(8); 555–559. (in Japanese)
6. Sheppard S.C. Parameter Values to Model the Soil Ingestion Pathway. *Environ Monit Assess*, 1995; 34(1): 27–44.
7. Paustenbach D.J. The Practice of Exposure Assessment: A State-of-the-art Review. *J Toxicol Environ Health B Crit Rev*, 2000; 3(3): 179–291.
8. Sheppard S.C. and Evenden W.G. Concentration Enrichment of Sparingly Soluble Contaminants (U, Th and Pb) by Erosion and by Soil Adhesion to Plants and Skin. *Environ Geochem Health*, 1992; 14(4): 121–131.
9. Sheppard S.C. A model to Predict Concentration Enrichment of Contaminants on Soil Adhering to Plants and Skin. *Environ Geochem Health*, 1995; 17(1): 13–20.

10. Ikegami M. Yoneda M., Tsuji T., Bannai O. and Morisawa S. Effect of Particle Size on Risk Assessment of Direct Soil Ingestion and Metals Adhered to Children's Hands at Playgrounds. *Risk Anal*, 2014; 34(9): 1677–1687.
11. Ruby M.V., Schoof R., Brattin W., Goldade M., Post G., Harnois M., Mosby D.E., Casteel S.W., Berti W., Carpenter M., Edwards D., Cragin D. and Chappell W. Advances in Evaluating the Oral Bioavilability of Inorganic in Soil for Use in Human Health Risk Assessment, *Environ Sci Technol*, 1999; 33(21): 3697–3705.
12. Oomen A.G., Hack A., MMinekus M., Zeijdner E., Cornelis C., Schoeters G., Verstraete W., Wiele T.V.D, Wragg J., Rompelberg C.J.M., Sips A.J.A.M., and Wijnen J.H.V. Comparison of Five In Vitro Digestion Models To Study the Bioaccessibility of Soil Contaminants, *Environ Sci Technol*, 2002; 36(15): 3326–3334.
13. Wragg J. and Cave M.R. In-vitro Methods for the Measurement of the Oral Bioaccessibility of Selected Metals and Metalloids in Soils: A Critical Review: Environment Agency; 2003. 34 p. R&D Technical Report P5-062/TR/01.
14. Ruby M.V., Davis A., Schoof R., Eberle S. and Sellstone C.M. Estimation of Lead and Arsenic Bioavailability Using a Physiologically Based Extraction Test. *Environ Sci Technol*, 1996; 30(2): 422–430.
15. Cave M.R., Wragg J., Palumbo B. and Klinck B.A. Measurement of the Bioaccessibility of Arsenic in UK Soils, Environment Agency; 2003. 108 p. R&D Technical Report P5-062/TR02.
16. Ministry of the Environment, Government of Japan. On the Dojo ganyuryo tyosa ni kakaru sokutei houhou wo sadameru ken, Ministry of Environment Notification No. 19 2003. (in Japanese)
17. ICRP. Limits for Intakes of Radionuclides by Workers. ICRP Publication 30 (Part 1), *Ann. ICRP* 2(3–4), 1979.

18. ICRP. Age-dependent Doses to Members of the Public from Intake of Radionuclides—Part 1. ICRP Publication 56, Ann. ICRP 20(2), 1990.

19. ICRP. Age-dependent Doss to Members of the Public from Intake of Radionuclides— Part 2 Ingestion Dose Coefficients. ICRP Publication 67, Ann. ICRP 23(3/4), 1993.

20. ICRP. Age-dependent Doses to Member of the Public from Intake of Radionuclides—Part 3 Ingestion Dose Coefficients. ICRP Publication 69, Ann. ICRP 25(1), 1995.

21. ICRP. Age-dependent Doses to the Member of the Public from Intake of Radionuclides—Part 5 Compilation of Ingestion and Inhalation Coefficients. ICRP Publication 72, Ann. ICRP 25(1), 1995.

22. Beresford N.A., Mayes R.W., Cooke A.I., Barnett C.L. Howard B.J., Lamb C.S. and Naylor G.P.L. The Importance of Source-Dependent Bioavailability in Determining the Transfer of Ingested Radionuclides to Ruminant-Derived Food Products. *Environ Sci Tech*, 2000; 34(21): 4455–4462.

23. Bannai O., Tsuji T., Yoneda M. and Morisawa S. Measurement of Heavy Metal Pollution and Health Risk Evaluation of Oral Soil Intake. Environmental Engineering Research, 2003; 40: 659–666. (in Japanese)

24. Japan Industrial Standard. Test Method for Ignition Loss of Soils 2009. JIS A 1226:2009.

25. Livens F.R. and Baxter M.S. Particle Size and Radionuclide Levels in Some West Cumbrian Soils. *Sci Total Environ*, 1988; 70: 1–17.

26. He Q. and Walling D.E. Interpreting Particle Size Effects in the Adsorption of ^{137}Cs and Unsupported ^{210}Pb by Mineral Solids and Sediments. *J Environ Radioactivity*, 1996; 30(2): 117–137.

27. Spezzano P. Distribution of pre- and post-Chernobyl Radio Caesium with Particle Size Fractions of Soils. *J Environ Radioactivity*, 2005; 83: 117–127.

28. Sakaguchi A., Yamamoto M., Hoshi M., Imanaka T., Apsalikov K.N. and Guesv B.I. Radiological Situation in the Vicinity of Semipalatinsk Nuclear Test Site: Dolon, Mostik, Cheremushka and Budene Settlements. *J Radiat Res*, 2006; 47: Suppl., A101–A116.

29. Sakaguchi A., Tanaka K., Iwatani H., Chiga H., Fan Q., Onda Y. and Takahashi Y. Size Distribution Studies of ^{137}Cs in River Water in the Abukuma Riverine System Following the Fukushima Dai-ichi Nuclear Power Plant Accident. *J Environ Radioactivity*, 2015; 139: 379–389.

30. Hamel S.C., Buckley B. and Lioy P.J. Bioaccessibility of Metals in Soils for Different Liquid to Solid Ratios in Synthetic Gastric Fluid. *Environ Sci Technol*, 1998; 32(3): 358–362.

31. Yamagata N., Iwashima K., Nagai T., Watari K. and Iinuma T.A. In Vivo Experiment on the Metabolism of Cs in Human Blood with Reference to Rubidium and Potassium. *J Rad Res*, 1966; 7(1): 29–46.

32. LeRoy G.V., Rust J.H. and Hasterlik R.J. The Consequences of Ingestion by Man of Real and Simulated Fallout. *Health Phys*, 1966; 12(4): 449–473.

33. Beresford N.A., Mayes R.W., Howard B.J., Eayres H.F., Lamb C.S., Barnett C.L. and Segal M.G. The Bioavailability of Different Forms of Radiocaesium for Transfer across the Gut of Ruminants. *Radiat Protect Dosim*, 1992; 41(2-4): 87–91.

34. Beresford N.A., Mayes R.W., Barnett C.L., MacEachern P.J., Crout N.M.J., Variation in the metabolism of radiocaesium between individual sheep. *Radiat Environ Biophys*, 1998; 37: 277–281.

35. Henrichs K., Paretzke H.G., Voigt G. and Berg D. Measurements of Cs Absorption and Retention in Man. *Health Phys*, 1989; 57(4): 571–578.

36. Cooke A.I., Green N., Rimmer D.L., Weekes T.E., Wilkins B.T., Beresford N.A., Fenwick J.D. Absorption of Radiocaesium by Sheep after Ingestion of Contaminated Soils. *Sci Total Environ*, 1996; 192(1): 21-9.

37. Talbot R.J., Newton D. and Segal M.G. Gastrointestinal Absorption by Rats of ^{137}Cs and ^{90}Sr from U_3O_8 Fuel Particles: Implications for Radiation Doses to Man after a Nuclear Accident. *Radiat Protect Dosim*, 1993; 50(1): 39–43.

38. Ellickson K. M., Schopfer C. J. and Lioy P. J., The Bioaccessibility of Low Level Radionuclides from Two Savannah Site Soils. *Health Phys*, 2002; 83(4): 476–484.

39. McKay W.A., and Memmott S.D. An Investigation of the Availability of ^{137}Cs and $^{239+240}Pu$ for Gut Absorption in Winkles following cooking and in vitro Simulated Gastro-intestinal Digestion. *Food Addit. Contaminatns.* 1991; 8(6): 781–786.

40. Sheppard S.C., Evenden W.G. and Schwartz W.J. Ingested Soil: Bioavailability of Sorbed Lead, Cadmium, Cesium, Iodine, and Mercury. *J Environ Qual*, 1995; 24(3): 498–505.

41. Rosoff B., Cohn S.H. and Spencer H. Cs-137 Metabolism in Man, *Radiat Res*, 1963; 19(4): 643–654.

42. Belli M., Blasi M., Capra E., Drigo A., Menegon S., Piasentier E. and Sansone U. Ingested Soil as a Source of ^{137}Cs to Ruminants. *Sci Total Environ*, 1993; 136(3): 243–249.

43. Hansen H.S. and Hove K. Radiocesium Bioavailability: Transfer of Chernobyl and Tracer Radiocesium to Goat Milk. *Health Phys*, 1991; 60(5): 665–673.

44. Ward G.M., Keszthelyi Z., Kanyar B., Kralovanszky U.P. and Johnson J.E. Transfer of ^{137}Cs to Mils and Meat in Hungary from Chernobyl Fallout with Comparisons of Worldwide Fallout in the 1960s. *Health Phys*, 1989; 57(4): 587–592.

45. Cristy M., Eckerman K.F. Specific Absorbed Fractions of Energy at Various Ages from Internal Photon Sources, ORNL/TM-8381/V1-V7, 1987; Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA.

46. Cristy M., Eckerman K.F.SEECAL: Program to Calculate Age-dependent Specific Effective Energies, ORNL/TM-123551, 1993; Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA.

47. Eckerman K.F., Leggett R.W., Crist M., Nelson C.B., Ryman J.C., Sjoreen A.L. and Ward R.C. User's Guide to the DCAL System, ORNL/TM-2001/190, 2006; Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA.

48. ICRP. Human Alimentary Tract Model for Radiological Protection. ICRP Publication 100, Ann. ICRP 36(1–2), 2006.

49. ICRP. Compendium of Dose Coefficients based on ICRP Publication 60. ICRP Publication 119, Ann. ICRP 41, 2012.

50. ICRP. Occupational Intakes of Radionuclides: Part 1. ICRP Publication 130, Ann. ICRP 44(2), 2015.

51. Finley B. and Paustenbach D. The benefits of probabilistic exposure assessment: three case studies involving contaminated air, water, and soi, *Risk Anal*, 1994; 14(1): 53–73.

52. Cullen A.C. and Frey H.C. Probabilistic techniques in exposure assessment: a Handbook for dealing with Variability and Uncertainty in Models and Inputs. New York: Plenum Publishing Corporation; 1999. 335 p.

53. Finley B, Proctor D., Scott P., Harrington N., Paustenbach D. and Price P. Recommended Distributions for Exposure Factors Frequently Used in Health Risk Assessment. *Risk Anal*, 1994; 14(4): 533–553.

54. Finley B.L., Scott P.K. and Mayhall D.A. Development of a Standard Soil-to-Skin Adherence Probability Density Function for Use in Monte Carlo Analyses of Dermal Exposure. *Risk Anal*, 1994; 14(4): 555–569.

55. Stenek III E.J., Calabrese E.J. and Zorn M. Soil Ingestion Distribution for Monte Carlo Risk Assessment in Children. *Hum Ecol Risk Assess*, 2001; 7(2): 357–368.

56. USEPA. Risk Assessment Guidance for Superfund: Volume III – Part A, Process for Conducting Probabilistic Risk Assessment, United States Environmental Protection Agency; 2001. EPA 540-R-02-002.

57. Ikegami M. Yoneda M. and Morisawa S. The Validity of Default Value of Daily Soil Ingestion Evaluated by the Amount of Soil on Children's Hands. Environmental Engineering Research, 2007; 44: 653–659. (in Japanese)

58. MEXT. Results of Deposition of Radioactive Cesium of the Airborne Monitoring Survey in the Areas to which Evacuation Orders have been issued (Decay correction: November 19, 2014). Available at: http://emdb.jaea.go.jp/emdb/en/ (accessed 31 July 2016)

59. MEXT. Results of Deposition of Radioactive Cesium of the Eighth Airborne Monitoring Survey and Airborne Monitoring Survey Outside 80 km from the Fukushima Daiichi NPP (Decay Correction: November 19, 2014). Available at: http://emdb.jaea.go.jp/emdb/en/ (accessed 31 July 2016)

60. UNSCEAR. Annex A: Levels and Effects of Radiation Exposure due to the Nuclear Accident After the 2011 Great East-Japan Earthquake and Tsunami. United Nations; 2014. UNSCEAR 2013 Report: Sources, Effects and Risks of Ionizing Radiation.

61. Liu Q., Homma T., Nishimaki Y., Hayashi H., Terakado M. and Tamura S. GSALab Computer code for Global Sensitivity Analysis. Japan Atomic Energy Agency; 2010. 57 p. JAEA-Data/Code 2010-001. (in Japanese).

62. Date City. Results of analysis for actual measurements on doses from external exposures, 2013. (in Japanese) Available at: http://www.city.date.fukushima.jp/uploaded/attachment/10030.pdf (accessed 19 July 2016).

63. Hayano R.S., Tsubokura M., Miyazaki M., Satou H., Sato K., Masaki S. and Sakuma Y. Comprehensive whole-body counter surveys of Miharu-town school children for three consecutive years after the Fukushima NPP accident. *Proc Jpn Acad*, 2014; Ser. B 90: 211–213.

64. Hayano R.S., Tsubokura M., Miyazaki M., Satou H., Sato K., Masaki S. and Sakuma Y. Whole-body counter surveys of Miharu-town School Children for Four Consecutive Years after the Fukushima NPP accident. *Proc Jpn Acad*, 2015; Ser. B 91: 92–98.

65. Tsubokura M., Kato S., Nomura S., Morita T., Sugimoto A., Gilmour S., Kami M., Oikawa T. and Kanazawa Y. Absence of Internal Radiation Contamination by Radioactive Cs among Children Affected by the Fukushima Daiichi Nuclear Power Plant Disaster. *Health Phys*, 2015; 108(1): 39–43.

66. Tsubokura M., Kato S., Morita T., Nomura S., Kami M., Sakaihara K., Hanai T., Oikawa T. and Kanazawa Y. Assessment of the Annual Additional Effective Doses amongst Minamisoma Children

during the second Year after the Fukushima Daiichi Nuclear Power Plant Disaster. PLoS One, 2015; 10(6): e0129114.doi:10.1371/journal.pone.0129114.

67. Wallace L.A., Duan N. and Ziegenfus R. Can Long-Term Exposure Distributions Be Predicted from Short-Term Measurements. *Risk Anal*, 1994; 14(1): 75–85.

68. Sternek III E.J., Calabrese E.J. and Xu L. A Caution for Monte Carlo Risk Assessment of Long Term Exposures based on Short-Term Exposure Study Data. *Hum Ecol Risk Assess*, 1998; 4(2): 409–422.

Figure captions

Figure 1. The biokinetic model recommended by ICRP Publication 30 to describe the transfer of radionuclides in the gastrointestinal tract.

Figure 2. Relationship between the extractability determined by the extraction test with 1N HCl and the bioaccessibility determined by PBET for ^{137}Cs adsorbed on soils.

Figure 3. Dependence of the bioaccessibility of ^{137}Cs from soils via the PBET and the extractability of those via an extraction test with 1N HCl on soil-to-fluid ratio.

Figure 4. The committed effective dose coefficient of ^{134}Cs and ^{137}Cs for various values of bioaccessibility.

Figure 5. The distribution of committed effective doses from hand-to-mouth ingested soils contaminated by radioactive Cs for children in Date City during the period between March 15, 2011 and March 15, 2012.

Figure 6. Projection results of the time-dependence of committed effective doses from hand-to-mouth ingested soils contaminated by radioactive Cs for children in Date City.

Table captions

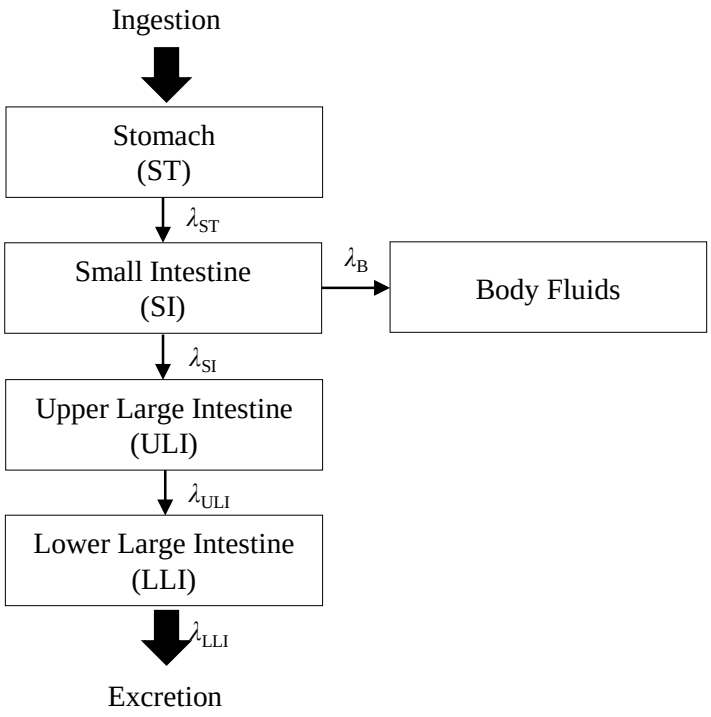
Table I. Characteristics of soil samples collected in Date City in Fukushima Prefecture.

Table II. Radioactive concentration of ^{137}Cs in soils before and after size classification.

Table III. Results of extractions with 1N HCl and PBET.

Table IV. Bioavailability or bioaccessibility of radioactive Cs for various types of dietary sources.

Table V. Values of parameters to assess doses from soil ingested via hand-to-mouth activities.



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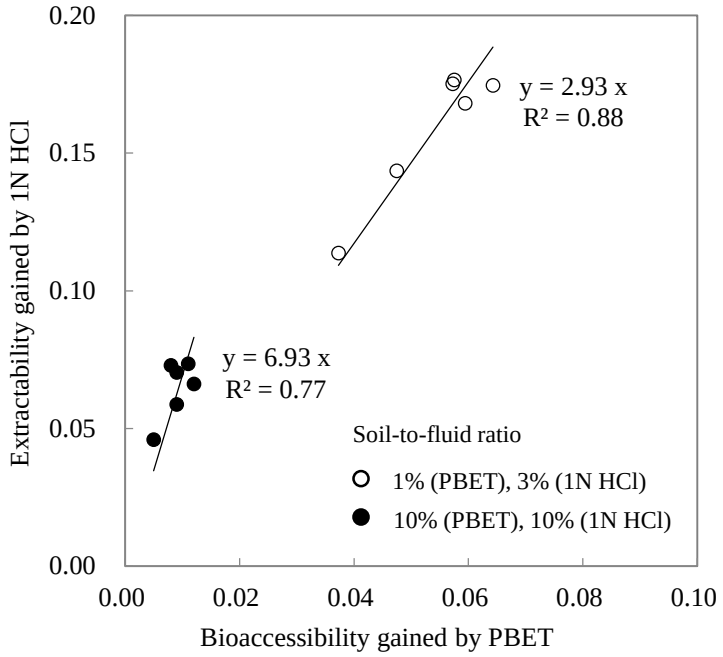

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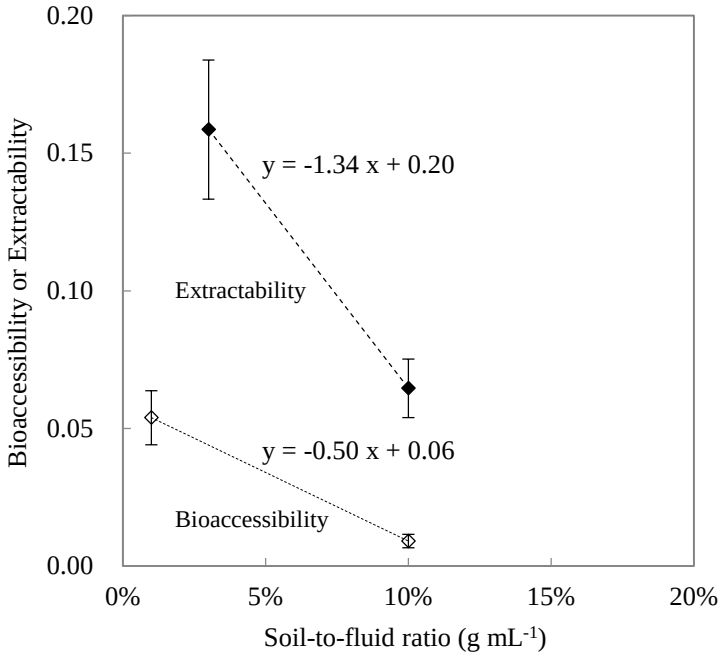

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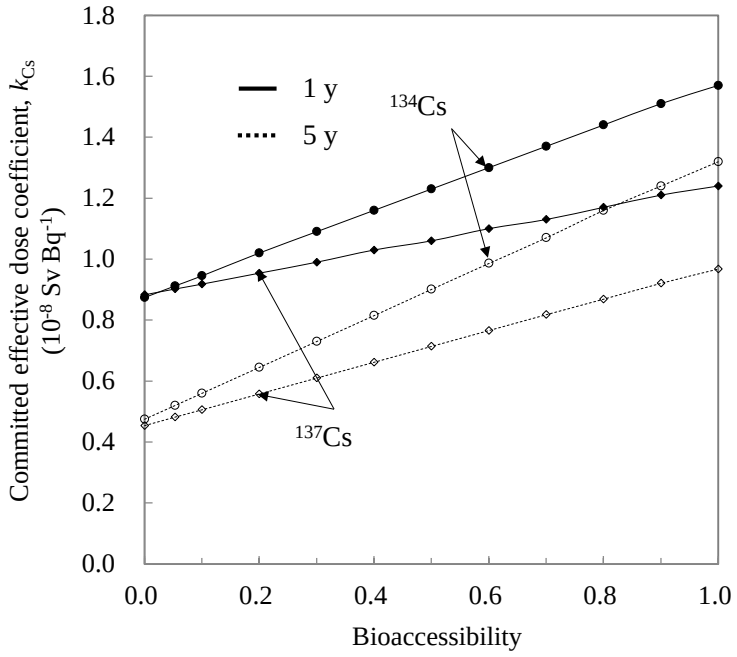

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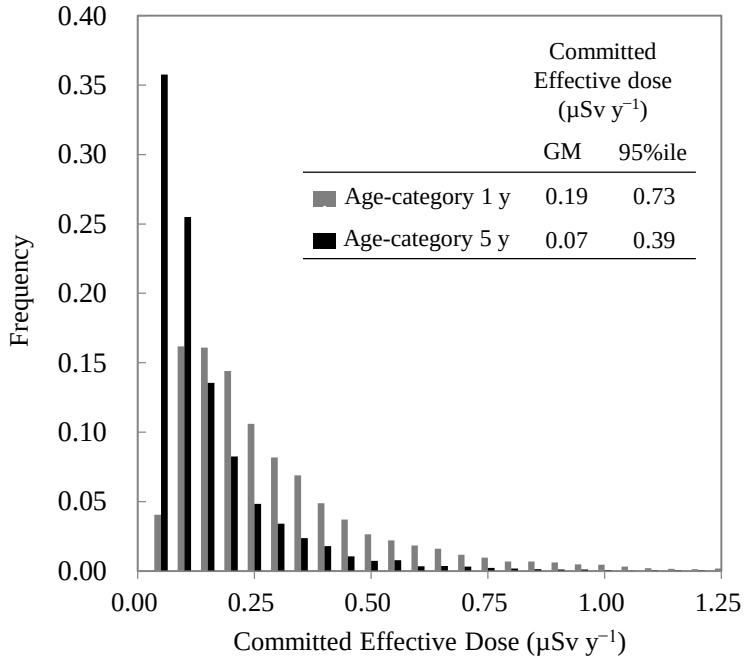

Fig. 5. The distribution of committed effective doses from hand-to-mouth ingested soils contaminated by ^{134}Cs and ^{137}Cs for children in Date City during the period between March 15, 2011 and March 15, 2012.

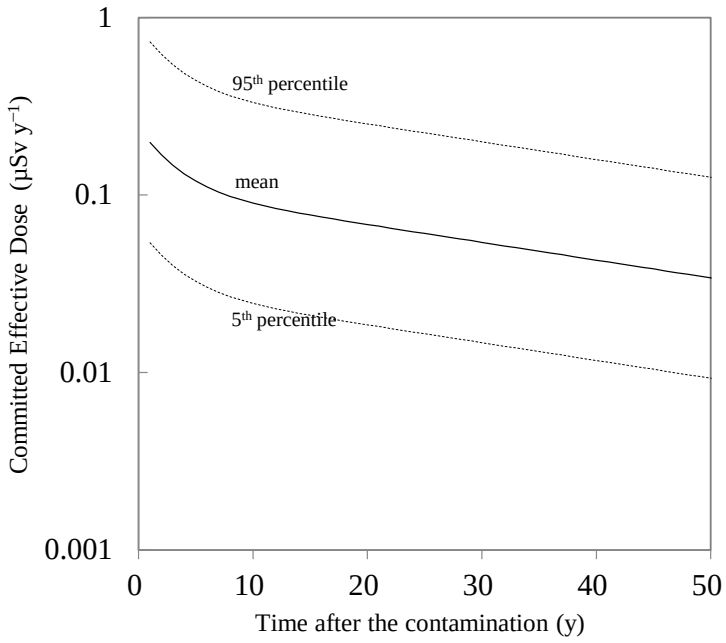

Fig. 6. Projection results of the time-dependence of committed effective doses from hand-to-mouth ingested soils contaminated by ^{134}Cs and ^{137}Cs for children in Date City.

Table I. Characteristics of soil samples collected in Date City in Fukushima Prefecture.

Sample No.	Sampling depth (cm)	Dried Soil (g dry wt.)	Sieved Soil by 100 μm (g dry wt.)	pH	Organic matter (%)
1	1	108	22.1	5.3	13%
2	1	118	24.4	5.1	14%
3	1	108	22.2	5.0	13%
4	1	106	20.9	5.1	13%
5	1	103	19.5	5.1	15%
6	1	83.1	18.7	5.2	14%

Table II. Radioactive concentration of ^{137}Cs in soils before and after size classification.

Sample No.	Dried Soil (Bq / kg (dry wt.))	Sieved Soil by 100μm (Bq / kg (dry wt.))	Sieved Soil by 100 μm/ Dried Soil
1	5.18×10^4	1.56×10^5	3.0
2	7.16×10^4	1.81×10^5	2.5
3	6.73×10^4	1.74×10^5	2.6
4	5.73×10^4	1.55×10^5	2.7
5	3.74×10^4	9.86×10^4	2.6
6	3.25×10^4	6.67×10^4	2.1

*Activities shown in this table are decay-corrected to the sampling date, i.e., September 11, 2014.

Table III. Results of extractions with 1N HCl and PBET.

Sample No.	Extraction with 1N HCl			Extracted with PBET		
	Soil-to-Fluid Ratio (g / mL)	Radioactivity (Bq / 50 mL)	Extractability[(1)]	Soil -to-Fluid Ratio (g / mL)	Radioactivity (Bq / 50 mL)	Bioaccessibility (Small Intestine)
1	3%	39	17%	1%	4.6	5.9%
2		48	18%		5.2	5.7%
3		46	18%		5.6	6.4%
4		41	18%		4.5	5.7%
5		21	14%		2.3	4.7%
6		11	11%		1.2	3.7%
1	10%	57	7.3%	10%	6.2	0.8%
2		67	7.4%		7.5	1.1%
3		58	6.6%		8.2	1.2%
4		55	7.0%		5.9	0.9%
5		29	5.9%		3.5	0.9%
6		15	4.6%		1.3	0.5%

[(1)] Extractability is defined as the ratio of the extracted radioactivity to that in soils sieved with a 100-μm filter.

Table IV. Bioavailability or bioaccessibility of radioactive Cs for various types of dietary sources.

In vivo or *in vitro* experiment	Dietary source	species	Bioavailability or Bioaccessibility	Ref.
In vivo	CsCl	human	0.85	(31)
	CsCl	human	0.89	(32)
	CsCl	sheep	0.84	(33)
	CsCl	sheep	0.87	(22, 34)
	CsCl	sheep	0.78	(22)
	CsCl	sheep	0.84	(22)
	CsCl	cattle	0.71	(22)
	Grass hay	sheep	0.73	(22)
	Clover hay	sheep	0.76	(22)
	Upland Grass	sheep	0.88	(33)
	Pasture grass	cattle	0.23	(22, 34)
	Calluna Valgaris	sheep	0.67	(33)
	Venison	human	0.78	(35)
	Lowland peat soil	sheep	0.03	(22, 36)
	Silty soil	sheep	0.13	(33)
	Alluvial gley soil	Sheep	0.19	(22, 36)
	Peaty podzol soil	Sheep	0.20	(22)
	Peaty podzol soil	Sheep	0.02	(22)
	Insoluble fallout particle	human	0.032	(32)
	Insoluble irradiated fuel particle	rat	0.075	(36)

In vitro	Savannah River Site soil	enzymolysis procedure	0.060–0.38	(38)
	Winkle with soil	enzymolysis procedure	0.13–0.63	(39)
	Alfalfa with soil	enzymolysis procedure	0.18	(40)

Table V. Values of parameters to assess doses from soil ingested via hand-to-mouth activities.

Parameter	Distribution		Value	
C_{Cs134}	—		1	
EF	—		2.6	
BA[(1)]	—		0.053	
k_{Cs}[(2)] (10^{-9} Sv Bq^{-1})	—	Age category	^{134}Cs	^{137}Cs
		1 y	9.1	9.0
		5 y	5.2	4.8
SI[(3)] (10^{-6} kg day^{-1})	Lognormal	Age category	GM	GSD
		1 y	13.4	2.0
		5 y	9.4	2.6
A_{Cs137} (Bq kg^{-1})	Lognormal	GM		GSD
		866		1.6

(1) The same values were adopted for ^{134}Cs and ^{137}Cs.

(2) The values of k_{Cs} shown in this table were calculated using a bioaccessibility of 0.053.

(3) Amounts of soil ingested were determined based on an actual survey in a Japanese kindergarten[(57)].