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Transfer factors of radionuclides and stable elements from soil to rice and wheat

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Abstract. It is important to obtain local transfer factors (TFs) of long-lived radionuclides for assessment of radioactive waste disposal, because climates, soil types and vegetation can affect TFs. Global fallout ^{90}Sr and ^{137}Cs are good tracers to obtain the TFs under natural conditions, however, their radioactivity levels in plants are extremely low. To close the gap, analyses of stable isotopes and natural radioisotopes in rice and wheat grains and their associated soils collected throughout Japan were carried out in order to obtain TFs under equilibrium conditions. We focused on rice and wheat, because the consumption of cereals is very high in Japan and other Asian countries. About 40-50 elements such as Cs, Sr, Th and U in plant and soil samples were measured by ICP-MS and ICP-OES. From the data, TFs of stable elements were calculated; among the values, TFs of Cs and Sr for rice and wheat were compared with the TFs of ^{137}Cs and ^{90}Sr. Using the elements' concentrations data, we proposed a 'Reference rice' (brown rice and polished rice) and a 'Reference paddy soil' to provide elemental composition data which can serve as reference values in mathematical transfer models and which allows the comparison of data for other crops.

1. INTRODUCTION

It is necessary to obtain the variations of transfer parameters that are used in mathematical models for a precise long-term radiological assessment. Among the parameters used in these models, the soil-to-crop transfer factor (TF) is a key parameter that directly affects the internal dose assessment for the ingestion pathway. As the TF can differ by areas due to different climates, soil types and vegetation, local TFs should be observed. Previously, Uchida and Okabayashi [1] surveyed TF values for various crops collected in Japan and around the world. The reported TF data obtained under natural conditions were limited especially in Asian and South American countries. Despite efforts to collect TF data in these areas through a coordinated research programme entitled "Transfer of radionuclides from air, soil, and freshwater to the foodchain of man in tropical and sub-tropical environments" by the Food and Agriculture Organization of the United Nations (FAO) and the International Atomic Energy Agency (IAEA) in cooperation with International Union of Radioecologists, the data are still limited.

The TFs of radionuclides should be obtained under equilibrium conditions for assessing the environmental transfer of the radionuclides from routine releases. Thus, TFs observed under natural conditions using global fallout ^{137}Cs and ^{90}Sr should fit this purpose, however, the concentrations of ^{137}Cs and ^{90}Sr in crops are close to the lower detection limits of radiation measurements, thus, few TF data are available under natural conditions. 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 as alternatives to TFs of ^{137}Cs and ^{90}Sr. We chose inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectrometry (ICP-OES) to measure stable Cs and Sr. About 40-50 elements including Th and U were also measured in the cereals and the associated soils.

We particularly focused on the TFs for Cs from paddy soil to rice grains, because rice plants are grown under waterlogged conditions, which are completely different from wheat cultivation conditions. Additionally, rice is a diet staple for people living in Southeast Asian countries including Japan. The TFs for wheat were also compared with those of rice, because both crops are classified as cereals and many TFs have been reported for wheat in Europe and North America.

Using the elemental composition data of rice and associated soils obtained in this study, we proposed 'Reference rice' (brown and polished rice) and a 'Reference paddy soil'. The idea was born from a 'Reference plant' [2, 3], presented in the same vein as the 'Reference Man' that was developed by the International Commission on Radiological Protection [4]. For Reference plant, only components of green plants, which are native to central Europe or North America, were included in the evaluation [2]. However, in works to obtain local TFs, cereals, not green plants, should be considered for the subject countries. In the present study case, rice cultivated in Asian and South American countries is of interest, thus, it is necessary to establish a new reference.

2. EXPERIMENTAL

2.1 Soil, rice and wheat samples

Paddy soils (plowed soil layer: 0 - 20 cm) have been collected nationwide from 11 - 13 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 at 8 sampling sites by the National Institute of Agro-Environmental Sciences. 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 grains were husked. The soil samples were air-dried and passed through a 2-mm mesh sieve. The soil and crop samples were ground into fine powder.

2.2 Analytical methods for stable isotopes and natural radioisotopes

Solution samples are usually used for ICP-MS and ICP-OES; thus, samples were digested with mineral acids (a mixture of HNO_3, HF and $HClO_4$) using a microwave digester (CEM, MARS5). Sample amounts used were 100 mg for soils and 500 mg for crops. The digestion samples were made in duplicate. After the microwave digestion, each sample was evaporated to near 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 40-50 elements, including Cs, Sr, Th and U, in both crop and soil samples were measured using ICP-MS (Agilent 7500, Yokogawa) and ICP-OES (VISTA-Pro, Seiko) after diluting the acid solutions 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

3.1 Transfer factors of stable elements and natural radioisotopes

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 a 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). Figure 1 shows the results of TFs (geometric mean) for 23 elements. Some elements showed TF values higher than 0.1: Mg, K, Mo and Cd for wheat; Mg, K, Zn and Mo for brown rice; and Zn and Mo for polished rice. The TFs of Th and U were also of interest: the values for brown rice were 0.0001 and 0.00005; those for polished rice were 0.00015 and 0.0002; and those for husked wheat were 0.0008 and 0.0002, respectively. Except for several trace elements, TFs were usually highest in wheat followed by brown rice and then polished rice.

There are TF data for ^{137}Cs and ^{90}Sr in brown rice, polished rice and husked wheat [5-7] and the data were summarized recently [8]; thus, the obtained TF-stable Cs and Sr values were compared. The geometric-mean-TFs of ^{137}Cs were 0.0026 (brown rice) and 0.0011 (polished rice) for the samples collected in 1999 while those of stable Cs were 0.0013 (brown rice) and 0.0007 (polished rice). The geometric-mean-TFs for ^{90}Sr were 0.015 for brown rice and 0.0059 for polished rice, while those for stable Sr were 0.0038 for brown rice and 0.0013 for polished rice. From these results, apparently, these two elements were associated mostly with the rice bran. It should be noted that the values were close to each other, but the TFs of ^{137}Cs and ^{90}Sr were usually higher than those of stable Cs and Sr. 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 and ^{90}Sr were more mobile and more easily adsorbed by plants than stable Cs and Sr in the soil. Possibly, some of the stable Cs and Sr are found in soil structures that ^{137}Cs and ^{90}Sr cannot easily enter, so that these isotopes have not reached an equilibrium condition. However, because the TF values for radioactive and stable isotopes were almost the same, the TF-stable Cs and Sr could be used for long-term transfer of ^{137}Cs and ^{90}Sr in the environment.

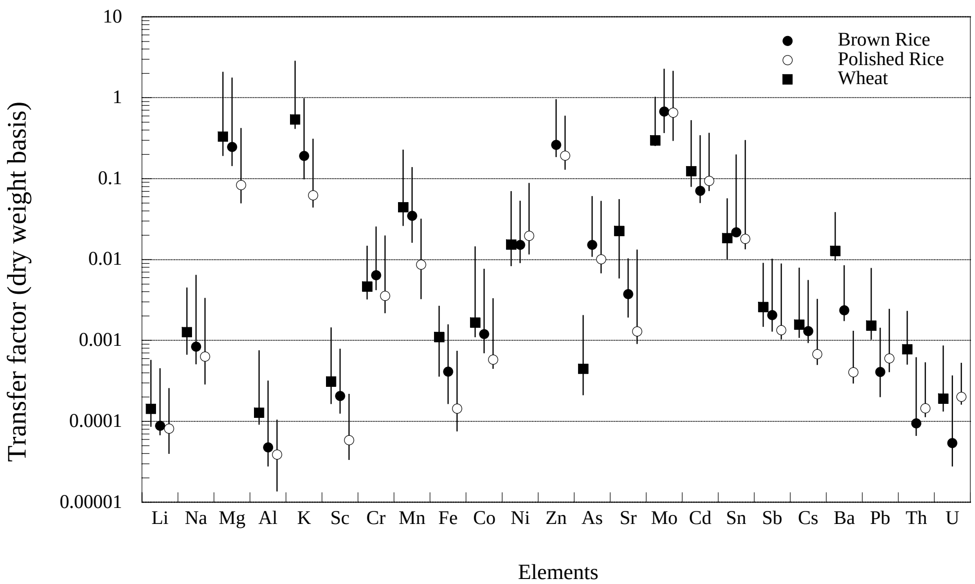

Figure 1. Geometric means of TFs (dry weight basis) for wheat, brown rice and polished rice. Both ends of bar show maximum and minimum TF values.

The geometric-mean-TFs of ^{137}Cs and ^{90}Sr in husked wheat were 0.0029 and 0.083, respectively [8], and those of stable Cs and Sr were 0.0016 and 0.022, respectively. The IAEA [10] reported expected TF values for cereals collected in a temperate climate; the TF-Cs and TF-Sr were 0.01-0.083 and 0.02-0.21, respectively. Both expected values were slightly higher than the TFs of ^{137}Cs, ^{90}Sr, stable-Cs and -Sr we obtained in Japan. Probably, climates, soil types and vegetation influence the parameters. Although our data are limited, further study is necessary to collect local TF values for precise radiological assessment.

3.2 Reference rice and paddy soil

To obtain information about the possible effects of radiation doses on humans, ICRP developed data for the chemical, physical, anatomical, and physiological composition of the ‘Reference man’ [4]. Analogous to ‘Reference man’, Markert [2] established a ‘Reference plant’. Although Reference plant only includes element concentration data of green plants in European and North American countries,

Table 1. Contents of elements in the Reference rice and SRM 1568a in mg/kg dry weight. Empty boxes indicate no data are available.

Z	Element	Brown rice (mg/kg)	Polished rice (mg/kg)	SRM 1568a (mg/kg)	Z	Element	Brown rice (mg/kg)	Polished rice (mg/kg)	SRM 1568a (mg/kg)
1	H				47	Ag			
2	He				48	Cd	0.027	0.041	0.022
3	Li	0.003	0.002		49	In			
4	Be		0.001		50	Sn	0.072	0.078	0.005*
5	B	2.5			51	Sb	0.0015	0.0016	0.0005*
6	C	415000			52	Te			
7	N	10090			53	I			0.009*
8	O				54	Xe			
9	F				55	Cs	0.006	0.003	
10	Ne				56	Ba	0.8	5.5	
11	Na	11	6.7	6.6	57	La	0.0012	0.0009	
12	Mg	1350	470	560	58	Ce	0.0031	0.0015	
13	Al	4.6	3.0	4.4	59	Pr	0.0009	0.0003	
14	Si	220			60	Nd	0.0015	0.0007	
15	P	2520		1530	61	Pm			
16	S			1200	62	Sm	0.0011	0.0003	
17	Cl			300*	63	Eu	0.0003	0.0005	
18	Ar				64	Gd	0.0011	0.0004	
19	K	2310	770	1280	65	Tb	0.0001	0.0005	
20	Ca	78	32	118	66	Dy	0.0005	0.0002	
21	Sc		0.001		67	Ho	0.0001	0.0003	
22	Ti	0.80			68	Er	0.0004	0.0002	
23	V	0.007	0.005	0.007*	69	Tm	0.0001	0.0002	
24	Cr	0.29	0.21		70	Yb	0.0005	0.0003	
25	Mn	21	5.8	20	71	Lu		0.0003	
26	Fe	15	5.4	7.4	72	Hf			
27	Co	0.012	0.007	0.018*	73	Ta			
28	Ni	0.3	0.4		74	W			0.001*
29	Cu	2.9	2.5	2.4	75	Re			
30	Zn	25	18	19	76	Os			
31	Ga	0.008	0.005		77	Ir			
32	Ge				78	Pt			
33	As	0.08	0.08	0.29	79	Au			
34	Se	0.03		0.38	80	Hg			0.006
35	Br			8	81	Tl		0.0004	
36	Kr				82	Pb	0.010	0.018	<0.010*
37	Rb	5.0	2.2	6.1	83	Bi			
38	Sr	0.33	0.14		84	Po			
39	Y	0.002	0.001		85	At			
40	Zr				86	Rn			
41	Nb				87	Fr			
42	Mo	0.69	0.67	1.46	88	Ra			
43	Tc				89	Ac			
44	Ru				90	Th	0.0007	0.0008	
45	Rh				91	Pa			
46	Pd				92	U	0.0002	0.0005	0.0003*

*Noncertified values.

each plant species can be inorganically characterized by a specific element distribution pattern in accumulation or rejection of elements compared to ‘Reference Plant’. Using the elemental composition data, chemical fingerprints can be described for brown rice and polished rice, however, it is more useful to develop ‘Reference rice’ in order to provide the elemental composition data directly as reference values in mathematical transfer models and to compare the data with other crops.

Also, when TF is considered, chemical data for paddy soil are also of interest so that the data for ‘Reference paddy soil’ were also collected.

Table 2. Contents of elements in the Reference paddy soil and continental crust [11] in mg/kg dry weight. Empty boxes indicate no data are available.

Z	Element	Paddy soil (mg/kg)	Continental crust (mg/kg) [11]	Z	Element	Paddy soil (mg/kg)	Continental crust (mg/kg) [11]
1	H			47	Ag		0.07
2	He			48	Cd	0.3	0.10
3	Li	24	18	49	In		0.05
4	Be	1.2	2.4	50	Sn	1.7	2.30
5	B	17	11	51	Sb	0.7	0.30
6	C		1990	52	Te		
7	N		60	53	I		0.8
8	O		472000	54	Xe		
9	F		525	55	Cs	3.4	3.40
10	Ne			56	Ba	260	584
11	Na	9120	23600	57	La	14	30
12	Mg	6920	22000	58	Ce	32	60
13	Al	74700	79600	59	Pr	3.4	6.7
14	Si	273000	288000	60	Nd	13	27
15	P	1740	757	61	Pm		
16	S		697	62	Sm	2.9	5.3
17	Cl		472	63	Eu	0.7	1.3
18	Ar			64	Gd	2.8	4.0
19	K	13800	21400	65	Tb	0.4	0.7
20	Ca	12200	38500	66	Dy	2.8	3.8
21	Sc	11	16	67	Ho	0.6	0.8
22	Ti	4560	4010	68	Er	1.7	2.1
23	V	100	98	69	Tm	0.3	0.3
24	Cr	50	126	70	Yb	1.8	2.0
25	Mn	650	716	71	Lu	0.3	0.4
26	Fe	38700	43200	72	Hf		4.9
27	Co	11	24	73	Ta		1.1
28	Ni	22	60	74	W		1
29	Cu	25	25	75	Re		0.0004
30	Zn	110	65	76	Os		0.00005
31	Ga	15	15	77	Ir		0.00005
32	Ge		1	78	Pt		0.0004
33	As	7.9	1.7	79	Au		0.0025
34	Se	0.5	0.1	80	Hg	0.22	0.04
35	Br		1	81	Tl	0.43	0.52
36	Kr			82	Pb	23	14.8
37	Rb	36	78	83	Bi		0.085
38	Sr	78	333	84	Po		
39	Y	15	24	85	At		
40	Zr		203	86	Rn		
41	Nb		19	87	Fr		
42	Mo	1.1	1.1	88	Ra		
43	Tc			89	Ac		
44	Ru		0.0001	90	Th	5.2	8.5
45	Rh		0.00006	91	Pa		
46	Pd		0.0004	92	U	2.5	1.7

The results are listed in Tables 1 and 2, for rice and soil, respectively. Only the data obtained in this study were used. From Table 1, comparison of the data for brown rice and polished rice indicated that the concentrations of essential elements, e.g., Ca, Fe, K, Mg and Mn, in brown rice were usually higher than those in polished rice, however, the concentrations of other non-essential elements, e.g., Cd, Sb, Th and U, were almost the same in brown rice and polished rice. Since Reference rice is established from short grain rice, the results can be compared with long grain rice, which is also a popular grain type in Asian countries. Certified values of SRM 1568a from NIST are also listed in Table 1. The standard reference material is fine rice powder of long grain type rice, husked (brown rice). When Reference brown rice and SRM 1568a were compared, for some non-essential elements, one order of magnitude differences were observed, however, concentrations of most elements were the same order of magnitude; that is, their elemental composition trend was almost the same. However, because the data are limited, further study is needed.

Concerning the reference paddy soil, we compared element concentrations with continental crust values [11]. Concentrations of alkali and alkaline earth metals in the Reference paddy soil were slightly lower than those in the continental crust. Possible reasons are (i) the rice paddy fields are covered with water during rice planting, so these elements might be leached from the soil by the irrigation water, and (ii) Na, K, Ca, and Mg are major nutrients for plants so that they should be absorbed by the rice plants from the paddy soil.

The data listed in Tables 1 and 2 for the Reference rice and paddy soil will have to undergo frequent modification. Multi-element analyses are available to give the chemical composition so that by adding data Reference rice and paddy soil will be sufficiently verified statistically.

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