Transfer Factors of Radionuclides from Soil to Rice and Wheat Collected in Japan

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Abstract. Obtaining local soil-to-crop transfer factor (TF) is important because climates, soil types and vegetations influence on the factor. In this study, analyses of stable isotopes and natural radioisotopes in rice and wheat grains and the associated soils collected throughout Japan were carried out in order to obtain TF under equilibrium conditions. The studied samples had been collected for the measurement of global fallout ^{137}Cs and ^{90}Sr at National Institute of Agro-Environmental Sciences. Inductively coupled plasma mass spectrometry and inductively coupled plasma optical emission spectrometry were used to measure about 30 elements, including Cs, Sr, Th and U. The geometric-mean-TFs of Cs, Sr, Th and U for polished rice collected in Akita, Japan, were 0.0024, 0.0037, 0.0001 and 0.000053, respectively. The results indicated that TF-Cs was usually lower than TF-^{137}Cs (geometric mean=0.0033). Possibly, the physico-chemical forms of stable Cs differed from those of global fallout ^{137}Cs in the soil samples, that is, a part of the stable Cs was strongly fixed in the soil particles, while ^{137}Cs was more mobile in the soil. However, the difference of TF-Cs and -^{137}Cs was small, thus, the TF-Cs can be used for long-term transfer of ^{137}Cs in the environment. The methodology would be applied to the other elements, though physico-chemical forms of natural and non-natural isotopes should be clarified.

1. Introduction

From a viewpoint of precise long-term radiological assessment, it is necessary to obtain the variations of transfer parameters that are used in mathematical models. Ingestion of crops and livestock products contaminated with radionuclides is the most important pathway through which the nuclides from routine releases are taken into the human body [1]. Eating habits, however, differs by countries. For example, in European and North American countries, livestock products, i.e. meat, eggs and milk, make a big contribution, whereas, in Asian and South American countries, crops including cereals and vegetables are the main contributors [2].

Among the parameters used in the model, a soil-to-crop transfer factor (TF) is the key parameter that directly affects the internal dose assessment for the ingestion pathway. The TF could differ by areas due to different climates, soil types and vegetations, therefore, local TFs should be observed. Previously, Uchida and Okabayashi surveyed TF values for various crops collected in Japan and in the world [3]. The reported TF data obtained under natural conditions were limited especially in Asian and South American countries.

Obtaining the TFs of radionuclides under equilibrium conditions is indeed necessary for assessing the environmental transfer of the radionuclides from routine releases. TFs observed under natural conditions using global fallout ^{137}Cs and ^{90}Sr should be fitted to this purpose. In recent years, however, the concentrations of ^{137}Cs and ^{90}Sr in crops are found to be close to the lower detection limits of radiation measurements. Thus, in this study, we observed the TFs of stable Cs and Sr from paddy soils to rice and from upland soils to wheat to obtain the local TFs alternatively to TFs of ^{137}Cs and ^{90}Sr. Because inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectrometry (ICP-OES) were applied to measure stable Cs and Sr, about 30 elements including Th and U, were also measured in the soil and crop samples. The studied samples had been collected for global fallout ^{137}Cs and ^{90}Sr measurements at National Institute of Agro-Environmental Sciences (NIAES), Japan, so that the TF-Cs was compared with the TF-^{137}Cs.

We particularly focused on the TFs for Cs from paddy soil to rice grains, because rice plants are grown under waterlogged conditions. Besides, rice is staple diet for the people who live in Southeast Asian countries including Japan. The TFs for wheat was also compared with rice, because both crops are classified into cereals and there are many TFs for wheat in Europe and North America.

2. Materials and methods

2.1. Soil, rice and wheat samples

Paddy soils (plowed soil layer: 0 - 20 cm) have been collected nationwide from 12 - 15 sampling sites and rice plants grown on these soils have also been collected in the harvesting season. Rice varieties grown traditionally in Japan are classified as short grain-types. Wheat plants and the associated soils were collected in Akita Prefecture in 1994. All the samples were collected by NIAES. Each rice sample was processed into brown rice (with bran) and polished rice (without bran). The average weight ratio of the polished rice to the brown rice was 0.9 (90% yield). The wheat sample was husked. The ^{137}Cs and ^{90}Sr activities were measured by Komamura and her co-workers and the results had already been reported in elsewhere [4-6].

2.2. Analytical methods for stable isotopes and natural radioisotopes

The soil and crop samples were ground up into fine power. Then, 100 mg of the soil and 500 mg of the crop samples were digested with mineral acids using a microwave digester (CEM, MARS5). Each sample was evaporated to dryness and the residue was dissolved in 20 mL of 2% HNO_3. All the acids used were ultra-pure analytical grade (Tama Chemicals, AA-100). About 30 elements, including Cs, Sr, Th and U, in both crop and soil samples were measured by ICP-MS (Agilent 7500, Yokogawa) and ICP-OES (VISTA-Pro, Seiko) after diluting the solution to a suitable concentration.

Standard solutions of known concentrations, 0-100 ng/mL for ICP-MS and 0-20 μg/mL for ICP-OES, were prepared by diluting a multi-element standard solution (XSTC-1, -7, -21, and -355, SPEX Ind. Inc.) with 2% HNO_3. For ICP-MS, In, Rh, Tl or Bi was used as an internal standard.

3. Results and discussion

The TFs were calculated from the concentrations of the radioactive or natural isotope in both crop and soil samples. The TF is defined as the concentration of an isotope in crop (in Bq/kg or mg/kg dry weight (DW)) divided by the concentration of the isotope in soil (in Bq/kg or mg/kg DW).

3.1. Survey of reported TFs of ^{137}Cs and ^{90}Sr

Figure 1 shows example results of TF-^{137}Cs for polished rice collected in Akita reported by Komamura *et al.* [6]. Although samples have been collected since 1961, we assumed that the soil-to-rice plant system was not in equilibrium before 1987. Thus, the ^{137}Cs-TF data obtained from 12 - 15 sites nationwide between 1987 and 2001 were used for the following analysis. Direct contamination on the rice grains was considered negligible during the period, because the radioactive fallout levels observed over the last decade were extremely low compared with the total fallout level.

The TF-^{137}Cs for polished rice collected in 1987-2001 had a log-normal probability distribution of TFs (the data not shown). The variation of TF was about three orders of magnitude and the median was 0.002. The maximum TF was 0.029 and the minimum was 0.00005, giving a ratio of maximum TF to minimum TF of 550. The geometric and arithmetic means were 0.0019 and 0.0033, respectively.

3.1.1. TFs for brown rice and polished rice

In Table I, TFs of ^{137}Cs and ^{90}Sr those were calculated for the present study from the reported data [4-6] are listed. The maximum TF-^{137}Cs values for brown rice and polished rice were 0.02 and 0.008, respectively, while the minimum-TFs were 0.0009 (brown rice) and 0.0002 (polished rice). The

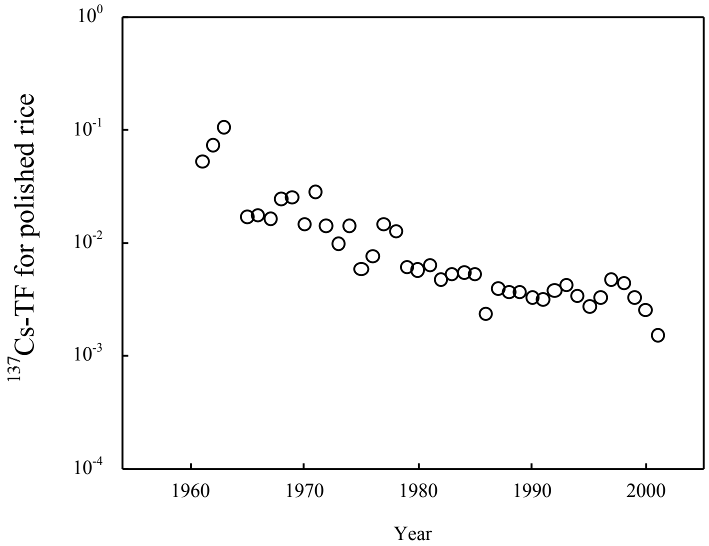

FIG. 1. Variations of ^{137}Cs-TFs for polished rice collected in Akita as a function of time.

Table I. Concentrations of ^{137}Cs and ^{90}Sr for brown rice, polished rice and husked wheat and soil samples and the TFs of ^{137}Cs and ^{90}Sr collected in Japan in 1990's.

	TF-^{137}Cs			TF-^{90}Sr		
	Brown rice (1999)	Polished rice (1999)	Husked wheat (1994)	Brown rice (1990)	Polished rice (1990)	Husked wheat (1994)
Max	0.031	0.029	0.0056	0.035	0.023	0.20
Min	0.0013	0.00032	0.00066	0.0044	0.0019	0.020
Geomean	0.0043	0.0021	0.0029	0.015	0.0059	0.083
Number of samples	12	15	10	14	14	10

geometric-mean-TFs were 0.0026 (brown rice) and 0.0011 (polished rice) for the samples collected in 1999. The geometric-mean-TFs for ^{90}Sr were 0.015 for brown rice and 0.0059 for polished rice.

The distributions of ^{137}Cs and ^{90}Sr in the polished rice samples were calculated. The average distribution ratios in the polished rice to brown rice were 0.35 for ^{137}Cs and 0.36 for ^{90}Sr, indicating that most of the activity from these two radionuclides was associated with the bran. About 35% of each nuclide was distributed in the polished rice.

The variations of TFs-^{137}Cs for polished rice obtained from 1987 to 2001 at 8 sampling points were studied using the Komamura's data [4-6]. The medians were 0.0025 for Sapporo, 0.0033 for Akita, 0.0015 Joetsu, 0.0036 for Morioka, 0.0043 for Sendai, 0.0014 for Mito, 0.0013 for Tsukuba and 0.0035 for Tsukushino. From these results, it was clear that the TF values observed at these sampling sites were almost the same. At each site, the variation of TFs, that is, the ratios of the maximum to the minimum TF, were very small, within the range of two orders of magnitude.

3.1.2. TFs for husked wheat

For TFs of ^{137}Cs and ^{90}Sr calculation, the wheat samples collected in 1994 from 10 sampling points throughout Japan were used [4, 5]. The results for husked wheat are also listed in the Table I. The geometric-mean-TFs ^{137}Cs and ^{90}Sr were 0.0029 and 0.083, respectively. The TFs of ^{137}Cs and ^{90}Sr for husked wheat and brown rice were in the same order of magnitude, however, the TFs for the polished rice were one order of magnitude lower than those for brown rice.

IAEA [7] reported expected TF values for cereals collected in the temperate environment and the TF-Cs and TF-Sr were 0.02-0.083 and 0.02-0.21, respectively. The expected values were slightly higher than the TFs of ^{137}Cs and ^{90}Sr calculated in this study. Probably, climates, soil types and vegetations influence the parameters. Although our data are limited, further study is necessary to collect local TF values for precise radiological assessment.

3.2. TFs of stable isotopes and natural radioisotopes for rice and wheat

3.2.1. TFs for polished rice

The TFs of stable isotopes and natural radioisotopes in the rice plants collected in Akita was observed because the TFs of ^{137}Cs and ^{90}Sr showed the narrowest year-to-year variation. The results are shown in Table II. Although 10 samples were measured (one sample for each sampling year), some elements were not measured in several samples due to their low concentrations in polished rice. It is necessary to adjust the ICP-MS and the ICP-OES operation conditions to measure more elements at ultra low-levels.

Among the measured elements, TFs higher than 0.1 were observed for Cu, Zn, Rb and Cd. Cupper and zinc are essential elements for plants so that their TF would be high. For Rb, it might act as K, which

Table II. Transfer factors of elements from paddy soil to polished rice collected in 1986 – 1995.

Element	n	Min	Max	Geomean	Element	n	Min	Max	Geomean
Li	7	2.30E-05	3.70E-04	1.00E-04	La	9	1.30E-05	5.40E-04	8.30E-05
Be	7	1.50E-05	6.20E-04	1.00E-04	Ce	10	6.20E-06	5.20E-04	4.60E-05
Cr	10	2.90E-03	3.40E-02	1.20E-02	Pr	6	4.00E-05	4.30E-04	1.00E-04
Co	10	3.50E-04	1.50E-03	8.10E-04	Nd	10	8.90E-06	4.20E-04	4.60E-05
Ni	10	1.00E-02	5.80E-02	2.90E-02	Sm	8	1.40E-05	3.80E-04	1.00E-04
Cu	10	1.00E-01	3.60E-01	1.30E-01	Eu	7	3.20E-04	8.00E-03	2.80E-03
Zn	10	9.40E-02	1.70E-01	1.20E-01	Gd	9	4.00E-05	3.50E-04	1.00E-04
Ga	10	1.90E-04	5.50E-04	3.10E-04	Dy	9	1.40E-05	3.30E-04	6.00E-05
As	10	4.60E-03	2.10E-02	8.00E-03	Er	8	1.30E-05	3.20E-04	8.00E-05
Se	10	2.00E-02	6.30E-02	2.70E-02	Tm	7	6.10E-06	6.30E-04	1.30E-04
Rb	10	1.10E-01	3.00E-01	1.60E-01	Yb	9	6.40E-06	2.50E-04	5.00E-05
Sr	8	5.40E-05	3.20E-02	3.70E-03	Lu	8	3.20E-05	6.00E-04	1.50E-04
Y	9	7.40E-06	4.60E-04	8.90E-05	Pb	10	1.30E-04	1.00E-02	6.00E-04
Cd	10	9.60E-02	4.90E-01	2.30E-01	Th	8	6.80E-06	7.50E-04	1.00E-04
Cs	10	1.10E-03	3.50E-03	2.40E-03	U	10	3.70E-06	2.80E-04	5.30E-05

is one of the essential elements. TFs between 0.1-0.001 were found for Cr, Ni, As, Se, Sr, Cs and Eu; for example, the TFs of Cs and Sr were 0.0024 and 0.0037, respectively. It was reported for mushrooms that the uptake mechanisms of K and Cs were independent [8]. Higher plant also can differentiate K from Cs, and thus, the TF of Cs was lower than that of Rb. The TFs of other elements were lower than 0.001 for the polished rice, indeed, TFs of Th and U were 0.0001 and 0.000053, respectively.

The TF values of ^{137}Cs and stable Cs for polished rice were compared (Fig. 2). The values were close from each other but the TF-^{137}Cs was usually higher than the TF-Cs. Tsukada *et al.* [9] also reported that TF-^{137}Cs was approximately 3 times higher than TF-Cs. The phenomena could be explained as follows; fallout ^{137}Cs was more mobile and more easily adsorbed by plants than stable Cs in the soil.

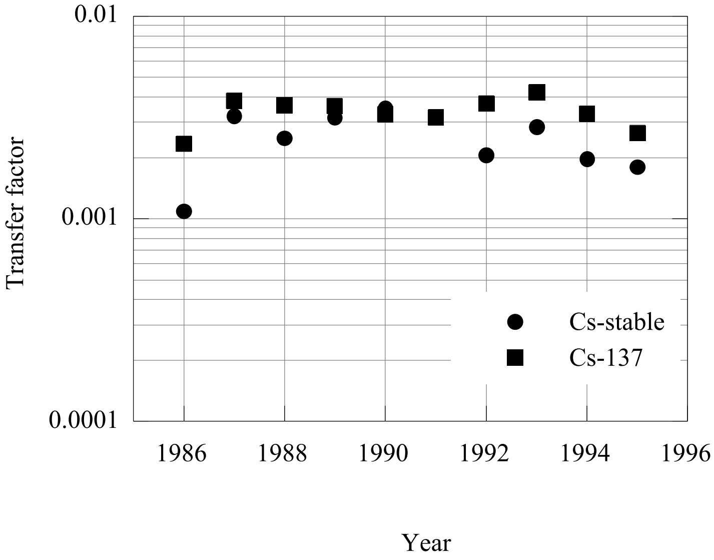

FIG. 2. Transfer factors of ^{137}Cs and stable Cs for polished rice.

Possibly, a part of stable Cs is found in soil structure that is hard to replace with ^{137}Cs so that these isotopes have not reach to an equilibrium condition. However, the TF-Cs can be used for long-term transfer of ^{137}Cs in the environment. For TF-Sr and TF-^{90}Sr, the difference was also small, but the TF-^{90}Sr was higher than TF-Sr. The same result was reported by Komamura *et al.* [10]. Somehow, the TFs of the other stable isotopes and natural radioisotopes would differ from the TFs of added radioisotopes due to their different physico-chemical forms in natural environment. However, the TFs of natural isotopes can be applied in the mathematical models for long-term radiological assessment, although it is necessary to clarify why the TF difference occurred between added radioisotopes and natural isotopes.

3.2.2. TF for husked wheat

As a preliminary test, one wheat sample was measured the concentrations of stable isotopes and natural radioisotopes by ICP-MS and ICP-OES to survey the TF levels (Table III). The TFs of stable Sr for wheat was higher than polished rice as found for TF-^{90}Sr, while the TF-Cs was in the same level with that for rice in this experiment. We are collecting the TF data for wheat and the results will be discussed in the near future.

Table III. Transfer Factor of elements from upland soil to wheat (n=1).

Element	TF	Element	TF	Element	TF	Element	TF
Li	5.70E-05	As	2.30E-04	La	2.00E-04	Er	8.10E-06
Be	2.20E-03	Se	5.80E-02	Ce	1.30E-04	Tm	6.60E-05
Cr	2.90E-03	Rb	1.90E-01	Pr	2.00E-04	Yb	-
Co	7.70E-04	Sr	1.70E-02	Nd	1.70E-04	Lu	1.30E-05
Ni	2.00E-02	Y	-	Sm	9.30E-05	Pb	6.20E-03
Cu	1.50E-01	Cd	4.00E-01	Eu	3.80E-04	Th	2.60E-04
Zn	3.20E-01	Cs	3.80E-03	Gd	8.10E-05	U	5.70E-05
Ga	5.20E-04	Ba	2.10E-02	Dy	-		

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