Soil-to-Crop Transfer Factors of Naturally Occurring Radionuclides and Stable Elements for Long-Term Dose Assessment

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ABSTRACT

A soil-to-crop transfer factor, TF, is a key parameter that directly affects the internal dose assessment for the ingestion pathway, however, obtaining TFs of various long-lived radionuclides occurred during operation of nuclear power plants is difficult because most of them could not be found in natural environments. In this study, therefore, we collected crops and their associated soils throughout Japan and measured more than 50 elements to obtain TFs under equilibrium conditions. The TFs were calculated for 42 elements (Li, Na, Mg, Al, Si, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Y, Mo, Cd, Sn, I, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Ho, Er, Tl, Pb, Th and U) from their concentrations 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) divided by the concentration of the isotope in soil (in Bq.kg or mg/kg dry weight). Probability distributions of TFs for 62 upland field crops were usually log-normal type so that geometric means (GMs) were calculated. The values for the elements of interest from the viewpoint of long-term dose assessment were 2.5E-02 for Se, 7.9E-02 for Sr, 3.1E-03 for Cs, 4.2E-04 for Th and 4.6E-04 for U. Leafy vegetable showed the highest TFs for all the elements among the crop groups. It was clear that these data were usually within the 95% confidence limits of TFs compiled by IAEA in Technical Report Series 364.

INTRODUCTION

It is necessary to obtain the variations of transfer parameters that are used in mathematical models for precise long-term radiological assessment of nuclear facilities, especially deep underground disposal facilities. 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. Many reports on TF have appeared, and the data have been summarized [1-3] The TFs of radionuclides should be obtained under equilibrium conditions for assessing the environmental transfer of the radionuclides from routine releases. Obtaining TFs of various long-lived radionuclides such as Sr-90 and Cs-137 in natural environments is difficult because their concentrations are extremely low in crop and soil samples. Thus, few TF data are available under natural conditions.

A possible approach is to understand behavior of native elements as analogues of radioisotopes. In order to check differences between native elements and radioisotopes in their behaviors, previously, TFs of fallout Cs-137 and native Cs [4, 5] were reported. TFs of fallout Cs and native Cs did not have the same values but the differences were usually no larger than one order of magnitude. This result suggested the potential use of native stable elements as analogues of long-lived radionuclides. In our previous study, TFs of stable elements from paddy soils to rice and from upland soils to wheat were measured to obtain the local TFs as alternatives to TFs of radionucldies [6]. It was found that the TF-stable Cs and Sr could be used for long-term transfer of Cs-137 and Sr-90 in the environment.

In this study, more TFs of stable elements in various crops such as cabbage, spinach, carrot, radish and potato were measured. About 40-50 elements such as Sr, Cs, Th and U in plant and soil samples were determined by inductively coupled plasma mass spectrometry (ICP-MS) and inductively coupled plasma optical emission spectrometry (ICP-OES).

EXPERIMENTAL

Sixty two crops, e.g., cabbage, spinach, carrot, radish, potato, and wheat, and associated soils (plowed soil layer: 0 - 20 cm) were collected throughout Japan. The soil samples were air-dried and passed through a 2-mm mesh sieve. Edible parts of crop samples were taken and then washed with deionized water to remove dust and soil particles. The washed parts were chopped and freeze-dried. Finally, the soil and crop samples were thoroughly ground into fine powder. They were transferred into a glass vials and stored at room temperature.

Each sample, 100 mg for soils and 500 mg for crops, was digested with mineral acids (a mixture of HNO_3, HF and $HClO_4$ for soil samples and HNO_3, HF and H_2O_2 for plant samples) using a microwave digester (CEM, Mars 5). All the acids used were ultra-pure analytical grade (Tama Chemicals, AA-100). After the microwave digestion, each sample was evaporated to near dryness on a hot plate at 140˚C, and the residue was dissolved in 20 mL of 2% HNO_3. The digestion samples were made in duplicate. More than 50 elements, including Cs, Sr, Th and U, in both crop and soil samples, were measured by ICP-MS and ICP-OES 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.

Standard reference materials, such as SRM-1573a (National Institute of Standards and Technology, tomato leaves), GBW-07603 (Institute of Geophysical and Geochemical Exploration, bush twigs and leaves), JB-3 (Geological Survey of Japan, igneous rock) were also analyzed together with the samples to check the accuracy of the method.

RESULTS AND DISCUSSION

Elemental concentrations in soils and crops, respectively; 54 elements and 52 elements, respectively were measured for soils and crops. The TFs were calculated from the concentrations of the radioactive or natural isotope in both crop and soil samples. The TF is defined as follows.

$$TF = \frac{\text{The concentration of an isotope in a crop (in Bq/kg or mg/kg dry weight)}}{\text{The concentration of the isotope in soil (in Bq/kg or mg/kg dry weight)}}$$

TFs of C (most of which is accumulated from the atmosphere), N, and 10 elements (numbers of samples for which concentrations were determined were less than 80% of total measured numbers) were not included so that TF values of 40 elements (Li, Na, Mg, Al, Si, P, K, Ca, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Rb, Sr, Y, Mo, Cd, Sn, Cs, Ba, La, Ce, Pr, Nd, Sm, Eu, Gd, Dy, Ho, Er, Pb, Th and U) obtained for all the crop samples were obtained. Probability distributions of TFs were usually log-normal type; examples for Ni, Se, Sr, Cs, Th, and U are shown in Fig.1. Thus, geometric mean (GM) was calculated for each element and the values for 40 elements are plotted in Fig.2. The TF-GM for each element was from 2.3E-4 (Al) to 2.1 (K).

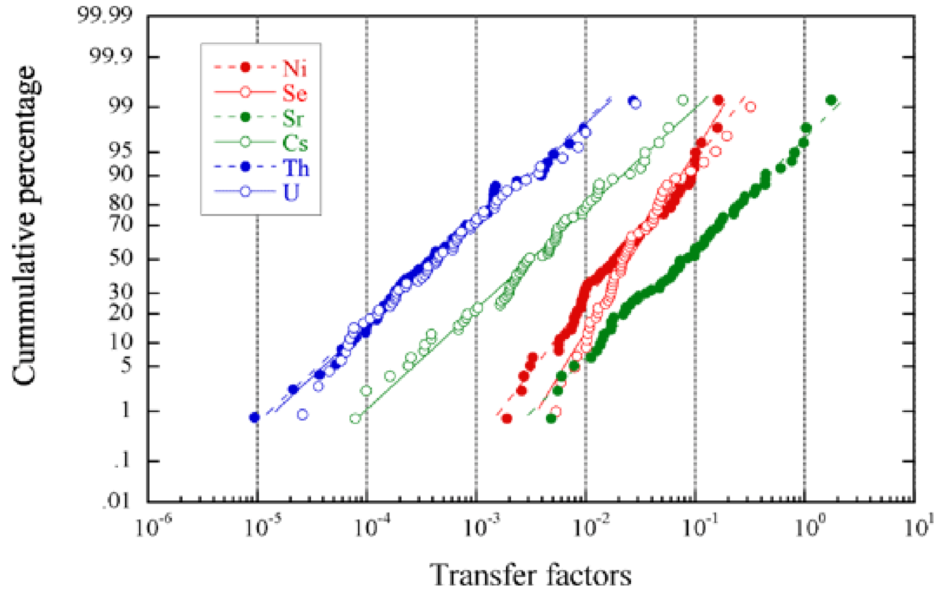

Fig.1. Probability distributions of TFs of Ni, Se, Sr, Cs, Th and U.

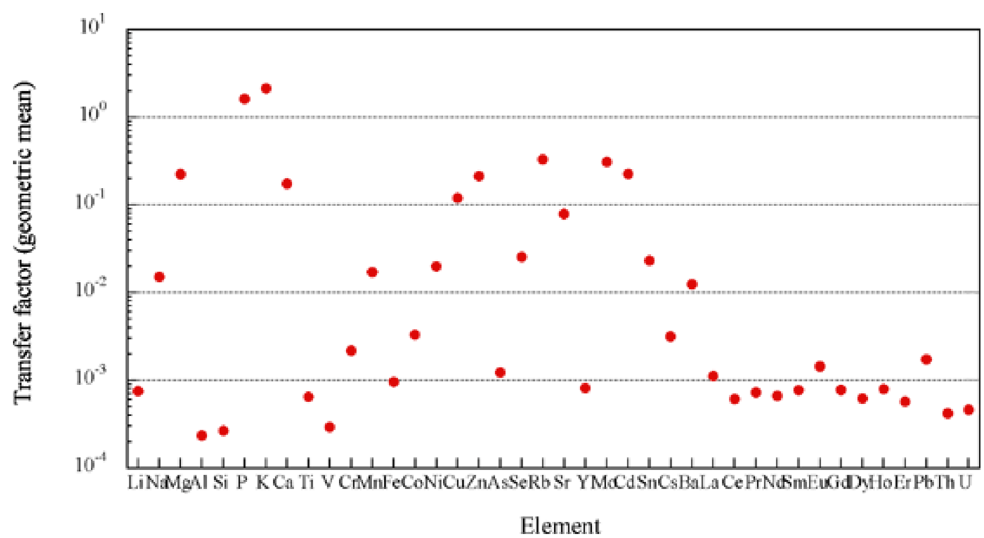

Fig.2. Geometric means of TFs of 40 elements for all samples.

The TF-GMs of essential elements for plants (i.e. Mg, P, K, Ca, Mn, Ni, Cu, Zn and Mo) were usually higher than nonessential elements, such as rare earth elements, Th and U. TF-GM of Rb was higher than that of Cs though it is not an essential element, but its ionic radius is close to that of K and Rb which may be taken up by plants alternatively.

Table I shows the TFs of Mn, Co, Zn, Sr, Cs, Pb, Th and U for 4 crop groups, i.e. leafy vegetable, fruit vegetable, potato and root vegetable samples. As can be seen from the table. leafy vegetable showed the highest TFs for most of the elements among the crop groups. The obtained data are also compared with the TF data compiled by IAEA [1], TRS-364, and it was clear that the present data were usually within the 95% confidence limits of TRS-364, but usually lower than the geometric mean values in TRS-364. Only exception was TFs of Th for potato, which was one order of magnitude higher than the value in TRS-364. However, number of samples was small so that it is necessary to obtain more data for further data analysis.

Table I. Transfer factor (Minimum, Maximum and Geometric mean values) of 9 elements for crop groups collected in Japan and comparison with the expected data in TRS-364 [1]

	Leafy vegetable (N=18)			Fruit vegetable (N=12)			Potato (N=6)			Root vegetable (N=8)		
Element	min	max	geomean	min	max	geomean	min	max	geomean	min	max	geomean
Mn	6.0E-03	1.2E-01	2.5E-02	4.6E-03	7.9E-02	1.5E-02	3.1E-03	1.3E-02	5.7E-03	4.9E-03	3.7E-02	8.8E-03
(TRS-364)			*(2.4-8.6)E-02*			-			*4.7E-02*			*2.6E-01*
Co	1.9E-03	3.9E-02	5.8E-03	1.1E-03	8.2E-03	3.6E-03	2.5E-03	6.2E-03	4.2E-03	8.7E-04	2.7E-02	3.4E-03
(TRS-364)			*(0.44-2.9)E-01*			-			*6.0E-02*			*(1.2-1.3)E-01*
Zn	1.3E-01	1.3E+00	2.7E-01	1.2E-01	3.1E-01	2.1E-01	1.0E-01	1.6E-01	1.3E-01	7.7E-02	2.3E-01	1.4E-01
(TRS-364)			*3.3E+00*			-			*3.5E+01*			-
Se	8.3E-03	3.2E-01	3.3E-02	5.4E-03	9.1E-02	1.8E-02	1.2E-02	4.8E-02	2.2E-02	1.0E-02	2.4E-02	1.6E-02
(TRS-364)			-			-			-			-
Sr	7.3E-02	1.0E+00	2.5E-01	6.0E-03	1.8E-01	4.4E-02	4.8E-03	7.2E-02	2.0E-02	2.2E-02	1.7E+00	1.3E-01
(TRS-364)			*(0.26-3.0)E+00*			-			*(0.2-2.6)E-01*			*(1.1-1.4)E+00*
Cs	3.3E-04	7.7E-02	5.4E-03	3.9E-04	3.5E-02	5.6E-03	8.3E-04	5.7E-02	4.5E-03	9.7E-04	1.3E-02	3.2E-03
(TRS-364)			*(1.8-4.6)E-01*			*2.2E-01*			*(0.7-2.7)E-01*			*(1.1-4.0)E-02*
Pb	3.0E-04	6.0E-02	2.9E-03	2.2E-04	4.3E-03	1.1E-03	4.1E-04	1.8E-03	7.9E-04	9.0E-04	9.3E-03	2.3E-03
(TRS-364)			*1.0E-02*			-			*1.3E-03*			*2.0E-02*
Th	9.8E-05	2.7E-02	7.6E-04	9.4E-06	1.4E-03	1.5E-04	2.5E-04	2.4E-03	7.2E-04	7.9E-05	1.3E-03	5.5E-04
(TRS-364)			*1.8E-03*			-			*5.6E-05*			*(0.003-3.9)E-02*
U	7.0E-05	2.8E-02	6.6E-04	4.5E-05	7.3E-04	1.9E-04	1.8E-04	2.3E-03	6.9E-04	9.4E-05	1.9E-03	5.8E-04
(TRS-364)			*8.3E-03*			-			*1.1E-02*			*1.4E-02*

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