

The transfer of fallout plutonium from paddy soil to rice: A field study in Japan

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ABSTRACT

Reported transfer factor (TF) values of Pu from paddy soil to rice are rather scarce, despite the radiotoxicity of Pu and the irreplaceable role of rice in Asian peoples' diets. Here, we conducted a field study to investigate the transfer of global fallout Pu from paddy soil to rice grain (hulled rice) in Japan. The ²⁴⁰Pu/²³⁹Pu atomic ratios in two rice grain samples out of 16 samples were determined and the ratios corresponded well with the global fallout value. The soil-to-rice TF_{Pu} in 12 Japanese prefectures ranged from 4.5×10^{-6} to 1.2×10^{-4} with a geometric mean of 3.3×10^{-5} . The TFs of rice obtained in this study were compatible to the TFs for the broad heading "cereals" compiled in the IAEA Technical Report Series No. 472. Weak correlations were found between the TF and the investigated soil characteristics such as soil pH and loss on ignition. Regarding the TFs for cerium (Ce) and thorium (Th) which are commonly considered as Pu analogues, we observed no significant correlations between the log(TF_{Pu}) and log(TF_{Ce}) or log(TF_{Pu}) and log(TF_{Th}). On the other hand, interestingly, a significantly positive correlation ($r = 0.795$, $p < 0.001$) was observed between log(TF_{Pu}) and log(TF_U). In view of the observed similarity of TF values for U and Pu from soil to rice, we thought that using the easy-to-measure TF_U to estimate TF_{Pu} from soil to rice might be suggested although the mechanism was unclear.

1. Introduction

In the terrestrial environment, radionuclides may be transferred to humans by inhalation and food ingestion and thus, there is a potential for internal radiological damage to the body. Plant uptake constitutes a vital link in the relocation of radionuclides from soil to humans. After being taken up by crops, the radionuclides can translocate towards different parts of the crops along with the minerals which are necessary for crop growth and reproduction (James et al., 2011).

Anthropogenic radioactive plutonium (Pu) has been released into the environment since the middle of the 20th century as a consequence of human nuclear activities such as the nuclear weapons tests, the occurrence of nuclear accidents, and operating nuclear reprocessing plants. Specifically, its main isotopes, ²³⁹Pu and ²⁴⁰Pu, have been categorized as "Group 1" radionuclides by the IAEA and they are among the most radiotoxic radionuclides (IAEA, 1973). They have long physical half-lives (24100 y for ²³⁹Pu and 6537 y for ²⁴⁰Pu) (Lariviere et al., 2006) and remain in the human body a long time due to their long

biological half-lives (e.g. 20 y in the liver and 50 y in bones) (Clarke et al., 1996); this gives rise to long-term cumulative exposure to human body once ingested through the food chain and local irradiation sources are formed (Sokolik et al., 2004). In this respect, the importance of studying the transfer of Pu from soil to crops should be highlighted.

The soil to plant transfer factor is a key factor employed in mathematical transfer models for estimating the radionuclides' concentration in crops or plants and assessing possible dose impacts to the human body (Mollah, 2014). Besides, due to the globalization of food production and trade, if a nuclear accident occurs, the possible local food contamination could rapidly present world-wide safety challenges to authorities (Emanuele et al., 2017). With the rapid boom in the nuclear power industry in Asian countries other than Japan, it would be instructive to investigate the transfer of Pu from soil to crops from the viewpoint of possible nuclear accidents as well as for use in long-term dose assessment of high-level radioactive waste disposal.

Among widely consumed crops, rice is a staple food crop in humid tropical regions worldwide and it represents a critical source of calories

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for more than half of the world's population (Uchida et al., 2009). However, despite the irreplaceable role of rice in human diets, studies on the transfer of Pu from paddy soil to rice are extremely scarce compared with those for other cereals such as wheat and barley; most studies on plant uptake of radionuclides have had a Western viewpoint and have been focused on the crops which grow in Western countries under their edaphic and climatic conditions (Adriano et al., 1981).

In 2010, the IAEA published the Technical Report Series No. 472 (TRS 472) that compiles literature transfer parameter data including the transfer factor (TF) of radionuclides from soil to crops (IAEA, 2010). No TF data, however, were presented specifically for Pu transfer from soil to rice. It might be possible to use the TF_{Pu} for cereals as a substitute for the TF_{Pu} for rice since data for other related cereals such as wheat and barley are much more abundant. However, this should be done with caution as the farming practices and field conditions for cereals and rice are significantly different. In waterlogged rice paddy fields, the soils are likely to be under anaerobic conditions due to the quick depletion of dissolved oxygen by the respiration of soil microorganisms and plant roots (Uchida et al., 2009). In such circumstances anaerobiosis occurs together with the reduction of mineral nutrients; in addition, the soil pH would also increase with the soil reducing condition (Uchida et al., 2009). The use of TF for cereals to replace that for rice has been a point of argument for another reason since the cultivation methods can affect the plant uptake of radionuclides greatly (IAEA, 2010). For this reason, newly derived TFs of Pu from paddy soil to rice is highly desired to clarify the uptake and to provide data to the existing TF_{Pu} database (Wang et al., 2015).

Additionally, many research studies aiming at investigating the transfer of Pu from soil to crops have been carried out with pot studies or under controlled experimental conditions using artificially contaminated soils and plants (Cataldo et al., 1975; Cline and Schreckhise, 1987; Schulz and Ruggieri, 1981). However, because the transfer of radionuclides from soil to crop generally depends on many environmental factors such as soil pH, organic matter contents, physicochemical forms of the radionuclides, species of crops, farming practices and climate conditions, the direct practical use of such transfer factors of Pu obtained in controlled conditions without taking into consideration the complex multifunctional natural processes should be reconsidered (Coughtrey and Thorne, 1983; Kozhakhyanov et al., 2014).

In this work, we investigated the transfer of fallout Pu from paddy soil to hulled rice grain samples collected throughout Japan. Possible influences of the soil characteristics on the transfer of Pu from soil to rice grain were evaluated. Furthermore, we discussed the feasibility of using easy-to-determine elements as analogues for Pu to study the transfer from soil to rice.

2. Material and methods

2.1. Sample collection

To investigate the transfer factor of Pu from paddy soil to rice, 15 rice samples (No. 1 to No. 15) together with the corresponding paddy soil samples were collected from 11 prefectures in Japan in 2005–2015. In addition, one rice sample (No. 16) and the corresponding soil were collected in Fukushima Prefecture at a place 55 km northwest of the Fukushima Daiichi Nuclear Power Plant (FDNPP) in October 2013, the third harvesting season after the nuclear accident. The sampling sites are indicated in Fig. 1. As a part of a ten-year project to study the transfer of elements in the biosphere, the soil characteristics as well as stable element concentrations in some of the rice and corresponding soil samples used in this study had been determined and partially reported previously (Tagami and Uchida, 2018; Uchida et al., 2007). The original data are provided in Tables A.1–A.3 in Appendix A, and the organic matter content in these soils was estimated during the dry-ashing step of the soil as loss on ignition (LOI); section 2.2 provides more details.

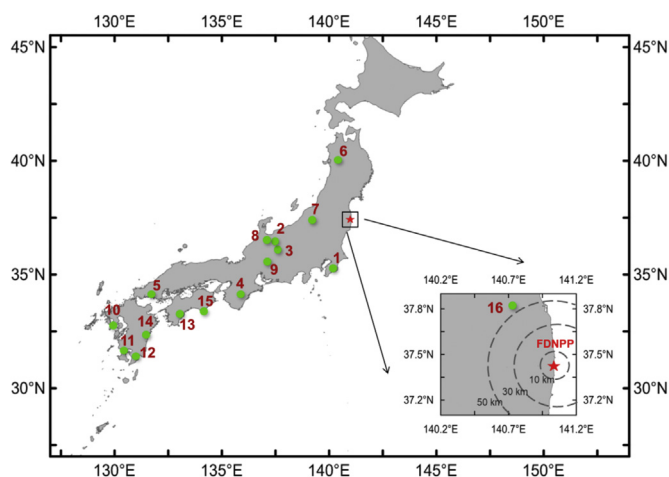


Fig. 1. Map showing the sampling sites in this study.

2.2. Chemical separation of Pu

For dried soil samples, 2 g soil sample was firstly calcined at 450 °C for 4 h to decompose organic matter before transferring to PTFE vessel. The LOI was calculated as the difference of soil weight before and after the calcination. After spiking with 0.57 pg ^{242}Pu tracer, 20 mL concentrated HNO_3 was added to leach Pu by heating at 180 °C for 4 h. After filtration, NaNO_2 was used at 40 °C for 30 min to adjust Pu to the tetravalent state. Then, Pu in the sample solution was purified with a well-established two-stage anion exchange chromatography method using successively AG 1X8 and AG MP-1M resins; the details have been described elsewhere (Bu et al., 2014). Finally, 0.8 mL 4% ultrapure HNO_3 containing 0.02 ng/mL ^{233}U as internal standard was added to reconstitute the sample in preparation for ICP-MS measurement.

For rice grain (hulled rice) samples, the sample pretreatment was more complicated than for the soil since a much larger amount of sample was required, which could bring about severe matrix effects. To mitigate the matrix effects as much as possible, we followed three strategies. First, the rice grain samples were combusted at 450 °C for 10 h to obtain roughly-volume-reduced samples. In order to improve the ashing efficiency, one rice sample (about 3–3.5 kg) was divided into 10–20 fractions (200–300 g each) during the dry-ashing and these fractions were combined after the first ashing. Then, an additional 10 h ashing was carried out at 450 °C. Typically, 15 g rice ash remained from 1 kg hulled rice after this dry ashing step. Second, as we had estimated that the analysis of approximately 15 g ash was required to obtain a sufficient ICP-MS instrumental signal, we separated each of the 15 g rice ash into five subsamples; each subsample had a weight of about 3 g. The subsamples were processed separately as below after adding the ^{242}Pu yield tracer. Third, we employed extra wet-ashing steps to process the 3 g rice ash sub-samples, which was accomplished by repeatedly digesting them with $\text{HNO}_3\text{-H}_2\text{O}_2$ at 200 °C, followed by filtrating the sample, and re-digesting the filtrated solution with $\text{HNO}_3\text{-H}_2\text{O}_2$ until the sample solution turned transparent. After further evaporating the solution to dryness and dissolving the residue with 50 mL 8 M HNO_3 , NaNO_2 was added for valence adjustment of Pu and the AG 1X8 resin was used for the first-stage separation. The 0.1 M $\text{NH}_4\text{I-8.5 M HCl}$ eluting solutions from the AG 1X8 resin carrying Pu were heated to dryness and reconstituted with 4 mL concentrated $\text{HCl/H}_2\text{O}_2$ solution. In this step, the five subsamples divided from the same sample were re-combined before being loaded onto the AG MP-1M resin. The subsequent preparation steps were the same as those for soil samples.

2.3. Pu measurement

To measure Pu isotopes, a high efficiency sample introduction

Table 1
Results of Pu in rice samples and corresponding soil samples.

Sample ID	Prefecture	^{239}Pu in soil mBq/g	$^{239+240}\text{Pu}$ in soil mBq/g	$^{240}\text{Pu}/^{239}\text{Pu}$ in soil	^{239}Pu in rice mBq/g	TF
1	Chiba	0.23 ± 0.01	0.39 ± 0.03	0.181 ± 0.013	$(5.3 \pm 1.5) \times 10^{-6}$	$(2.3 \pm 0.7) \times 10^{-5}$
2	Toyama	0.24 ± 0.01	0.39 ± 0.07	0.165 ± 0.012	$(4.9 \pm 1.7) \times 10^{-6}$	$(2.0 \pm 0.7) \times 10^{-5}$
3	Gifu	0.55 ± 0.03	0.92 ± 0.01	0.180 ± 0.014	$(2.5 \pm 0.9) \times 10^{-6}$	$(4.5 \pm 1.6) \times 10^{-6}$
4	Nara	0.12 ± 0.00	0.20 ± 0.02	0.179 ± 0.006	$(1.3 \pm 0.1) \times 10^{-5}$	$(1.1 \pm 0.1) \times 10^{-4}$
5	Yamaguchi	0.13 ± 0.01	0.21 ± 0.01	0.172 ± 0.024	$(9.0 \pm 6.5) \times 10^{-7}$	$(6.9 \pm 5.1) \times 10^{-6}$
6	Akita	0.56 ± 0.02	0.89 ± 0.03	0.162 ± 0.008	$(1.9 \pm 1.7) \times 10^{-5}$	$(3.4 \pm 3.0) \times 10^{-5}$
7	Niigata	0.28 ± 0.01	0.45 ± 0.00	0.166 ± 0.010	$(8.0 \pm 2.9) \times 10^{-6}$	$(2.9 \pm 1.0) \times 10^{-5}$
8	Toyama	0.22 ± 0.01	0.36 ± 0.01	0.171 ± 0.008	$(4.5 \pm 1.7) \times 10^{-6}$	$(2.0 \pm 0.8) \times 10^{-5}$
9	Gifu	0.36 ± 0.02	0.36 ± 0.02	0.182 ± 0.013	$(9.0 \pm 2.2) \times 10^{-6}$	$(2.5 \pm 0.6) \times 10^{-5}$
10	Nagasaki	0.15 ± 0.01	0.15 ± 0.01	0.160 ± 0.011	$(1.2 \pm 0.7) \times 10^{-5}$	$(8.0 \pm 4.3) \times 10^{-5}$
11	Kagoshima	0.05 ± 0.01	0.07 ± 0.01	0.160 ± 0.038	$(5.5 \pm 3.6) \times 10^{-6}$	$(1.2 \pm 0.8) \times 10^{-4}$
12	Kagoshima	0.14 ± 0.01	0.23 ± 0.01	0.174 ± 0.018	$(5.5 \pm 2.4) \times 10^{-6}$	$(3.9 \pm 1.7) \times 10^{-5}$
13	Kochi	0.28 ± 0.01	0.48 ± 0.02	0.184 ± 0.019	$(2.5 \pm 0.3) \times 10^{-5}$	$(8.6 \pm 1.1) \times 10^{-5}$
14	Miyazaki	0.10 ± 0.01	0.17 ± 0.02	0.196 ± 0.039	$(8.1 \pm 2.1) \times 10^{-6}$	$(8.2 \pm 2.3) \times 10^{-5}$
15	Kochi	0.10 ± 0.01	0.16 ± 0.01	0.169 ± 0.035	$(2.9 \pm 0.9) \times 10^{-6}$	$(3.0 \pm 1.0) \times 10^{-5}$
16	Fukushima	0.24 ± 0.01	0.39 ± 0.03	0.167 ± 0.012	$(6.6 \pm 2.5) \times 10^{-6}$	$(2.8 \pm 1.1) \times 10^{-5}$
				GM ^a	6.3×10^{-6}	3.3×10^{-5}
				GSD ^b	2.2	2.4

^a GM-Geometric mean.

^b GSD-Geometric standard deviation.

system Apex-Q was combined with the sector field ICP-MS (Element XR, Thermo Scientific, Bremen, Germany) to achieve high instrumental sensitivity and decrease the formation rate of $^{238}\text{UH}^+ / ^{238}\text{U}$. The detailed instrumental setups and parameters of this measurement system have been described elsewhere (Zheng, 2015). The ICP-MS was daily tuned with 0.02 ng/mL U standard solution to optimize the instrumental performance and all the measurements were completed in the low-resolution mode. As benefits from the abovementioned efforts, we achieved a typical instrumental sensitivity of 4×10^7 to 5×10^7 cps/ppb for ^{238}U in our study. Standard reference materials IAEA-soil-6 (soil) and IAEA-446 (seaweed) were analyzed together with the soil and rice samples for quality control of the analytical method (results are shown in Table A.4).

3. Results and discussion

3.1. Source of Pu in the present study

The Pu results in the 16 rice samples and the corresponding soil samples as well the deduced TF_{Pu} values are listed in Table 1.

In the Japanese terrestrial environment, there have been two prominent contamination sources of Pu, i.e. the global fallout as a result of nuclear weapon tests in the 20th century (Kelley et al., 1999) on a nationwide scale and the Nagasaki atomic bomb explosion in 1945 (Saito-Kokubu et al., 2008; Yamamoto et al., 1983a) on a local scale. Besides, it was found that a very small amount Pu release from the FDNPP nuclear accident in 2011 occurred compared to the Chernobyl accident (Zheng et al., 2012, 2013); and we considered that the FDNPP-sourced Pu was presented in an area close to the accident site (local scale deposition). These sources are distinguishable using their characteristic $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios, as the typical $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios of the global fallout, the Nagasaki atomic bomb explosion and the FDNPP accident are 0.166–0.194 (for 71–30 °N, Kelley et al., 1999), 0.028–0.037 (Yamamoto et al., 1983a) and 0.303–0.330 (Zheng et al., 2012), respectively.

As shown in Fig. 2, the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio of the 16 soil samples ranged from 0.160 to 0.196. These results were within the range of global fallout and were distinctive from the other two sources, suggesting that the global fallout is the dominant Pu source for these soils, even for the sample collected from Nagasaki (sample No.10) and the sample collected near the FDNPP (sample No.16). In addition, the $^{239+240}\text{Pu}$ activity concentration in these soil samples was from 0.07 mBq/g to 0.92 mBq/g with an average value of 0.33 mBq/g. These

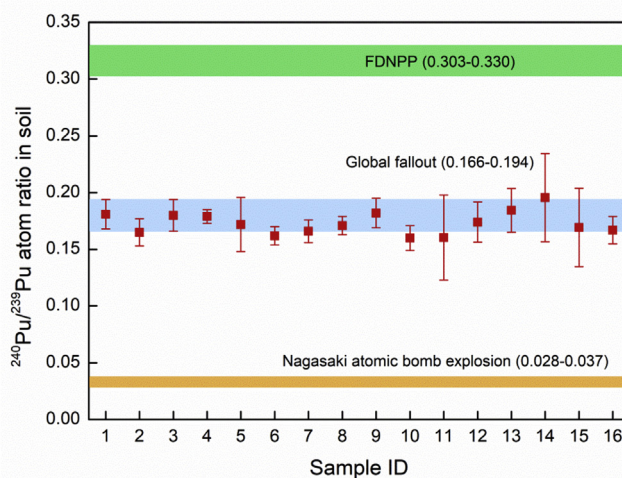


Fig. 2. The $^{240}\text{Pu}/^{239}\text{Pu}$ atomic ratios in Japanese soils and different sources. Data for the three sources were cited from Kelley et al. (1999), Yamamoto et al. (1983a), and Zheng et al. (2012).

values were in good agreement with those of other Japanese surface soils, e.g. 0.08–1.03 mBq/g reported by Yamamoto et al. (1983b), 0.15–4.31 mBq/g reported by Muramatsu et al. (2003) and 0.004–1.46 mBq/g reported by Yang et al. (2015), further suggesting the Pu in this study was derived from global fallout.

3.2. Soil-to-rice transfer factor (TF) of Pu

Generally, the transfer of a radionuclide from soil to a plant can be quantified by a dimensionless parameter characterizing the ratio of radionuclide concentration in the plant (mBq/g, dry weight) and its concentration (mBq/g, dry weight) in the corresponding soil. We note that there is no universal terminology concerning this parameter, and different terms such as “transfer factor (TF)”, “concentration factor (C_p)”, “concentration ratio (CR)” and “accumulation factor (F_v)” have been used by various researchers (Adriano et al., 1981; Caldwell et al., 2011; Kozhakhhanov et al., 2014; Nisbet and Lembrechts, 1990; Nishita, 1981; Sokolik et al., 2004). There are slight differences in the definitions of these parameters: TF, C_p , and F_v that have been recently employed by the IAEA should be gotten under equilibrium conditions, while equilibrium conditions are not a prerequisite for the CR.

However, not many researchers have considered that. We here employed the term “transfer factor (TF)” in this study, and it was calculated according to the following Eq. (1).

$$TF_{Pu} = \frac{C_{\text{rice}}(\text{Pu})}{C_{\text{soil}}(\text{Pu})} \quad (1)$$

where the TF_{Pu} is the transfer factor of Pu from soil to rice grain, the $C_{\text{rice}}(\text{Pu})$ stands for the ^{239}Pu concentration in the hulled rice grain (mBq/g, dry weight) and the $C_{\text{soil}}(\text{Pu})$ represents the ^{239}Pu concentration in the corresponding soil at harvest (mBq/g, dry weight). To facilitate our discussion, when other terms (C_f and F_v) were used in the cited references, we re-expressed them as “TF” in the text while when “concentration ratio” was used the cited papers, we also cited them as “concentration ratio” here.

Compared to the determination of Pu in soil samples, the analysis of Pu in rice grain samples was much more challenging due to the extremely low concentration of Pu in the rice grains and the matrix effects encountered during the ICP-MS measurements. In consequence, only the ^{239}Pu concentration was measured for the rice grain samples. The ^{239}Pu activity concentration in the hulled rice samples was from 9.0×10^{-7} mBq/g to 2.5×10^{-5} mBq/g as presented in Table 1. The concentration of Pu was far below the level regulated by the Japanese government (10 mBq/g for Pu and transuranic nuclides in vegetables, grains, etc.) (Food Safety Commission of Japan, 2011), illustrating that the radiological impact of Pu is negligible in Japanese rice, and it is the same for the rice samples collected in Nagasaki (sample No. 10) and in the contaminated area (northwest 55 km) around the FDNPP (sample No. 16). Based on the ^{239}Pu concentration in the rice samples and the corresponding soil samples, the TF_{Pu} from rice paddy soil to hulled rice grain was calculated (Table 1). For the 16 rice samples collected from 12 prefectures of Japan, the TF_{Pu} from paddy field soil to hulled rice varied from 4.5×10^{-6} to 1.2×10^{-4} , and the data distributed log-normally (Shapiro-Wilk test); the geometric mean of the TF_{Pu} was 3.3×10^{-5} .

Nishita (1981) studied the relative amount of uptake of ^{238}Pu and ^{239}Pu by wheat from soils contaminated by a soluble nitrate form of Pu. No appreciable difference in plant uptake between ^{238}Pu and ^{239}Pu was found in the pot experiment, indicating that isotopic fractionation was unlikely to occur in the transfer process of Pu from soil to wheat. Similarly, Schulz and Ruggieri (1981) systematically investigated the uptake and translocation of ^{238}Pu and $^{239+240}\text{Pu}$ from artificially contaminated soil to wheat plant and their results showed that both the uptake and translocation of Pu into various plant organs was identical for ^{238}Pu and $^{239+240}\text{Pu}$ when they were both added in the same nitrate form. However, Adriano et al. (1981) reported different concentration ratios of ^{238}Pu (10^{-5} – 10^{-4}) and $^{239+240}\text{Pu}$ (10^{-3} – 10^{-2}) from soil to rice grains for their greenhouse pot study using floodplain soil that was contaminated by the eluent from a nuclear facility. In our present study, ^{240}Pu was measured in two rice grain samples which had relatively high ^{239}Pu concentrations (sample No. 4 and No. 13). The obtained $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in No. 4 and No. 13 rice samples were 0.195 ± 0.049 and 0.181 ± 0.020 , respectively. To the best of our knowledge, this is the first report showing the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio that was from the global fallout in rice grain samples. Considering the measurement uncertainties, they agreed with the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios of the corresponding soils (0.179 ± 0.006 and 0.184 ± 0.019 , respectively) as listed in Table 1 and illustrated that the Pu in the rice grain was derived from the global fallout. Since the $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios in rice grain samples corresponded well with those in the soil samples, no appreciable difference between the investigated Pu isotopes (^{240}Pu and ^{239}Pu) was observed during the transfer of Pu from paddy soil to rice. Similar findings were obtained by Nishita (1981) and Schulz and Ruggieri (1981) that Pu isotopes (^{238}Pu vs. $^{239+240}\text{Pu}$, ^{238}Pu vs. ^{239}Pu) had identical transfer fates from soil to wheat plant. The difference with the findings of Adriano et al. (1981) was supposed to be due to the different forms of ^{238}Pu and $^{239+240}\text{Pu}$, which we discuss in

section 3.3. Therefore, the TF_{Pu} from paddy soil to rice was presumably identical to the values calculated with ^{239}Pu data in our study and thus could be used for intercomparison with TF_{Pu} values reported in the literature, many of which were calculated based on $^{239+240}\text{Pu}$.

3.3. Comparison of soil-to-rice TF_{Pu} with literature values

As aforementioned, despite the irreplaceable role of rice in peoples' diets (especially Asian peoples), studies on the transfer of Pu from paddy soil to hulled rice are scarce. As far as we know, there have been only three published papers including sporadic data on TF_{Pu} values from soil to rice. An overview of studies on the soil-to-rice TF_{Pu} is presented in Table A.5. Duffa et al. (2002) obtained 3 soil-to-rice TF_{Pu} values for soils in France. These paddy fields had been irrigated with river water that carried transuranic elements from a nuclear reprocessing plant. However, the $^{239+240}\text{Pu}$ in these soils was low (0.125–0.197 mBq/g) and no $^{240}\text{Pu}/^{239}\text{Pu}$ isotopic ratio data were available to confirm the Pu source although the $^{241}\text{Am}/^{239+240}\text{Pu}$ activity ratios in these samples were close to the characteristic ratio of global fallout. The TF_{Pu} ranged from 6.7×10^{-4} to 2.0×10^{-3} in that study. The reason for the nearly two orders of magnitude higher TF of Pu than our results was unclear, but we thought most likely due to the farming practices and soil characteristics. Also, the rice cultivar in their study was long-grain Koral rice while Japanese rice has short translucent grains, Japonica type. This difference in rice cultivars is another likely reason for the discrepancy in TF_{Pu} values between the study of Duffa et al. (2002) and our study.

Adriano et al. (1981) cultivated four types of rice in a greenhouse pot study. The soil used for the rice planting was collected from the floodplain of a stream receiving liquid effluents from a chemical separation facility at the US Department of Energy's Savannah River Site. Variation in the concentration ratios of Pu was insignificant among the plant types. However, the concentration ratio of ^{238}Pu (1.7×10^{-4}) from soil to hulled rice grain was approximately two orders of magnitude lower than that of $^{239+240}\text{Pu}$ (1.0×10^{-2}) for the same rice plant. Although the authors did not give any explanation for this difference, we assumed it might be a consequence of the different physical chemical forms of ^{238}Pu and $^{239+240}\text{Pu}$ in the liquid waste effluent which could lead to differences in their bioavailability, and that should also contribute to the higher concentration ratio of Pu in their study compared with our results where Pu originated from global fallout. As an illustration of this, it has been reported the uptake of nitrate form of Pu was 10–20 times greater than that of the oxide form of Pu for peas and barley (Cline and Schreckhise, 1987); more obviously, Adams et al. (1974) reported TF_{Pu} from soil to barley grown in nitrate-Pu contaminated soils was three orders of magnitude greater than that when barley was grown in oxide-Pu contaminated soils.

Zhang et al. (1989) conducted a field study to investigate the transfer of Pu from soil to several crops in residential areas around the Chinese nuclear test site in Xinjiang Province. They concluded that the mean ($n = 6$) TF_{Pu} from soil to rice was 2.0×10^{-3} in the investigated areas. However, as these authors mentioned, foliage contamination following the resuspension of dust served as a significant pathway of Pu transfer from soil to plant in that area. In arid, windy environments, airborne dust or soil particles could deposit or be intercepted by foliar surfaces followed by intake and translocation to other plant organs such as roots and seeds (Cataldo et al., 1975). Higher TF values of Pu and Am from soil to plants have been observed in such types of environments (e.g. the former US Nevada Test Site and Rocky Flats nuclear weapons production facility) where resuspension prevails (Dahlman et al., 1976). Therefore, it is reasonable to suspect that the much higher soil-to-rice TF_{Pu} value obtained by Zhang et al. (1989) when compared with our results was an overestimation of the root uptake. In summary, our field study showed that the TF_{Pu} values from Japanese rice paddy soil to rice were about two orders of magnitude lower than those sporadic data in other studies. These differences might be attributable to the different

Pu sources, physiochemical Pu forms, field and climate conditions, soil characteristics and different rice cultivars. In addition, the rice treatment (unhulled, hulled or polished) is also likely a relevant factor for these discrepancies, unfortunately, not all the reported studies on soil-to-rice TF_{Pu} has provided this crucial information.

In IAEA TRS 472, rice was categorized separately on account of its significant role in human diets (IAEA, 2010). However, although the transfer factors of many radionuclides from soil to rice have been compiled in this report, no transfer factor of Pu was included, most likely due to the lack of sufficient data. Instead, the transfer factors of Pu were only compiled for the broad heading “cereals” which covers several types of crops such as barley and wheat. Researchers might consider using the TF_{Pu} of cereals as a substitute for the TF_{Pu} of rice in cases where related data were lacking. Nevertheless, before doing that, it should be noted that since the field conditions of upland crops and rice are distinct, the bioavailability of Pu as well as plant uptake ability in these situations *presumably* vary, in which case separate consideration of the TF_{Pu} for rice and other upland cereals might be called for. For this reason, enhancing the soil-to-rice TF_{Pu} data with newly derived values is crucial not only to enlarge the database of TF_{Pu} but also to verify whether the TF_{Pu} for rice which grows in unique field conditions could be assigned to the TF_{Pu} for the general category of cereals. In the IAEA TRS 472, the TF_{Pu} for cereals showed a wide range that covers four orders of magnitude, i.e. from 2.0×10^{-7} to 1.1×10^{-3} with a geometric mean of 9.5×10^{-6} . Compared with that, the paddy soil-to-hulled rice TF_{Pu} obtained in our field study varied from 4.5×10^{-6} to 1.2×10^{-4} with the mean value of 3.3×10^{-5} , which was a narrower range than that for cereals as illustrated in Fig. 3. However, if the sporadic data in the literature (Table A.5) were considered as well regardless of the Pu sources and the site-specific diversities, the total soil-to-rice TF_{Pu} varied from 4.5×10^{-6} to 1.5×10^{-2} ($n = 29$). This range is slightly wider but still close to that for the soil-to-cereals TF_{Pu} . Fig. 3 shows the TF_{Pu} values for cereals and rice. Supported by these comparison results, we conclude that the TF_{Pu} for rice could be assigned to the TF_{Pu} for the cereals; and in cases where no or few site-specific data are available, the soil-to-cereals TF_{Pu} could be adopted to represent the soil-to-rice TF_{Pu} .

3.4. Relationship between the transfer of Pu and soil characteristics and element contents

Since the concentration of Pu in rice and the TF_{Pu} are log-normally

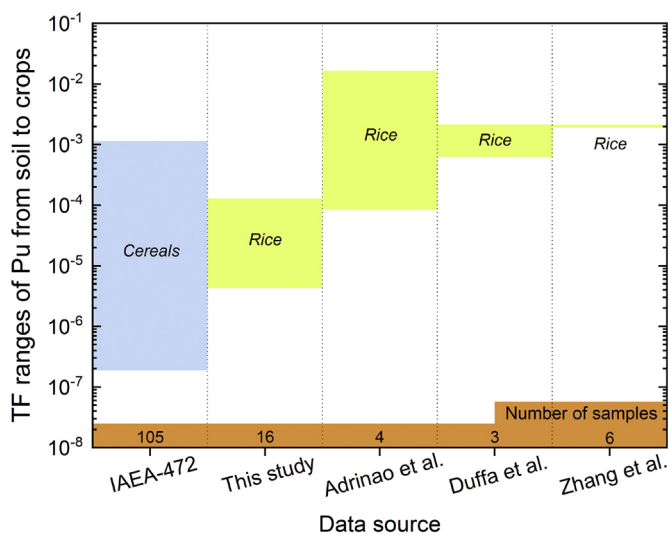


Fig. 3. Ranges of the TF_{Pu} values from soil to crops (cereals and rice). Data for the cereals were cited from IAEA (2010). Data for the rice were cited from Adriano et al. (1981), Duffa et al. (2002) and Zhang et al. (1989).

distributed (Shapiro-Wilk test), we calculated the Pearson correlation coefficients between the log(Pu concentration in rice) as well as the log(TF_{Pu}) and the various soil characteristics and some concentrations of common trace elements in soil to study the factors that are likely to influence the transfer of Pu from soil to rice as in Table A.6. The original data for the related terms are provided in Table A.1. In general, both the logarithmic Pu concentration in rice and the log(TF_{Pu}) showed no significant correlation with the investigated soil parameters, while both of them were moderately correlated with the Ce concentration in soil at the 0.05 confidence level. Hanušik (1994) quantified the effect of soil parameters on the soil-to-plant transfer of Pu, Np, Am and Cm using selected data of other studies from the literature. However, the author found that the correlations between the transfer factors of Pu from soil to wheat and the same soil parameters (e.g. organic matter, CEC, and pH) were opposite in two of their selected studies. In the pot study conducted by Vyas and Mistry (1981), increasing the organic matter content in soil led to less Pu uptake by plants. However, based on our present data, no significant correlation was found between the soil-to-rice transfer of Pu and soil LOI. These poor correlations indicated that the influences of soil characteristics on the transfer of Pu were complex and involved multifactorial processes.

3.5. Possible analogue for Pu in the transfer from paddy soil to hulled rice

In situations where no data or few data are available to evaluate the transfer of a radionuclide, its analogue may be used to provide relevant information in screen models on the environmental behavior of the radionuclide of interest (IAEA, 2010). Finding a proper analogue contributes to better, more reliable descriptions of related research (Varga et al., 2009). It is also advantageous to find some easy-to-measure elements as analogues for elements that are difficult to measure. For this purpose, we analyzed the correlations between Pu concentration and concentrations of some other elements in rice and the results are shown in Table A.7 and the correlations between their TFs are shown in Table A.8. We note that as the concentrations of Pu and the investigated elements and their TF values were log-normally distributed, the correlation coefficients were calculated based on their logarithmic values. The original data of the element concentrations in rice and their corresponding TF values are provided in Table A.2 and Table A.3. The logarithmic Pu concentration in rice was significantly correlated with the logarithmic concentrations of Al, Fe, La, Ce, Nd, Sm, Th and U in rice (Table A.7), even with the elements that are basic for plant growth (Fe, Al). Similar findings were reported by Caldwell et al. (2011) who found significant correlations between Pu concentration and Al, Ce, Fe, Mn, Sm concentrations in several types of vegetation grown in a contaminated site. Furthermore, as illustrated in Table A.8, the log(TF_{Pu}) was positively correlated with those of Al, Fe, La, Nd, Sm and U ($p < 0.05$), among which the correlations with Al and U were significant at the 0.01 confidence level.

The lanthanides are often considered as analogues for the trivalent actinides (Krauskopf, 1986; Varga et al., 2009). Pu (III) could also be present under reducing conditions (Choppin, 2006, 2007), providing the possibility of using trivalent lanthanides as an analogue for Pu in such circumstances. Also, the 4+ oxidation state of cerium (Ce), neighboring the 3+ oxidation state of most lanthanides, is thought to be presumable to act as an analogue for tetravalent Pu (Varga et al., 2009). However, in our study that focused on the transfer process of Pu from soil to rice, a weak correlation ($r = 0.418$, $p = 0.201$) was observed between log(TF_{Pu}) and log(TF_{Ce}) (Table A.8). For comparison, the correlations between log(TF_{Pu}) and those of the other lanthanides La, Nd and Sm were stronger and more significant ($p < 0.05$) although these elements are only present in the trivalent state. On account of this, the suitability of using these lanthanides as analogues for Pu transfer from the waterlogged paddy field soil to rice deserves further systematic study.

Thorium (Th) is thought to be the most useful and a common

analogue for Pu in the tetravalent state (Choppin, 2006, 2007; Krauskopf, 1986; Varga et al., 2009). Both Th and Pu can exist in the stable form of a dioxide, and the dissociated hydroxides are the principal dissolved species in equilibrium with them (Krauskopf, 1986). Moreover, stability constants of Th and Pu for complexes with common ligands are similar, while Th(IV) has somewhat weaker complexation and hydrolytic than Pu(IV) (Choppin, 2006; Krauskopf, 1986). The logarithmic Pu concentration in rice in our study showed a significant correlation with the logarithmic Th concentration in rice ($r = 0.623$, $p < 0.05$, Table A.7), while a poor correlation was observed between $\log(\text{TF}_{\text{Pu}})$ and $\log(\text{TF}_{\text{Th}})$ ($r = 0.481$, $p = 0.070$). Since both Pu and Th are not essential nutrient elements for rice growth, we speculate that the most probable reason for the inconsistent trend of TF_{Pu} and TF_{Th} is that the plant bioavailability of Pu and Th differed from each other in the paddy soils, leading to the poor correlation of TFs even though their concentrations in rice correlated very well. Specifically, Th is a naturally existing radionuclide in soil minerals (which are hard to decompose) while anthropogenic fallout Pu was deposited and has bound to soil surfaces. The different existing forms for Th and Pu are likely to have large influences on their bioavailability because not all the Th and Pu present in the soil are available for plant uptake as generally only water soluble and exchangeable fractions of an element are regarded as bioavailable (Kennedy et al., 1997; Zhang et al., 2010). However, the bioavailability of the elements was not considered when using the conventional transfer factor, which requires further investigation in the future. If significant correlations between the bioavailable fraction of Pu and Th could be confirmed, the use of Th as an analogue for Pu in the soil-to-rice transfer process would be more convincing.

Interestingly, both the logarithmic Pu concentration in rice and $\log(\text{TF}_{\text{Pu}})$ were significantly correlated with those of U, respectively, as shown in Table A.7 and Table A.8. As illustrated in Fig. 4, the correlation between $\log(\text{TF}_{\text{Pu}})$ and $\log(\text{TF}_{\text{U}})$ was significant ($\log(\text{TF}_{\text{Pu}}) = 0.847 \cdot \log(\text{TF}_{\text{U}}) - 1.050$, $r = 0.795$, $p < 0.001$). Since the slope of the $\log(\text{TF}_{\text{Pu}})$ and $\log(\text{TF}_{\text{U}})$ fitting curve was 0.847, the TF values of Pu and U from soil to rice are thought to be close to each other thus further suggesting the possible similarity between Pu and U in their transfer processes from paddy soil to rice.

To explain this similarity, we considered that in oxic surface waters, the hexavalent state is the dominant valence state of U, while under anoxic conditions, U(IV) is presumably the major form (Markich, 2002). In the long-term waterlogged rice paddy soils, oxygen could be quickly depleted by microbiological activities and that would lead to anoxic conditions. Thus, comparable environmental behavior of the

two actinides U and Pu is possible under this specific field condition if they both present in the tetravalent state during their uptake by rice plant roots. It is generally known that both the U(IV) and Pu(IV) are particle reactive. Therefore, both the Pu and U in the waterlogged anoxic paddy fields were presumably largely bounded to the soil particles in this soil-water system. Although the mechanisms of the root uptake dynamics for U and Pu from soil solution are not well understood, evidence has indicated that the uptake process of U and Pu could be facilitated by the complexation with ligands that increase their solubility (Neu et al., 2002; Vandenhove et al., 2007). As the stability constants of these two actinides for many complexation reactions (e.g. with HPO_4^{2-} , DTPA etc.) are comparable (Brown et al., 2012; Choppin, 2007; Lemire and Tremaine, 1980), similar uptake of U and Pu from soil solution by rice plant root might be expected. In the IAEA TRS 472, a log equation $\log(\text{TF}) = \log(\text{B}) - \log(\text{K}_d)$ where B is the radionuclide plant to soil solution ratio ($\text{Bq} \cdot \text{kg}^{-1}$ in dry weight plant tissue/ $\text{Bq} \cdot \text{L}^{-1}$ in soil solution) and K_d is the distribution coefficient of radionuclide ($\text{Bq} \cdot \text{kg}^{-1}$ in solid phase/ $\text{Bq} \cdot \text{L}^{-1}$ in liquid phase) has been successfully used to predict radiocaesium and radiostrontium transfer factors, while so far its applicability for actinides remains inconclusive. However, if we assume the rationality of this equation for Pu and U, the similarity between TF_{Pu} and TF_{U} could be explained. In that case, the comparable TFs of U and Pu could presumably be attributed to both the similarity in their concentration distribution between soil and soil solution and the likely similar uptake of these actinides from soil solution by the rice plant root. However, we must note again that the above assumption is only raised to give probable explanation for the similarity of the TF values. More exactly, the TF_{U} values are slightly higher than TF_{Pu} values for all the investigated rice samples, suggesting the transfer of U might be somewhat more significant, or Pu is less mobile in the paddy soils. Although a detailed investigation of the transfer mechanism is beyond the scope of the present work, we must suggest that further studies on the bioavailability of these elements are quite necessary to test, for example, whether the water soluble Pu and U in these soils are related to the rice uptake. Nevertheless, within the scope of this study, since the soil-to-rice TF_{U} values are much easier to measure, the TF_{U} may be a good indicator to estimate soil-to-rice TF_{Pu} levels.

4. Conclusions

In this study we investigated the transfer of fallout Pu from Japanese paddy soil to rice grain (hulled rice) in samples collected from 12 prefectures throughout Japan. The Pu concentrations in hulled rice samples were far below the regulation level set by the Japanese government. The TF_{Pu} values varied from 4.5×10^{-6} to 1.2×10^{-4} with a geometric mean of 3.3×10^{-5} for the 16 hulled rice samples. The TF_{Pu} of rice could be assigned into the TF values of cereals compiled in IAEA TRS 472 since the TF_{Pu} values of rice obtained in this study were distributed more narrowly. Weak correlations were found between the TF_{Pu} and the investigated soil parameters, such as soil pH, which might indicate their multifactorial and complex influences on the transfer process. Although Ce and Th are commonly considered as chemical analogues of Pu in environmental studies, no significant correlation was observed between the TF_{Pu} and TFs of Ce and Th. Interestingly, significantly positive correlations were observed between $\log(\text{TF}_{\text{Pu}})$ and $\log(\text{TF}_{\text{U}})$ as can be expressed by the equation $\log(\text{TF}_{\text{Pu}}) = 0.847 \cdot \log(\text{TF}_{\text{U}}) - 1.050$ ($r = 0.795$, $p < 0.001$). This might be attributed to the similar behavior of Pu(IV) and U(IV) as a consequence of the anoxic paddy field conditions. These results also highlighted the necessity to further study the bioavailability of Pu and those elements in the paddy soils that are closely relevant to their uptake by plants. However, in view of this significant correlation together with the similar TF values of U and Pu from soil to rice observed in this study, the easy-to-measure TF_{U} may be useful for rough estimation of TF_{Pu} levels for soil to hulled rice.

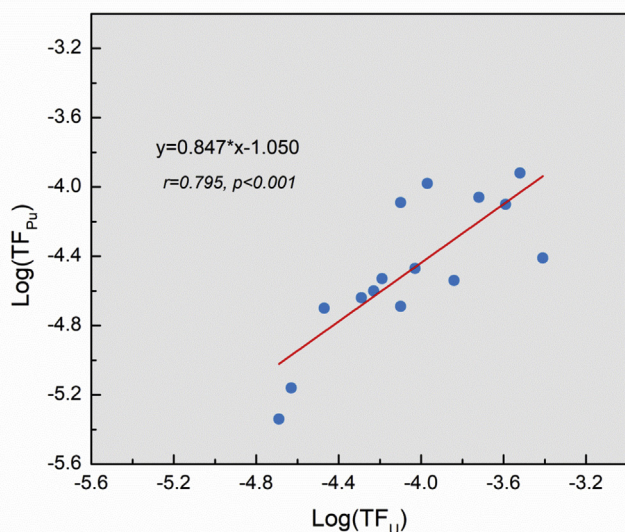


Fig. 4. Correlation between $\log(\text{TF}_{\text{Pu}})$ and $\log(\text{TF}_{\text{U}})$ from soil to rice grain.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvrad.2018.10.010>.

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