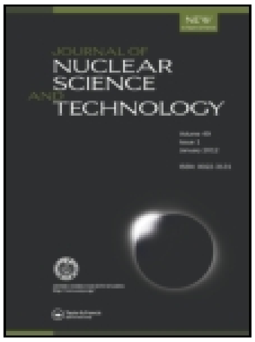

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ARTICLE

Soil-to-Plant Transfer Factors of Stable Elements and Naturally Occurring Radionuclides
(1) Upland Field Crops Collected in Japan

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In long-term dose assessment models for radioactive waste disposal, an important exposure pathway to humans is *via* ingestion of contaminated foods. In order to obtain soil-to-plant transfer factors (TFs) of radionuclides under equilibrium conditions, naturally existing elements were measured as analogues of radionuclides. Crops grown in upland fields and associated soil samples were collected from 62 sampling sites throughout Japan. The total concentrations of 52 elements in the crops and 54 elements in the soil samples were measured. The TFs 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) were calculated on a dry weight basis.

Among all the TF data, K showed the highest TF with a geometric mean (GM) of 2.1, followed by P. The GMs of TFs for rare earth elements, Th and U were on the order of 10^{-4}. Most of the TF-GMs for green vegetables were higher than GMs of all crops for the elements. The obtained TFs of some elements for green vegetables and potatoes were compared with those in the technical report series-364 (TRS-364) compiled by IAEA in 1994. The TF-GMs were usually lower than the best estimates (expected values) listed in TRS-364; however, the GMs of TF for La and TF for Th observed for potatoes were slightly higher than the expected values.

KEYWORDS: transfer factor, soil, plant, edible part, stable isotope, naturally occurring radionuclide, upland field condition, TRS-364

I. Introduction

Long-lived radionuclides released from nuclear facilities, such as deep underground disposal facilities, could reach humans through several transfer paths in the environment. According to some reports,[1,2] important radionuclides to be considered are ^{3}H, ^{14}C, ^{36}Cl, ^{60}Co, ^{59,63}Ni, ^{79}Se, ^{90}Sr, ^{93}Zr, ^{93m,94}Nb, ^{93}Mo, ^{99}Tc, ^{107}Pd, ^{108m}Ag, ^{126}Sn, ^{129}I, ^{135,137}Cs, ^{151}Sm, ^{210}Pb, ^{226}Ra, ^{227}Ac, ^{229,230}Th, ^{231}Pa, ^{233,234,235,236,238}U, ^{237}Np, ^{238,239,240,241,242}Pu, ^{241,242m,243}Am, and ^{244,245,246}Cm. One of the major paths of these radionuclides is food consumption. Consequently, obtaining data on the root uptake of radionuclides to an edible part of a crop, that is, the soil-to-plant transfer factor (TF), is important in long-term dose assessment models. Many reports have appeared, and the data have been summarized.[3–6] The TF data were usually obtained under laboratory conditions or in open fields using short-lived radionuclides, and typical observation periods were less than one year, such as from planting to harvest of crops. It is possible that a freshly added radionuclide would not be at equilibrium in the soil environment. For example, the TFs of Tc obtained under laboratory conditions are much higher than under field conditions.[7] To close the gap, longer time range studies have also been carried out, but usually not more than 5 years or so.[8,9]

For a precise long-term dose assessment, therefore, it is not clear whether such parameters are suitable to use in the models or not. Moreover, the critical paths of radionuclides and the critical foods in Japan are different from those in the European countries because agricultural products and food customs are different. For instance, livestock products including meat, eggs and milk make a big contribution in the European and North American countries, whereas, crops such as rice and vegetables are the main contributors in Japan. Consequently, safety assessment for people who live in Japan must consider rice and leafy vegetables as critical foods. However, the numbers of their data are limited.

Naturally occurring radionuclides (NORs), such as ^{226}Ra, ^{232}Th and ^{235,238}U, have been studied in order to understand their long-term behaviors and to estimate the radionuclide

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transfer to humans from the environment.[10,11] It is likely that fallout radionuclides derived from atmospheric nuclear weapons testing, such as ^{90}Sr and ^{137}Cs, can also be used for this purpose as more than 40 years have passed since the fallout peak was observed in 1963. However, data on NORs and fallout radionuclides would not be sufficient to cover the data for radionuclides generated in peaceful uses of atomic energy. Thus, even if some NORs and fallout radionuclides could be measured, there is still a general lack of knowledge on the environmental behaviors of many long-lived radionuclides of interest as listed above. A possible approach is to measure native elements as analogues of radionuclides of interest, however, it is necessary to test the differences of native elements and radioisotope behaviors. Vera Tome *et al.*[10] collected TF data of NORs together with stable elements for comparison. In some reports, TFs of fallout ^{137}Cs and native Cs,[12,13] and fallout ^{90}Sr and native Sr[14] were measured: TFs of fallout ^{137}Cs and native Cs did not have the same values but the differences were usually no larger than one order of magnitude. For ^{90}Sr, it was observed that the concentration ratio of ^{90}Sr/Sr had a constant value in the different components of rice plants in each sampling site. These results suggested the potential use of native stable elements as analogues of long-lived radionuclides.

For several radioisotopes, there is/are stable isotope(s), such as ^{88}Sr for ^{90}Sr, ^{127}I for ^{129}I and ^{133}Cs for ^{135,137}Cs, so that the stable isotopes can serve as analogues. However, for radioisotopes that have no stable isotopes, it might be possible to use other elements having biogeochemical characteristics that resemble the radioisotopes as a way to add informational values. For instance, Vandenhove *et al.*[15] reported that Ba is considered a better tracer for Ra based on the significant linear correlation found between Ra and Ba-TF. For ^{99}Tc, we found that Re can be used as the analogue.[16]

Thus, in this study, crops (edible parts) and associated soil samples were collected throughout Japan to obtain the TFs under equilibrium conditions. Here, the TFs of 40 elements are reported (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) for crops grown in upland field conditions. Among the TF data in the present study, 9 stable elements and 2 naturally occurring radionuclides related to the long-lived radionuclides mentioned above, *i.e.*, Co, Ni, Se, Sr, Mo, Sn, Cs, Sm, Pb, Th, and U, are reported, while rare earth elements give some ideas to behaviors of actinides. Further, in TRS-364,[3] the TFs of 37 elements (Na, Cr, Mn, Fe, Co, Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Mo, Tc, Ru, Rh, Ag, Sb, Te, I, Cs, Ba, La, Ce, Pr, Nd, W, Pb, Po, Ra, Th, U, Np, Pu, Am, and Cm) were compiled; the TFs of 19 elements presented in this study (Na, Fe, Cu, Zn, Rb, Y, and Ba together with Co, Ni, Sr, Mo, Cs, La, Ce, Pr, Nd, Pb, Th, and U: the latter 12 elements were already mentioned as long-lived radionuclides' related elements) will provide additional information to the TRS-364 data. Other elements will also be useful since informational value can be obtained from chemically similar elements, *e.g.*, Li, Mg, Ca, Sr, and Ba (alkaline earth metals).

Fig. 1 Sampling sites in Japan

II. Experiments

1. Sample Collection

Upland field soils (plowed soil layer: up to ca. 20 cm in depth) were collected nationwide from 62 sampling sites in 2002–2004 as shown in **Fig. 1**. Soils were classified as Fluvisols (27 sites), Andosols (21 sites), Cambisols (13 sites), and Regosols (1 site) according to the FAO/UNESCO soil taxonomy.

From one sampling field, 5 sub-samples, approximately 1 kg on a fresh weight basis each, were collected in the harvesting season and these sub-samples were mixed well. About 15 kg in amount (on a fresh weight basis) of edible parts of crops were also collected. Crops included 18 green vegetable samples (cabbage, Chinese cabbage, spinach, lettuce, and so on), 10 tuber samples (potatoes, sweet potatoes, and taro), 7 allium samples (leeks and onions), 2 legume samples (beans and peas), 12 fruit vegetable samples (cucumbers, tomatoes, sweet pepper, and so on), 8 root vegetable samples (carrots and Japanese radish), and 7 cereal samples (wheat and barley).

Three kg of the soil samples were air-dried and passed through a 2-mm mesh sieve. For crop samples, edible parts were washed with deionized water at least 3 times, and the removal of dust and soil particles was completed. The washed parts were paper-towel dried, chopped, and freeze-dried. For leeks, green and white parts were separated. Leaves of carrots and Japanese radish are also edible so that roots and leaves were separated and elemental compositions of both of them were measured. Finally, samples of 62 soils and 68 crop parts were obtained and they were separately and thoroughly ground into fine powders. The powders were transferred into glass vials and stored at the room temperature. Before measurements, the samples were dried in an oven at 80°C.

2. Measurements

The pH (H_2O) of the soils was measured at a soil:water ratio of 1:2.5. The contents of total carbon and total nitrogen were analyzed with the CHN analyzer (Euro Vector, Euro-EA3000) using 1–1.5 mg of the crop samples and 10 mg of the soil samples. Boron in the soils was measured by inductively coupled plasma (ICP) optical emission spectrometry (Perkin-Elmer Optima 3300DV) after alkaline fusion of the samples.

For the measurement of major and trace elements, samples were prepared as follows. The soil samples, 100 mg each, were digested with mineral acids (a mixture of HNO_3, HF, and $HClO_4$) using a microwave digester (CEM, Mars 5). For the crop samples, 500 mg were used. To them were added 10 m*l* HNO_3 and 4 m*l* HF. The mixtures were heated for 10 h at 80°C to decompose organic matter. After that, microwave digestion was carried out. Following their digestion, both the soil and crop samples were evaporated to near dryness at 140°C. The residue was dissolved with 1 m*l* of conc. HNO_3 and 0.5 m*l* of H_2O_2 and evaporated again. Finally, the residue was dissolved with 1 m*l* of 40% HNO_3 and diluted with deionized water. The digestion samples were made in duplicate. All the acids used were ultra-pure analytical grade (Tama Chemicals, AA-100). Water (>18.1 MΩ) which was treated by a Milli-Q water system (Millipore Co.) was used throughout the work.

After diluting the acid solutions to a suitable concentration, elements in both the crop and soil samples were measured using ICP mass spectrometry (Yokogawa, Agilent 7500c). Measurements of major elements such as Na, K, Ca, Mg, Fe, Al, and Ti were done by ICP optical emission spectrometry (Seiko, Vista Pro). Standard solutions were purchased from SPEX Certiprep Inc. (XSTC series).

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), and JB-3 (Geological Survey of Japan, igneous rock) were also analyzed together with the samples to check the accuracy of the method.

III. Results and Discussions

1. Elemental Compositions in the Soils and the Crops

Tables 1 and **2** summarize elemental concentrations in all the soils and the crops samples (on the dry weight basis), respectively, to understand elemental composition trends in upland field soils and crops. Fifty-four and 52 elements were measured for the soils and the crops, respectively. For the crop samples, B and Sb concentrations were usually close to or lower than the detection limits so that they were not determined. Further, for some crop samples, concentrations of some elements were lower than the detection limits so that numbers of the samples shown in Table 2 were not always 68. For example, U concentration was measured for 58 samples but not determined for 10 others.

For the soil samples, pH ranged from 4.3 to 8.1 with an arithmetic mean of 6.3. Almost all the studied elements were determined in the samples. Table 1 also lists median, arithmetic mean, and geometric mean (GM). The maximum/minimum ratios were less than 10 for 29 elements and 10–100 for 25 elements. The results were within the value ranges reported by Takeda *et al.*[17] and the present median values were almost same as the reported median values of 514 Japanese soils in the same reference (MV-514J-soils). However, because only the agricultural soils were used in this study, it is necessary to consider addition of some elements including U that originated from fertilizers. For U, although the maximum/minimum ratios were small, its GM was 1.4 times higher than MV-514J-soils, when U/Th concentrations were calculated using the GMs obtained in agricultural fields. Previously, it was estimated that about 48% of total U in upland field soils (range: 4–74%) came from phosphatic fertilizers.[18]

In order to compare elemental contents in the three soil groups, that is, Fluvisols, Andosols, and Cambisols, the GM of each element for each soil group was divided by MV-514J-soils, and the results are plotted in **Fig. 2**. Andosols are young soils from volcanic deposits and they are widely distributed around the Pacific Rim so that Andosol fields are a common type in Japan, but not in Europe. This unique soil type is one of the dominant Japanese agricultural field soils for upland crops;[19] thus, it is necessary to check the chemical composition differences among the soil types. In the studies of Andosols, GMs of C, N, Ca, and Cu were 2 times higher than MV-514J-soils while the GM of Rb was only 1/3 of that of MV-514J-soils. Also other alkaline metals had usually lower GMs than in other soil types. The GMs of light rare earth elements and Th were lower than those of MV-514J-soils; this trend has already been reported.[20] Compared to Andosols, elemental concentrations of Fluvisols and Cambisols were close. In Fluvisols, slightly higher concentrations of heavy metals and higher concentrations of rare earth elements were observed, compared with Cambisols, possibly due to parent material differences.

Most of the elemental data for the crop samples varied, compared with those of soils as shown in Table 2; essential elements such as C, N, P, Mg, K, Mn, Fe, Ni, Cu, Zn, and Mo (except Ca) showed a narrow distribution range (maximum/minimum ratio), usually less than two orders of magnitude, compared with other non-essential elements. Then the GMs of elemental composition for each crop group were compared with those of the reference plants (mean elemental composition values of whole green plant parts grown in central Europe and North America) reported by Markert[21] as shown in **Fig. 3**. Green vegetables usually showed the highest concentrations while cereals showed the lowest among the crop groups. The GMs for all the samples were almost same or slightly lower than the reference plant values, for example, GM/reference plant concentration ratios for Be, C, N, Na, Mg, P, K, Sc, and Ti were within 0.5–2. Among the essential elements, the ratio was lowest for Mn; since its maximum/minimum ratio was relatively lower than other essential elements, it is probable that the Mn content in the reference plants was higher than that in many plants. Indeed, leaves of *Vaccinium vitis-idea* (cowberry) and *Vaccinium myrtillus* (bilberry) were reported to contain quite high amounts of Mn (604–674 $mg\,kg^{-1}$ dry);[21] since only limited numbers of samples were used to establish

Table 1 Concentrations of 54 elements and pH for 62 upland field soil samples on dry weight basis

	Li mg kg^{-1}	Be mg kg^{-1}	B mg kg^{-1}	C g kg^{-1}	N g kg^{-1}	Na g kg^{-1}	Mg g kg^{-1}	Al g kg^{-1}	Si g kg^{-1}	P g kg^{-1}	K g kg^{-1}	Ca g kg^{-1}	Sc mg kg^{-1}	Ti g/kg	V mg kg^{-1}	Cr mg kg^{-1}	Mn g kg^{-1}	Fe g kg^{-1}	Co mg kg^{-1}
N[a)]	62	62	57	62	62	62	62	62	62	62	62	62	62	62	62	62	62	62	62
Minimum	8.1E+0	4.9E-1	6.5E+0	2.2E+0	5.7E-1	4.2E-1	3.6E-1	5.5E+1	1.7E+2	4.4E-1	1.3E+0	6.3E-1	3.7E+0	1.5E+0	3.0E+1	2.1E+1	3.1E-1	1.3E+1	3.1E+0
Maximum	7.9E+1	2.0E+0	8.1E+1	9.6E+1	6.4E+0	2.1E+1	2.6E+1	2.1E+2	3.7E+2	7.4E+0	3.7E+1	6.0E+1	3.5E+1	1.1E+1	3.8E+2	1.5E+2	2.0E+0	1.1E+2	3.9E+1
Max./Min.	10	4	12	45	11	51	72	4	2	17	28	95	10	7	13	7	6	8	13
Median	2.8E+1	1.4E+0	2.3E+1	2.6E+1	2.6E+0	1.0E+1	6.5E+0	8.3E+1	2.7E+2	2.2E+0	1.2E+1	1.1E+1	1.2E+1	4.8E+0	1.1E+2	6.2E+1	9.0E-1	4.3E+1	1.4E+1
Arithmetic mean	2.9E+1	1.4E+0	2.4E+1	3.3E+1	3.1E+0	1.0E+1	8.7E+0	8.6E+1	2.6E+2	2.4E+0	1.3E+1	1.5E+1	1.4E+1	4.8E+0	1.4E+2	6.3E+1	9.1E-1	4.7E+1	1.5E+1
Geometric mean	2.6E+1	1.3E+0	2.1E+1	2.7E+1	2.7E+0	8.8E+0	6.8E+0	8.3E+1	2.5E+2	2.1E+0	1.1E+1	1.1E+1	1.2E+1	4.5E+0	1.2E+2	5.9E+1	8.2E-1	4.3E+1	1.3E+1

	Ni mg kg^{-1}	Cu mg kg^{-1}	Zn mg kg^{-1}	Ga[b)] mg kg^{-1}	As mg kg^{-1}	Se mg kg^{-1}	Rb mg kg^{-1}	Sr mg kg^{-1}	Y mg kg^{-1}	Mo mg kg^{-1}	Cd mg kg^{-1}	Sn mg kg^{-1}	Sb mg kg^{-1}	Cs mg kg^{-1}	Ba mg kg^{-1}	La mg kg^{-1}	Ce mg kg^{-1}	Pr mg kg^{-1}	Nd mg kg^{-1}
N[a)]	62	62	62	51	62	61	62	62	62	62	62	62	62	62	62	62	62	62	62
Minimum	7.7E+0	7.1E+0	3.7E+1	1.2E+1	2.2E+0	1.2E-1	3.6E+0	1.2E+1	5.2E+0	3.0E-1	4.0E-2	8.6E-1	2.5E-1	7.6E-1	1.0E+2	3.8E+0	1.5E+1	1.1E+0	4.9E+0
Maximum	9.6E+1	4.0E+2	2.4E+2	3.2E+1	1.6E+2	1.6E+0	1.4E+2	2.4E+2	2.8E+1	3.4E+0	7.5E-1	1.4E+1	2.1E+0	1.1E+1	5.6E+2	3.8E+1	7.8E+1	8.7E+0	3.3E+1
Max./Min.	12	56	6	3	72	13	39	20	5	11	19	16	9	15	5	10	5	8	7
Median	2.4E+1	3.4E+1	1.2E+2	1.8E+1	9.6E+0	4.2E-1	3.2E+1	8.5E+1	1.6E+1	1.2E+0	3.4E-1	2.5E+0	7.8E-1	4.1E+0	2.4E+2	1.5E+1	3.3E+1	3.8E+0	1.5E+1
Arithmetic mean	2.6E+1	5.4E+1	1.2E+2	1.9E+1	1.4E+1	5.4E-1	4.2E+1	9.8E+1	1.6E+1	1.3E+0	3.7E-1	2.9E+0	8.1E-1	4.4E+0	2.9E+2	1.6E+1	3.6E+1	3.9E+0	1.5E+1
Geometric mean	2.2E+1	4.1E+1	1.2E+2	1.8E+1	1.0E+1	4.6E-1	3.2E+1	8.1E+1	1.6E+1	1.2E+0	3.4E-1	2.5E+0	7.3E-1	3.8E+0	2.6E+2	1.4E+1	3.3E+1	3.6E+0	1.4E+1

	Sm mg kg^{-1}	Eu mg kg^{-1}	Gd mg kg^{-1}	Tb mg kg^{-1}	Dy mg kg^{-1}	Ho mg kg^{-1}	Er mg kg^{-1}	Tm mg kg^{-1}	Yb mg kg^{-1}	Lu mg kg^{-1}	Hf[b)] mg kg^{-1}	W[c)] mg kg^{-1}	Tl mg kg^{-1}	Pb mg kg^{-1}	Th mg kg^{-1}	U mg kg^{-1}	pH (H_2O)
N[a)]	62	62	62	62	62	62	62	62	62	62	51	34	62	62	62	62	62
Minimum	1.1E+0	2.5E-1	1.1E+0	1.5E-1	9.6E-1	2.0E-1	6.5E-1	8.8E-2	6.3E-1	8.5E-2	9.3E-1	4.0E-1	1.3E-1	7.4E+0	1.2E+0	1.0E+0	4.3
Maximum	6.8E+0	1.4E+0	6.3E+0	9.3E-1	5.4E+0	1.1E+0	3.2E+0	4.8E-1	3.3E+0	5.0E-1	5.7E+0	2.9E+0	1.2E+0	4.6E+2	1.2E+1	4.2E+0	8.1
Max./Min.	6	6	6	6	6	5	5	6	5	6	6	7	9	62	10	4	
Median	3.4E+0	8.3E-1	3.4E+0	5.0E-1	3.0E+0	6.1E-1	1.8E+0	2.7E-1	1.8E+0	2.8E-1	3.2E+0	1.3E+0	4.6E-1	2.2E+1	5.3E+0	2.6E+0	6.3
Arithmetic mean	3.3E+0	8.3E-1	3.3E+0	4.9E-1	3.0E+0	6.2E-1	1.8E+0	2.7E-1	1.8E+0	2.8E-1	3.1E+0	1.3E+0	4.5E-1	3.2E+1	5.5E+0	2.6E+0	6.3
Geometric mean	3.1E+0	8.0E-1	3.1E+0	4.7E-1	2.9E+0	5.9E-1	1.8E+0	2.6E-1	1.8E+0	2.7E-1	3.0E+0	1.2E+0	4.1E-1	2.3E+1	5.0E+0	2.4E+0	

[a)]Numbers of samples for which concentrations were determined. [b)]11 samples were not measured. [c)]28 samples were not measured.

Table 2 Concentrations of 52 elements in 68 edible parts of 62 crop samples on dry weight basis

	Li	Be	C	N	Na	Mg	Al	Si	P	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co
	mg kg^{-1}	mg kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	mg kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	g kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}
N[a)]	68	37	68	68	68	68	68	68	68	68	68	44	62	63	68	68	68	68
Minimum	1.1E-3	2.1E-4	3.5E+2	4.0E+0	6.0E-3	5.5E-1	1.5E+0	6.0E-3	1.4E+0	4.2E+0	8.5E-3	1.3E-3	7.5E-1	4.5E-3	1.2E-2	3.4E+0	1.3E+1	4.2E-3
Maximum	8.6E-1	3.3E-2	6.1E+2	6.0E+1	5.8E+0	7.5E+0	1.6E+3	2.5E+0	7.8E+0	5.8E+1	3.4E+1	4.0E-1	9.3E+1	2.4E+0	3.2E+0	1.4E+2	9.3E+2	4.5E-1
Max./Min.	796	158	2	15	975	14	1095	421	6	14	4060	309	124	535	259	41	70	106
Median	1.6E-2	1.3E-3	3.9E+2	2.2E+1	1.4E-1	1.5E+0	1.6E+1	4.7E-2	3.4E+0	2.8E+1	3.0E+0	2.3E-2	2.3E+0	3.3E-2	9.1E-2	1.3E+1	3.7E+1	4.2E-2
Arithmetic mean	5.9E-2	2.7E-3	3.9E+2	2.4E+1	5.0E-1	1.8E+0	7.1E+1	2.3E-1	3.7E+0	2.8E+1	4.3E+0	4.3E-2	5.4E+0	1.3E-1	3.1E-1	2.0E+1	6.4E+1	6.8E-2
Geometric mean	1.9E-2	1.5E-3	3.9E+2	2.1E+1	1.3E-1	1.6E+0	2.0E+1	6.7E-2	3.4E+0	2.3E+1	2.0E+0	2.1E-2	2.9E+0	3.7E-2	1.3E-1	1.4E+1	4.1E+1	4.4E-2

	Ni	Cu	Zn	Ga[b)]	As	Se	Rb	Sr	Y	Mo	Cd	Sn	Cs	Ba	La	Ce	Pr	Nd
	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}
N[a)]	68	68	68	51	68	51	68	68	66	68	68	65	68	68	68	68	66	68
Minimum	5.8E-2	6.5E-1	4.5E+0	4.0E-3	2.1E-3	2.3E-3	6.0E-1	6.0E-1	8.0E-4	3.1E-2	4.9E-3	5.3E-3	4.8E-4	1.4E-1	1.4E-3	9.3E-4	2.0E-4	7.8E-4
Maximum	9.7E+0	6.4E+1	1.2E+2	5.3E-1	2.0E-1	2.4E-1	4.8E+1	8.8E+1	7.4E-1	5.6E+0	1.3E+0	1.2E+0	2.4E-1	5.1E+1	1.3E+0	2.0E+0	2.5E-1	9.1E-1
Max./Min.	166	99	26	133	96	105	80	147	931	181	268	216	504	370	949	2174	1209	1170
Median	4.1E-1	5.2E+0	2.5E+1	4.5E-2	9.8E-3	1.1E-2	1.2E+1	8.3E+0	1.2E-2	3.7E-1	7.9E-2	5.0E-2	1.4E-2	3.1E+0	1.4E-2	1.8E-2	2.5E-3	9.7E-3
Arithmetic mean	7.8E-1	6.8E+0	2.9E+1	9.4E-2	2.2E-2	2.1E-2	1.5E+1	1.4E+1	3.5E-2	6.3E-1	1.3E-1	1.5E-1	2.5E-2	7.2E+0	6.2E-2	7.8E-2	9.8E-3	3.5E-2
Geometric mean	4.5E-1	4.8E+0	2.5E+1	4.2E-2	1.2E-2	1.2E-2	1.0E+1	6.7E+0	1.3E-2	3.6E-1	7.6E-2	5.7E-2	1.2E-2	3.2E+0	1.6E-2	2.0E-2	2.7E-3	9.8E-3

	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu	Hf[c)]	W[c)]	Tl	Pb	Th	U
	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}
N[a)]	59	63	64	54	63	59	66	49	58	46	36	36	61	68	65	58
Minimum	2.2E-4	1.2E-4	1.5E-4	4.6E-5	7.4E-5	4.4E-5	4.9E-5	1.1E-5	1.3E-4	2.4E-5	3.2E-4	9.0E-4	4.0E-4	5.3E-3	8.3E-5	6.2E-5
Maximum	1.7E-1	3.4E-2	1.6E-1	2.2E-2	1.3E-1	2.4E-2	6.5E-2	8.8E-3	5.4E-2	8.0E-3	2.2E-2	1.2E-1	6.5E-1	1.4E+0	3.2E-1	8.8E-2
Max./Min.	785	280	1043	488	1720	541	1339	803	409	329	69	132	1608	256	3886	1407
Median	2.2E-3	9.5E-4	2.2E-3	4.2E-4	1.7E-3	4.2E-4	9.9E-4	2.2E-4	9.2E-4	2.1E-4	1.2E-3	5.1E-3	1.5E-2	3.6E-2	1.9E-3	1.2E-3
Arithmetic mean	7.5E-3	2.4E-3	7.2E-3	1.2E-3	5.5E-3	1.2E-3	3.0E-3	5.7E-4	2.8E-3	5.5E-4	2.3E-3	1.3E-2	5.4E-2	1.0E-1	9.9E-3	4.0E-3
Geometric mean	2.4E-3	1.2E-3	2.4E-3	4.4E-4	1.8E-3	4.6E-4	1.0E-3	2.3E-4	1.0E-3	2.3E-4	1.3E-3	5.9E-3	1.3E-2	4.0E-2	2.1E-3	1.1E-3

[a)]Numbers of samples for which concentrations were determined. [b)]17 samples were not measured. [c)]32 samples were not measured.

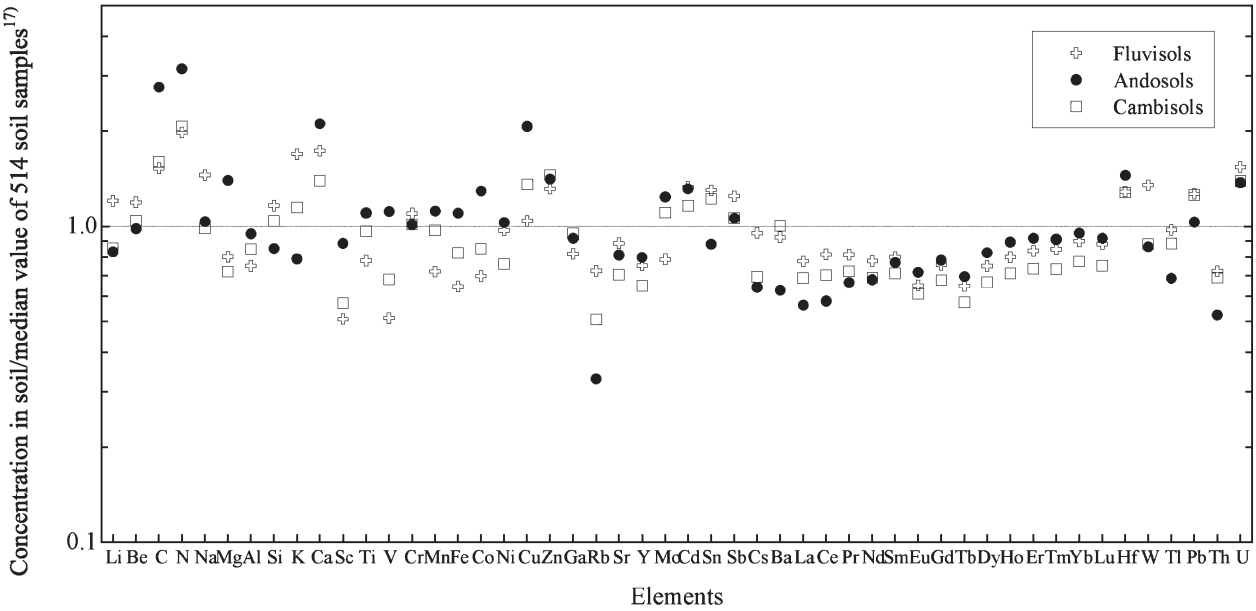

Fig. 2 Elemental compositions of Fluvisols, Andosols, and Cambisols normalized by mean values of Japanese soils (Takeda *et al.*)[17)]

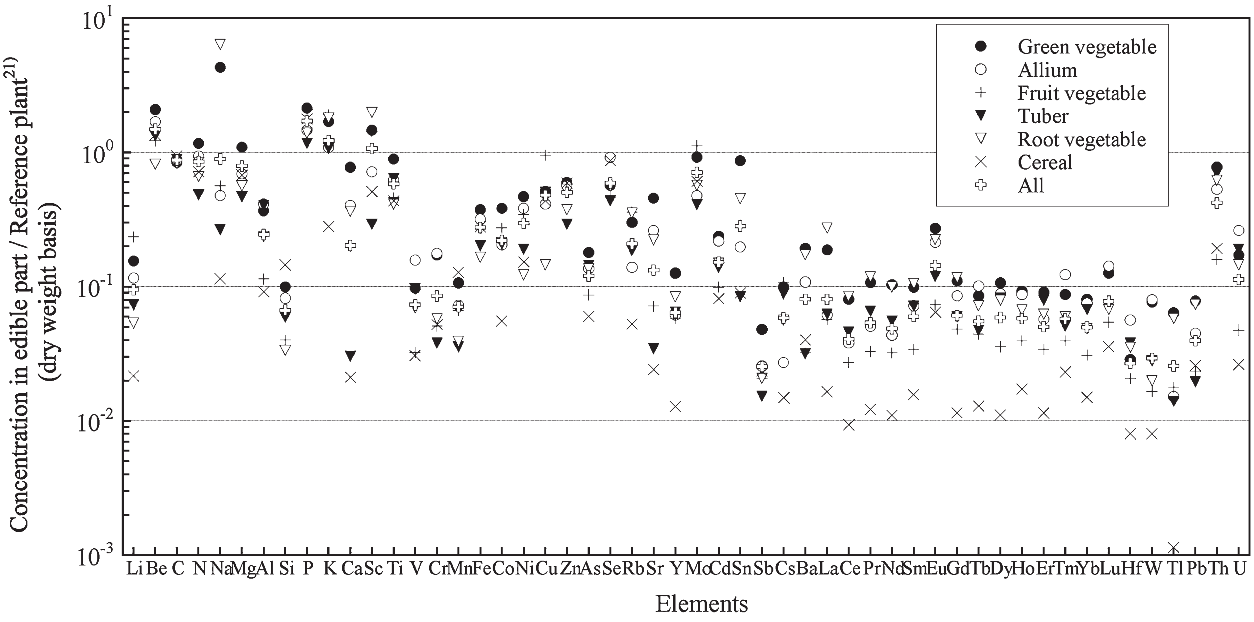

Fig. 3 Elemental compositions of some crop groups normalized by reference plant (Markert)[21)]

the reference plants, some modifications would be necessary in the future.

Following the GM of Mn, the GM of Ca was also low, especially in cereals and tubers, and due to these two crops, Ca concentrations for all the crops showed a wide range of distribution as shown in Table 2. The result is not a unique case, since low Ca concentrations in cereals and tubers are the well-known facts as listed in the food composition database.[22)] The other elements which had lower than 1/10 content of the reference plants were Li, Si, V, Cr, Cs, Ba, rare earth elements, Hf, W, Tl, and Pb. The results implied that these elemental concentrations in edible parts of agricultural crops were lower than those in other green plants such as trees and bushes.

2. Soil-to-plant Transfer Factors of 40 Elements

Soil-to-plant transfer factors, that is, the uptake of elements by plants from the soils, TFs, were calculated using the element concentration data in the soils and the crop samples. As described in TRS-364,[3)] the TF is defined as the concentration of a radionuclide in a crop (in $Bq\,kg^{-1}$ dry weight) divided by the concentration of the radionuclide in the soils (in $Bq\,kg^{-1}$ dry weight). The equation was also used for stable elements by using the unit "in $mg\,kg^{-1}$"

for both crop and soil samples. The 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 are listed in **Table 3**. The TFs of C (most of which is accumulated from the atmosphere), N, and 10 elements (numbers of samples for which concentrations were determined to be less than 80% of the total measured numbers) are not listed.

The probability distributions of the TFs for Mn, Co, Sr, Cs, La, Th, and U are presented in **Fig. 4**. The TFs for green vegetables (Fig. 4, left) had lognormal type distributions. Other crop groups have smaller numbers of samples so that it is difficult to test the distribution type, instead, the probability distributions of the TFs for all the samples are plotted (Fig. 4, right). As shown in the figure, each of the seven elements had a lognormal type distribution. The TFs of other elements for all the samples also showed a similar trend (data not shown). Thus, in this study, the arithmetic mean was not used for the TF analysis, but the GM and 95% lower and upper confidence limits were calculated as reported in TRS-364.

The GM of TF data (TF-GM) of each element for all the crops with 95% upper and lower confidence limits are plotted in **Fig. 5**. The TF-GMs of essential elements for plants were usually higher than nonessential elements, such as U, Th, and rare earth elements. Among all the TF data, K showed the highest TF with a GM of 2.1, followed by P. The TF-GM of Fe, an essential element for plants, was low because Fe is the major component of the soils. The TF-GMs of Al, Si, and V were also some of the lowest due to their high contents in the soils. The TF-GM of Rb was higher than that of Cs though Rb is not an essential element, but its ionic radius is close to the radius of K, and thus, Rb may be taken up by plants alternatively. The same tendency was observed in the TF-GMs of Sr and Ba, which are in the same group as Ca. Interestingly, the TF-GM of Cd was also high, though it is not essential to plants. The mechanisms have not been clarified sufficiently to explain the results.

Figure 5 also presents the TF-GMs of green vegetables, allium, fruit vegetables, tuber, root vegetables, and cereal samples. The TF-GMs of most of the elements for green vegetables were the highest among the crop types studied. As it is already known, the TFs depend on the plant types, the parts of the plants in question, the soil characteristics, and the physico-chemical form in the soils. A plant leaf, which is a major organ for photosynthesis, has a higher transpiration rate than that other plant organs have. Thus, leaves need more water and essential elements than other plant organs, therefore, elements are necessarily accumulated in the leaves. Indeed, the TFs of green vegetables have usually been higher in the previous reports.[3] Thus, the same trend was observed in this study.

Unfortunately, it is difficult to compare area differences in Japan because each crop type did not have a sample size large enough for making comparisons. Besides, the differences might be caused by the differences of climate and agricultural conditions. Further studies are needed for upland crops. However, since the numbers of data compiled in TRS-364 were sometimes less than 10, the present data could provide more information about the TFs of radionuclides under equilibrium conditions.

3. Comparison of the TFs between the Previous Reports and This Study

Some TF data have already been reported for Mn, Co, Zn, Sr, and Cs on Japanese upland field crops, therefore, the present data were compared with those reported values. **Table 4** lists the TF-GMs for cabbage, Chinese cabbage, Japanese radish and carrots. The TF data for Mn, Co, and Cs obtained by the radiotracer experiments[23,26] were, by 1-2 orders of magnitude, higher than those in the present and field observation studies, possibly due to the aging effect of the soil samples. Freshly added radiotracers are usually in mobile forms such as water soluble and ion exchangeable forms, while stable elements in the soils have various physico-chemical forms such as organically-bound and specifically adsorbed forms and, frequently, they are occluded in weathering-resistant particles.

On the other hand, differences of the TFs of Zn and Sr were usually within 10 times. According to soil-soil solution distribution coefficients reported by Yasuda *et al.*,[27] Sr has a low ability to sorb on the soil solid phase, and, possibly, it is relatively mobile in the actual environment. Thus, the present TF-GMs of Sr were close to or just slightly lower than the radiotracer experiment results. Concerning the TF-GMs of Zn, although its mobility in upland fields is low,[27] the present results showed high uptake ratios for these four crops. It was previously reported that the water soluble and ion exchangeable Zn in an Andosols sample and a Gray lowland soils sample was strongly adsorbed on the soils after addition to the samples, however, strongly adsorbed or weakly complexed fractions were relatively high, that is, extracted-^{65}Zn/added-^{65}Zn ratios in the soil samples were more than 0.2 for 6 months.[28] Zinc is an essential element for plants; thus, Zn in strongly adsorbed or weakly complexed fractions might still be available to plants, though further studies are needed to clarify the Zn behavior. Table 4 also lists the TF data of the stable elements for crops collected in Aomori Prefecture[24,25] and the results were almost same as those observed in this study. However, the sample size for each crop was small so that it is difficult to discuss the differences between the results obtained in Aomori and in other sites throughout Japan.

The TFs for green vegetables and potatoes for selected elements, Mn (only for potatoes), Co, Sr, Cs, La, Pb, Th, and U, were compared with TRS-364 (**Fig. 6**). The values listed in TRS-364 were "expected values" and the 95% confidence range. The expected values were essentially best estimates; the compiled data were analyzed to estimate an expected value for the TF so that the value is considered 'typical' or most likely to occur. In the TF table of TRS-364, the TFs were classified by the type of the soils if experimental conditions were known. For green vegetables, "clay, loam pH = 6" was selected if there was any choice in the literature because the median of soil pH was 6.3 and the percentage of peat or sandy soils was negligible in the sampled upland fields.

Table 3 Soil to crop transfer factors of 40 elements on dry weight basis

No.	Latin name	English name	Li	Na	Mg	Al	Si	P	K	Ca	Ti	V
1	*Brassica oleracea* var. *capitata*	Cabbage	7.3E-4	5.1E-2	2.6E-1	6.6E-5	6.7E-5	4.4E+0	1.9E+0	4.8E-1	n.d.	1.4E-4
2	*Brassica oleracea* var. *capitata*	Cabbage	9.6E-4	2.3E-2	1.2E-1	2.1E-4	1.5E-4	9.6E-1	3.8E+0	2.7E-1	n.d.	1.3E-4
3	*Brassica oleracea* var. *capitata*	Cabbage	1.1E-3	1.7E-2	1.7E-1	1.1E-4	1.5E-4	3.8E+0	1.3E+0	4.9E-1	n.d.	1.2E-4
4	*Brassica oleracea* var. *capitata*	Cabbage	5.7E-4	5.1E-2	4.3E-1	2.6E-4	3.0E-4	1.4E+0	2.5E+0	9.2E-1	9.1E-4	4.8E-4
5	*Brassica oleracea* var. *capitata*	Cabbage	6.0E-3	4.1E-2	2.8E-1	9.9E-5	4.5E-5	6.4E+0	1.4E+0	7.6E-1	4.3E-4	1.5E-4
6	*Brassica oleracea* var. *capitata*	Cabbage	6.4E-4	3.7E-2	3.4E-1	2.0E-4	1.6E-4	3.3E+0	1.0E+0	1.5E+0	7.0E-4	3.4E-4
7	*Brassica oleracea* var. *capitata*	Cabbage	8.5E-4	2.9E-2	4.6E-1	5.9E-5	3.2E-5	1.0E+0	1.1E+0	3.1E-1	6.6E-4	2.4E-4
8	*Brassica rapa* L.	Chinese Cabbage	1.3E-3	4.0E-1	8.2E-2	5.0E-4	6.9E-4	3.7E+0	9.0E+0	9.7E-1	3.6E-4	3.5E-4
9	*Brassica rapa* L.	Chinese Cabbage	5.8E-4	4.6E-2	2.7E-1	7.1E-5	7.4E-5	4.5E+0	2.2E+0	1.4E+0	n.d.	n.d.
10	*Brassica rapa* L.	Chinese Cabbage	5.4E-4	1.6E-2	1.0E-1	2.1E-4	3.2E-4	3.0E+0	7.6E+0	2.7E-1	7.1E-4	2.0E-4
11	*Brassica rapa* L.	Chinese Cabbage	2.0E-4	5.3E-2	3.3E-1	6.3E-5	2.4E-4	2.0E+0	5.5E+0	2.2E+0	2.1E-4	3.1E-5
12	*Spinacia oleracea* L.	Spinach	4.7E-3	4.8E-1	1.5E+0	3.2E-3	2.3E-3	3.8E+0	3.3E+0	9.1E-1	2.5E-3	3.9E-3
13	*Spinacia oleracea* L.	Spinach	1.7E-3	6.9E-1	4.4E-1	5.5E-4	1.4E-3	1.8E+0	2.0E+1	6.0E-1	1.0E-3	8.3E-4
14	*Lactuca sativa* L.	Lettuce	3.9E-4	6.2E-2	2.4E-1	2.2E-4	5.8E-4	1.0E+0	6.2E+0	6.2E-1	3.3E-4	1.2E-4
15	*Lactuca sativa* L.	Lettuce	1.3E-3	3.7E-2	3.6E-1	4.0E-4	7.0E-4	2.2E+0	2.5E+0	3.1E-1	8.6E-4	1.9E-4
16	*Daucus carota* L.	Carrot (leaves)	2.5E-2	2.7E-1	2.2E-1	1.6E-2	9.7E-3	1.3E+0	2.7E+0	4.6E-1	1.6E-2	2.0E-2
17	*Raphanus sativus* L.	Radish (leaves)	4.5E-3	9.0E-1	2.8E-1	2.6E-3	2.7E-3	1.2E+0	7.4E-1	5.5E+0	2.1E-3	2.7E-3
18	*Brassica rapa* var. *hakabura*	Nozawana	4.7E-3	2.7E-2	3.6E-1	2.5E-3	3.4E-3	3.0E+0	3.8E+0	6.8E-1	3.5E-3	2.9E-3
19	*Solanum tuberosum* L.	Potato	7.3E-5	5.0E-4	7.5E-2	7.8E-5	6.9E-5	7.4E-1	2.6E+0	4.1E-3	n.d.	6.1E-5
20	*Solanum tuberosum* L.	Potato	2.6E-4	1.3E-3	2.0E-1	4.0E-4	2.1E-4	9.2E-1	1.4E+0	4.2E-2	3.9E-4	3.5E-4
21	*Solanum tuberosum* L.	Potato	1.5E-3	3.8E-3	2.0E-1	1.9E-3	8.9E-4	2.3E+0	1.7E+0	7.1E-2	1.7E-3	1.7E-3
22	*Solanum tuberosum* L.	Potato	6.6E-4	1.4E-3	1.2E-1	5.0E-4	3.0E-4	1.3E+0	1.8E+0	7.8E-4	6.0E-4	5.8E-4
23	*Solanum tuberosum* L.	Potato	1.0E-3	2.9E-2	1.9E+0	7.8E-4	4.4E-4	9.6E-1	1.6E+1	1.9E-1	4.3E-4	2.6E-4
24	*Solanum tuberosum* L.	Potato	5.7E-4	1.0E-2	7.8E-1	2.5E-4	2.1E-4	1.5E+0	7.3E-1	9.5E-2	7.2E-4	1.2E-3
25	*Ipomoea batatas* L.	Sweet potato	2.5E-4	6.8E-2	2.0E-1	2.5E-4	1.6E-4	4.3E-1	2.3E+0	1.7E-1	3.1E-4	1.9E-4
26	*Ipomoea batatas* L.	Sweet potato	1.8E-3	1.3E-2	1.1E-1	6.0E-4	8.6E-5	2.3E+0	7.3E-1	6.5E-2	4.3E-3	1.2E-3
27	*Colocasia esculenta*	Taro	1.9E-3	5.7E-4	1.7E-1	3.7E-4	1.7E-4	1.8E+0	1.8E+0	6.8E-2	7.3E-4	2.9E-4
28	*Colocasia esculenta*	Taro	3.9E-4	2.9E-2	3.5E-1	4.1E-4	3.7E-4	2.7E+0	1.5E+0	1.2E-1	9.0E-4	8.0E-4
29	*Allium fistulosum*	Leek (green part)	1.3E-3	6.2E-3	1.3E-1	5.0E-4	5.0E-4	7.5E-1	7.2E+0	9.4E-2	4.3E-4	4.6E-4
30	*Allium fistulosum*	Leek (white part)	3.6E-4	3.4E-3	8.7E-2	1.2E-4	1.8E-4	7.2E-1	5.5E+0	8.2E-2	1.5E-4	2.4E-4
31	*Allium fistulosum*	Leek (green part)	8.0E-3	9.7E-3	6.2E-1	4.7E-3	3.4E-3	1.8E+0	1.7E+0	1.4E+0	4.6E-3	5.6E-3
32	*Allium fistulosum*	Leek (white part)	1.1E-3	2.8E-3	1.8E-1	3.8E-4	2.7E-4	1.2E+0	1.0E+0	2.4E-1	3.1E-4	4.4E-4
33	*Allium fistulosum*	Leek (green part)	5.8E-3	2.2E-2	1.1E-1	6.4E-3	4.7E-3	1.3E+0	5.7E+0	3.6E-1	6.9E-3	6.3E-3
34	*Allium fistulosum*	Leek (white part)	6.8E-4	7.4E-3	4.2E-2	5.8E-4	5.7E-4	1.0E+0	3.5E+0	1.1E-1	7.4E-4	5.6E-4
35	*Allium fistulosum*	Leek (green part)	1.9E-3	4.6E-3	3.6E-1	1.2E-3	6.9E-4	1.3E+0	2.3E+0	9.9E-1	1.2E-3	1.1E-3
36	*Allium fistulosum*	Leek (white part)	3.2E-4	3.5E-3	2.0E-1	2.5E-4	1.3E-4	1.3E+0	1.8E+0	7.1E-1	6.7E-4	2.9E-4
37	*Allium cepa* L.	Onion	1.3E-3	6.2E-3	4.2E-1	7.3E-5	4.2E-5	1.9E+0	8.0E-1	2.5E-1	6.5E-4	n.d.
38	*Allium cepa* L.	Onion	4.4E-4	1.6E-2	1.7E-1	6.2E-5	5.6E-5	1.6E+0	1.3E+0	1.1E-1	2.8E-4	1.0E-4
39	*Allium cepa* L.	Onion	2.7E-4	8.9E-3	3.5E-1	1.2E-4	5.7E-5	1.7E+0	7.1E-1	6.4E-1	7.0E-4	2.2E-4
40	*Cucumis sativus* L.	Cucumber	5.4E-4	9.8E-3	2.4E-1	9.3E-5	1.8E-4	1.5E+0	7.1E+0	2.2E-1	4.2E-4	3.5E-5
41	*Cucumis sativus* L.	Cucumber	2.9E-4	8.6E-3	3.3E-1	1.5E-4	2.2E-3	1.2E+0	5.4E+0	1.6E-1	4.5E-4	1.1E-4
42	*Cucumis sativus* L.	Cucumber	1.2E-3	2.6E-3	4.3E-1	9.6E-5	6.5E-4	2.1E+0	4.8E+0	1.9E-1	4.2E-4	8.3E-5
43	*Solanum lycopersicum* L.	Tomato	2.8E-3	2.3E-2	2.5E-1	8.2E-5	9.5E-5	1.2E+0	1.4E+0	1.2E-1	6.0E-4	n.d.
44	*Solanum lycopersicum* L.	Tomato	7.7E-4	1.1E-1	3.5E-1	2.3E-5	5.8E-5	1.8E+0	2.4E+0	2.8E-1	4.0E-4	n.d.
45	*Solanum lycopersicum* L.	Tomato	1.7E-3	2.2E-2	2.4E-1	3.2E-4	1.6E-4	1.0E+0	2.5E+0	7.2E-2	4.7E-4	3.7E-4
46	*Solanum melongena* L.	Egg plant	6.2E-4	8.3E-3	2.4E-1	1.2E-4	3.8E-5	3.1E+0	5.3E+0	2.8E-1	2.6E-4	7.3E-5
47	*Solanum melongena* L.	Egg plant	1.7E-2	1.0E-2	4.2E-1	1.4E-4	2.7E-5	1.5E+0	1.4E+0	3.3E-1	5.8E-4	2.3E-4
48	*Solanum melongena* L.	Egg plant	8.8E-3	4.6E-3	1.4E-1	7.5E-5	2.0E-5	2.0E+0	1.9E+0	2.2E-1	5.0E-4	6.9E-5
49	*Capsicum annuum* L.	Sweet pepper	7.8E-3	1.0E-2	1.0E-1	9.3E-5	1.1E-4	3.8E-1	4.1E+0	3.6E-2	6.6E-4	7.2E-5
50	*Capsicum annuum* L.	Sweet pepper	5.6E-4	1.4E-2	1.9E-1	5.1E-4	3.5E-4	9.2E-1	4.1E+0	8.8E-2	5.5E-4	3.3E-4
51	*Momordica charantia*	Balsam pear	3.0E-3	5.0E-3	2.1E-1	3.8E-5	2.2E-3	8.1E+0	1.5E+0	9.3E-2	3.8E-4	3.5E-5
52	*Daucus carota* L.	Carrot	1.2E-3	7.5E-2	5.9E-2	8.8E-4	6.3E-4	1.2E+0	1.3E+0	6.6E-2	8.3E-4	1.0E-3
53	*Daucus carota* L.	Carrot	9.8E-4	9.6E-2	7.7E-2	2.2E-4	8.2E-5	1.1E+0	6.2E+0	1.1E-1	2.8E-4	1.4E-4
54	*Raphanus sativus* L.	Japanese radish	1.3E-4	2.4E-1	1.3E-1	8.0E-5	4.6E-5	8.0E-1	1.2E+0	5.2E-1	n.d.	8.9E-5
55	*Raphanus sativus* L.	Japanese radish	4.3E-4	1.6E-1	2.4E-1	1.5E-4	1.2E-4	9.5E-1	4.3E+0	4.7E-1	2.7E-4	3.5E-4
56	*Raphanus sativus* L.	Japanese radish	3.1E-4	1.1E-1	9.2E-1	4.2E-4	1.6E-4	2.2E+0	2.3E+0	1.3E+0	5.7E-4	3.3E-4
57	*Raphanus sativus* L.	Japanese radish	1.0E-3	4.6E-2	4.2E-2	2.0E-4	1.6E-4	6.9E-1	5.8E+0	8.4E-2	4.1E-4	2.4E-4
58	*Raphanus sativus* L.	Japanese radish	2.8E-4	1.9E-1	3.7E-1	2.2E-4	1.6E-4	7.1E-1	3.0E+0	7.2E-1	3.7E-4	3.6E-4
59	*Raphanus sativus* L.	Japanese radish	3.6E-4	1.5E-2	2.6E-1	9.1E-5	8.2E-5	9.4E-1	2.5E+0	1.3E-1	6.3E-4	6.7E-4
60	*Glycine max*	Soy bean	3.1E-5	1.0E-3	3.0E-1	7.0E-5	9.1E-5	4.4E+0	1.5E+0	1.3E-1	6.6E-4	7.4E-5
61	*Arachis hypogaea* L.	Peanut	1.6E-4	1.3E-3	3.8E-1	2.1E-4	1.1E-4	1.5E+0	1.4E+0	4.7E-2	3.9E-4	1.5E-4
62	*Triticum aestivum* L.	Wheat	1.6E-4	7.2E-4	6.3E-2	1.4E-4	2.7E-4	2.1E+0	6.2E-1	5.1E-3	3.3E-4	7.8E-5
63	*Triticum aestivum* L.	Wheat	5.9E-5	2.4E-3	1.6E-1	5.0E-5	2.2E-4	1.3E+0	1.1E+0	1.5E-2	3.2E-4	4.1E-5
64	*Triticum aestivum* L.	Wheat	2.9E-5	1.9E-3	2.0E-1	3.9E-5	2.0E-4	1.6E+0	4.0E-1	3.0E-2	3.2E-4	n.d.
65	*Triticum aestivum* L.	Wheat	5.5E-5	5.5E-4	1.0E-1	2.8E-5	1.4E-4	3.2E+0	2.8E-1	1.4E-2	3.2E-4	3.8E-5
66	*Triticum aestivum* L.	Wheat	2.0E-4	1.5E-3	1.2E-1	3.4E-5	2.0E-4	1.7E+0	3.1E-1	9.1E-3	3.6E-4	1.6E-4
67	*Hordeum vulgare* L.	Barley	9.5E-5	3.2E-3	4.3E-1	1.2E-4	8.7E-3	2.0E+0	4.1E-1	3.9E-2	4.8E-4	1.0E-4
68	*Hordeum vulgare* L.	Barley	1.2E-3	7.8E-3	3.0E-1	1.3E-3	7.4E-3	3.6E+0	6.9E-1	3.3E-2	1.5E-3	1.2E-3

n.d.: Not detected.

Table 3 (cont.-1).

No.	Cr	Mn	Fe	Co	Ni	Cu	Zn	As	Se	Rb	Sr	Y	Mo	Cd	Sn
1	3.1E-2	1.8E-2	9.4E-4	5.6E-3	1.6E-1	1.0E-1	1.3E-1	1.0E-3	n.d.	7.7E-1	1.4E-1	5.2E-4	5.3E-1	1.9E-1	4.7E-2
2	2.1E-2	2.1E-2	7.6E-4	5.8E-3	3.8E-2	4.3E-2	2.8E-1	6.3E-4	n.d.	4.0E-1	7.3E-2	3.4E-4	1.9E-1	1.3E-1	3.3E-1
3	4.8E-3	1.3E-2	3.0E-4	1.9E-3	5.1E-2	4.9E-2	1.4E-1	3.9E-4	n.d.	8.3E-2	1.7E-1	1.7E-3	7.9E-2	2.2E-1	4.7E-2
4	1.1E-3	2.2E-2	1.2E-3	3.9E-3	1.3E-2	5.1E-2	1.7E-1	1.1E-3	1.3E-2	3.7E-1	1.0E+0	9.6E-4	3.2E+0	8.2E-2	2.1E-2
5	5.0E-3	1.2E-1	8.4E-4	3.9E-2	1.6E-1	7.7E-2	4.0E-1	4.4E-4	3.6E-2	2.6E-1	1.1E-1	8.6E-3	3.6E-2	8.2E-1	1.3E-2
6	1.1E-3	2.3E-2	1.4E-3	3.7E-3	6.5E-2	6.3E-2	1.9E-1	2.1E-3	2.1E-2	1.3E-1	3.5E-1	7.3E-4	1.9E+0	1.5E-1	2.0E-2
7	5.9E-4	6.0E-2	1.6E-3	8.4E-3	2.0E-2	3.9E-2	1.7E-1	2.0E-3	1.1E-2	6.2E-2	7.3E-2	1.6E-4	6.9E-1	4.0E-2	4.9E-2
8	5.6E-3	6.0E-3	5.9E-4	2.2E-3	9.0E-3	3.1E-2	3.1E-1	3.3E-3	3.2E-1	2.2E+0	8.1E-1	1.2E-3	4.6E-1	3.1E-1	1.4E-1
9	5.1E-3	1.4E-2	8.9E-4	4.5E-3	7.3E-2	9.0E-2	2.8E-1	8.1E-4	n.d.	9.6E-2	3.6E-1	1.1E-3	4.2E-1	8.6E-1	2.6E-3
10	1.5E-3	1.0E-2	7.6E-4	2.5E-3	9.1E-3	2.1E-1	2.6E-1	1.3E-3	4.5E-2	4.5E+0	2.8E-1	9.0E-4	1.5E+0	3.9E-1	2.4E-1
11	3.4E-3	2.4E-2	6.7E-4	4.9E-3	7.8E-3	3.7E-2	2.8E-1	6.5E-4	1.5E-2	1.8E+0	7.6E-1	3.2E-3	4.7E-1	2.6E-1	4.9E-2
12	9.5E-3	4.1E-2	4.6E-3	1.4E-2	6.5E-2	3.8E+0	1.3E+0	6.9E-3	n.d.	5.4E-1	1.6E-1	4.0E-3	5.5E-1	2.6E+0	5.7E-1
13	2.1E-3	1.2E-2	1.3E-3	3.9E-3	9.4E-3	7.7E-2	6.2E-1	3.7E-3	4.1E-2	7.0E+0	4.3E-1	7.2E-3	1.2E-1	7.6E-1	6.9E-1
14	2.7E-3	2.8E-2	6.6E-4	2.6E-3	1.3E-2	2.4E-1	1.9E-1	5.6E-4	8.3E-3	1.4E+0	6.0E-1	8.1E-4	4.5E-2	4.5E-1	1.6E-1
15	1.1E-3	7.5E-2	2.7E-3	9.6E-3	4.0E-2	2.8E-1	3.6E-1	3.2E-3	1.7E-2	8.1E-2	1.2E-1	6.3E-4	1.2E-1	6.8E-1	1.4E-1
16	5.1E-2	4.3E-2	1.6E-2	2.5E-2	8.8E-2	2.9E-1	1.5E-1	2.5E-2	1.9E-1	1.6E-1	1.0E-1	2.7E-2	1.4E+0	2.4E+0	4.0E-2
17	7.2E-3	7.3E-2	3.2E-3	8.0E-3	1.6E-2	3.6E-1	2.2E-1	1.3E-2	n.d.	1.7E-1	9.7E-1	4.3E-3	3.1E-1	9.0E-1	3.4E-1
18	8.6E-3	3.0E-2	3.2E-3	8.0E-3	1.0E-1	5.8E-2	3.9E-1	4.0E-3	6.6E-2	1.1E+0	2.8E-1	4.3E-3	2.4E+0	4.2E-1	2.9E-2
19	3.5E-3	3.1E-3	2.5E-4	5.3E-3	1.1E-2	1.2E-1	1.1E-1	3.5E-4	n.d.	4.6E-1	4.8E-3	1.4E-4	1.5E-1	2.9E-1	3.6E-3
20	5.3E-4	3.3E-3	5.2E-4	3.9E-3	2.7E-3	1.1E-1	1.0E-1	4.1E-4	2.6E-2	8.1E-2	7.8E-3	5.2E-4	2.3E-1	3.0E-1	1.9E-2
21	1.9E-3	4.6E-3	2.0E-3	3.5E-3	9.8E-3	1.9E-1	1.6E-1	1.7E-3	2.1E-2	8.0E-2	2.3E-2	2.7E-3	3.5E-1	2.5E-1	3.1E-3
22	1.5E-3	5.6E-3	6.5E-4	2.5E-3	9.7E-3	1.6E-1	1.3E-1	7.9E-4	1.2E-2	3.6E-1	1.4E-2	1.0E-3	6.5E-2	3.4E-1	5.8E-3
23	1.3E-3	1.0E-2	5.2E-4	4.6E-3	4.1E-2	2.0E-1	1.5E-1	2.5E-3	4.8E-2	3.1E+0	7.1E-2	4.7E-3	1.0E-1	4.4E-1	1.0E-2
24	8.2E-4	1.3E-2	1.4E-3	6.2E-3	2.8E-2	1.6E-1	1.1E-1	1.0E-3	1.8E-2	6.3E-2	7.2E-2	7.8E-4	2.0E-1	6.5E-2	8.2E-3
25	5.1E-4	6.3E-3	4.5E-4	1.3E-3	5.8E-3	5.0E-2	7.2E-2	4.7E-4	1.1E-2	6.9E-1	1.4E-1	5.8E-4	4.2E-2	3.1E-2	3.1E-3
26	1.6E-3	4.2E-2	1.9E-3	4.3E-3	2.0E-2	3.2E-1	1.2E-1	3.9E-3	n.d.	6.1E-2	1.5E-2	2.6E-3	4.9E-1	1.2E-1	9.5E-3
27	7.7E-4	2.0E-2	8.8E-4	4.3E-3	1.1E-1	2.6E-1	9.6E-2	1.7E-3	2.2E-2	2.3E-1	1.3E-2	5.1E-4	3.1E-1	2.3E+0	n.d.
28	8.8E-4	1.4E-2	1.1E-3	2.6E-3	6.5E-3	1.8E-1	8.7E-2	5.8E-3	1.8E-2	1.4E-1	5.5E-2	4.1E-4	8.4E-1	1.7E-1	1.0E-3
29	7.2E-3	1.4E-2	9.3E-4	1.5E-3	3.1E-2	1.6E-1	3.5E-1	1.7E-3	n.d.	1.2E+0	5.0E-2	6.1E-4	1.3E-1	1.1E-1	5.1E-2
30	5.2E-2	5.9E-3	5.4E-4	2.0E-3	9.2E-2	8.7E-2	3.0E-1	8.1E-4	n.d.	6.9E-1	8.5E-2	1.3E-4	8.5E-2	1.1E-1	1.6E-2
31	1.6E-2	4.2E-1	6.5E-3	3.7E-2	9.7E-2	3.9E-1	3.6E-1	9.4E-3	1.2E-1	3.1E-1	2.5E-1	1.2E-2	1.8E-1	1.2E+0	1.5E-2
32	4.5E-3	4.5E-2	6.1E-4	1.6E-2	9.2E-2	2.1E-1	2.5E-1	1.3E-3	n.d.	1.6E-1	9.8E-2	1.1E-3	6.4E-2	8.0E-1	1.1E-2
33	1.1E-2	1.6E-2	5.8E-3	6.8E-3	9.1E-3	4.8E-2	2.2E-1	1.0E-2	5.5E-2	7.2E-1	1.6E-1	7.2E-3	6.5E+0	1.8E-1	7.1E-2
34	1.6E-3	5.2E-3	7.9E-4	1.0E-3	3.1E-3	2.6E-2	1.8E-1	2.0E-3	1.8E-2	3.7E-1	6.2E-2	1.0E-3	1.6E+0	1.4E-1	1.2E-2
35	4.1E-2	3.0E-2	1.8E-3	5.0E-3	6.0E-2	3.6E-1	3.6E-1	1.9E-3	4.6E-2	8.3E-1	4.4E-1	3.2E-3	2.2E-1	4.4E-1	3.0E-1
36	6.3E-3	1.4E-2	7.4E-4	2.3E-3	5.2E-2	2.1E-1	3.9E-1	1.1E-3	3.0E-2	5.2E-1	4.4E-1	7.2E-4	1.3E-1	5.4E-1	2.1E-2
37	7.8E-4	3.8E-2	1.6E-3	2.5E-3	3.8E-2	1.7E-1	4.6E-1	2.9E-3	n.d.	3.8E-2	6.0E-2	8.8E-5	4.4E-1	4.6E-1	n.d.
38	5.2E-4	1.0E-2	3.5E-4	2.8E-3	7.5E-3	5.8E-2	1.0E-1	1.0E-3	1.8E-2	3.7E-2	6.5E-2	1.5E-4	7.6E-2	3.7E-1	7.1E-3
39	1.0E-3	2.0E-2	1.3E-3	7.6E-4	3.3E-2	2.8E-1	2.7E-1	1.1E-3	n.d.	2.8E-2	1.8E-1	1.8E-4	5.2E-2	1.9E-1	3.9E-3
40	1.6E-3	1.2E-2	1.6E-3	1.3E-3	1.0E-1	4.4E-1	4.7E-1	2.4E-4	2.5E-2	5.5E-2	3.6E-2	8.9E-5	7.3E-1	9.0E-2	1.3E-2
41	6.2E-4	1.2E-2	3.4E-4	1.1E-3	1.8E-2	9.6E-2	2.1E-1	2.6E-4	1.7E-2	4.3E-1	6.3E-2	2.1E-4	1.6E+0	2.3E-1	7.5E-3
42	1.0E-3	9.4E-3	6.1E-4	1.3E-3	2.2E-2	1.5E-1	2.5E-1	1.2E-3	1.0E-2	1.9E+0	1.8E-1	1.7E-3	3.4E-1	1.8E-2	7.6E-3
43	3.4E-3	1.5E-2	5.9E-4	3.1E-3	2.1E-2	1.4E-1	3.1E-1	1.4E-3	2.6E-2	3.2E+0	9.7E-2	4.8E-4	7.1E-1	1.7E-2	4.4E-2
44	1.1E-3	1.5E-2	9.8E-4	5.9E-3	7.1E-2	2.9E-1	2.6E-1	4.7E-3	2.5E-2	2.5E+0	1.3E-1	4.7E-3	7.3E-1	7.2E-2	1.3E-1
45	7.9E-4	4.6E-3	1.5E-3	2.0E-3	3.1E-2	2.4E-1	1.5E-1	1.5E-4	3.5E-2	2.8E-1	1.1E-2	2.0E-4	2.3E-1	2.0E-1	6.8E-3
46	1.5E-3	2.0E-2	1.2E-3	6.1E-3	9.5E-3	2.5E-1	1.5E-1	3.1E-4	1.3E-2	2.7E-1	4.7E-2	4.1E-4	2.9E-1	4.5E-1	1.6E-2
47	2.5E-3	1.9E-2	1.3E-3	5.0E-3	1.2E-2	1.8E-1	1.2E-1	1.8E-3	5.9E-3	3.3E-1	2.9E-2	1.6E-3	8.2E-2	2.2E-1	3.5E-2
48	2.6E-4	1.3E-2	4.1E-4	2.3E-3	3.3E-3	1.1E-1	2.6E-1	6.2E-4	2.3E-2	1.8E+0	1.0E-1	9.1E-4	7.3E-1	2.3E-1	2.9E-2
49	3.3E-3	7.9E-2	1.5E-3	8.2E-3	6.0E-2	4.9E-1	2.3E-1	4.5E-3	n.d.	1.9E-1	2.5E-2	6.8E-4	7.5E-1	4.0E-1	9.0E-3
50	1.0E-3	3.4E-2	7.3E-4	5.1E-3	2.8E-2	3.5E-1	1.7E-1	2.2E-3	2.1E-2	3.0E-1	4.0E-2	3.5E-4	1.0E+0	2.1E-1	6.3E-2
51	7.4E-4	5.1E-3	4.5E-4	1.1E-3	1.7E-2	9.9E-2	2.4E-1	4.4E-4	n.d.	1.3E+0	6.0E-3	n.d.	3.6E-1	1.4E-1	2.2E-2
52	7.5E-3	8.9E-3	9.0E-4	7.5E-3	1.9E-2	1.7E-1	2.6E-1	7.7E-4	5.4E-3	9.2E-1	1.7E-2	1.6E-3	5.9E-2	3.3E-1	1.4E-2
53	9.4E-4	1.7E-2	1.0E-3	4.0E-3	3.3E-2	3.3E-1	2.4E-1	9.3E-4	9.1E-2	1.6E-1	1.2E-1	2.6E-4	2.7E+0	9.9E-2	3.4E-2
54	2.3E-3	5.3E-3	9.5E-4	2.1E-3	2.5E-2	1.5E-1	7.7E-2	2.5E-3	n.d.	8.1E-2	3.0E-2	1.5E-3	9.6E-2	1.0E+0	5.2E-2
55	1.0E-3	7.0E-3	3.8E-4	8.7E-4	7.8E-3	1.1E-1	1.6E-1	6.9E-4	1.0E-2	2.8E+0	2.2E-1	8.9E-4	6.6E-2	3.1E-1	5.2E-2
56	9.5E-4	6.0E-3	3.9E-4	1.7E-3	5.7E-3	3.4E-2	1.5E-1	6.1E-4	n.d.	2.8E-1	1.2E-1	1.5E-4	8.4E-2	1.9E-1	1.0E-2
57	1.1E-3	7.6E-3	5.1E-4	3.9E-3	1.0E-2	3.6E-3	1.1E-1	3.9E-4	1.0E-2	4.9E-1	2.3E-1	3.8E-3	1.6E-1	1.4E-1	5.0E-2
58	1.1E-3	2.4E-2	5.6E-4	1.9E-2	8.3E-3	4.7E-2	2.3E-1	1.0E-3	2.1E-2	9.3E-1	1.7E+0	4.2E-3	1.2E-1	3.2E-1	6.2E-3
59	6.6E-4	4.9E-3	4.6E-4	1.3E-3	2.6E-3	2.0E-2	1.7E-1	1.4E-3	2.4E-2	2.2E+0	2.2E-2	4.0E-4	8.9E-1	4.8E-2	7.8E-2
60	3.1E-3	3.7E-2	6.6E-4	2.7E-2	1.8E-2	2.9E-2	1.8E-1	1.1E-3	1.6E-2	6.5E-1	2.3E-1	3.0E-3	3.8E-1	2.9E-1	2.5E-2
61	1.1E-3	5.1E-3	6.0E-4	2.2E-3	1.5E-2	4.6E-2	1.2E-1	2.3E-3	2.3E-2	1.4E-1	4.8E-2	4.4E-4	1.2E+0	7.7E-2	4.5E-2
62	1.9E-3	1.9E-2	5.9E-4	2.8E-4	8.2E-3	1.1E-1	2.2E-1	2.5E-4	7.9E-3	2.4E-1	5.6E-3	1.6E-4	1.1E-1	1.2E-1	1.5E-2
63	2.0E-4	2.0E-2	5.7E-4	4.4E-4	1.9E-3	6.1E-2	3.4E-1	2.0E-4	1.5E-1	2.7E-1	1.7E-2	9.0E-5	3.4E-1	8.4E-2	2.3E-3
64	5.0E-4	1.1E-1	9.7E-4	6.0E-4	5.7E-3	1.3E-1	3.7E-1	3.2E-4	5.1E-2	7.6E-2	2.7E-2	n.d.	5.3E-1	2.1E-1	1.0E-2
65	2.1E-4	4.0E-2	1.1E-3	6.4E-4	7.4E-3	1.2E-1	3.2E-1	6.2E-4	4.9E-2	1.8E-2	1.4E-2	4.1E-5	3.6E-1	1.4E-1	n.d.
66	5.2E-4	7.5E-2	7.7E-4	7.4E-4	1.0E-2	1.1E-1	2.2E-1	1.2E-3	1.7E-2	2.4E-2	1.8E-2	9.5E-5	3.3E-1	2.3E-1	1.5E-3
67	1.6E-2	1.1E-2	7.8E-4	9.0E-4	1.4E-2	6.8E-2	2.0E-1	2.0E-4	4.2E-2	1.4E-1	2.3E-2	1.4E-4	2.0E-1	2.9E-2	3.7E-2
68	1.6E-2	2.2E-2	1.9E-3	2.2E-3	2.3E-2	2.8E-1	3.5E-1	2.4E-3	4.2E-2	1.4E-1	1.7E-2	1.3E-3	3.7E-1	8.4E-2	4.7E-3

n.d.: Not detected.

Table 3 (cont.-2).

No.	Cs	Ba	La	Ce	Pr	Nd	Sm	Eu	Gd	Dy	Ho	Er	Pb	Th	U
1	7.7E-2	1.1E-2	1.0E-3	5.0E-4	4.3E-4	4.4E-4	2.2E-4	n.d.	3.5E-4	2.0E-4	n.d.	1.5E-4	4.7E-4	n.d.	n.d.
2	1.8E-2	4.3E-2	9.5E-4	3.4E-4	2.7E-4	2.7E-4	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	8.9E-4	2.2E-4	n.d.
3	1.0E-3	2.8E-2	2.6E-3	1.7E-3	1.5E-3	1.5E-3	1.1E-3	1.4E-3	1.5E-3	9.4E-4	6.9E-4	6.5E-4	5.2E-3	9.9E-5	n.d.
4	6.9E-4	4.0E-2	8.0E-4	3.7E-4	5.4E-4	4.9E-4	5.4E-4	3.0E-3	6.1E-4	4.8E-4	5.3E-4	4.4E-4	4.9E-3	4.3E-4	3.6E-4
5	5.5E-3	4.4E-3	1.8E-2	8.7E-3	1.2E-2	1.1E-2	8.3E-3	8.4E-3	1.1E-2	7.0E-3	6.0E-3	4.8E-3	7.7E-4	1.8E-4	7.6E-5
6	2.3E-3	1.7E-2	9.3E-4	5.5E-4	6.3E-4	6.2E-4	6.2E-4	1.9E-3	6.6E-4	6.4E-4	7.8E-4	6.2E-4	1.4E-3	6.1E-4	4.5E-4
7	3.3E-4	1.6E-3	1.1E-4	9.1E-5	8.9E-5	9.5E-5	8.1E-5	2.8E-4	8.7E-5	1.2E-4	1.6E-4	1.1E-4	3.0E-4	9.8E-5	7.0E-5
8	2.5E-2	8.4E-2	3.8E-3	1.4E-3	1.8E-3	1.6E-3	1.2E-3	1.2E-3	1.1E-3	7.3E-4	6.1E-4	6.9E-4	7.5E-3	1.5E-3	1.5E-3
9	1.7E-3	4.0E-2	1.2E-3	1.6E-4	5.1E-4	5.6E-4	4.4E-4	1.1E-3	7.4E-4	4.6E-4	n.d.	3.6E-4	1.4E-3	n.d.	n.d.
10	2.5E-3	4.4E-2	1.3E-3	8.1E-4	7.9E-4	7.4E-4	6.1E-4	2.7E-3	5.9E-4	4.8E-4	4.3E-4	4.9E-4	7.4E-3	1.3E-3	8.8E-4
11	1.1E-2	9.2E-2	1.6E-2	1.0E-3	4.7E-3	4.0E-3	2.4E-3	1.1E-2	2.5E-3	1.5E-3	1.2E-3	1.2E-3	1.5E-3	4.2E-4	1.3E-4
12	4.7E-3	1.0E-1	4.0E-3	3.6E-3	3.8E-3	3.9E-3	3.5E-3	4.0E-3	3.7E-3	3.6E-3	3.9E-3	4.1E-3	7.0E-3	4.1E-3	3.8E-3
13	4.7E-2	1.5E-2	1.3E-2	3.6E-3	6.7E-3	6.0E-3	4.8E-3	5.4E-3	5.6E-3	4.5E-3	4.8E-3	4.4E-3	1.2E-2	4.5E-3	1.6E-3
14	4.5E-3	2.6E-2	1.5E-3	7.9E-4	7.7E-4	7.1E-4	4.3E-4	1.0E-3	4.5E-4	2.8E-4	3.6E-4	3.6E-4	2.3E-3	2.3E-4	1.1E-4
15	2.6E-3	4.8E-3	8.3E-4	6.5E-4	5.1E-4	5.2E-4	4.6E-4	7.9E-4	4.8E-4	4.4E-4	3.4E-4	4.7E-4	2.7E-3	3.6E-4	3.0E-4
16	3.2E-2	9.3E-2	3.5E-2	2.6E-2	2.8E-2	2.7E-2	2.6E-2	2.4E-2	2.5E-2	2.3E-2	2.3E-2	2.2E-2	6.0E-2	2.7E-2	2.8E-2
17	9.3E-3	6.1E-2	7.2E-3	3.6E-3	4.8E-3	4.6E-3	4.0E-3	5.2E-3	4.3E-3	3.9E-3	3.9E-3	4.0E-3	4.3E-2	3.9E-3	2.8E-3
18	6.6E-3	6.1E-2	5.8E-3	4.2E-3	4.7E-3	4.5E-3	4.5E-3	9.0E-3	4.5E-3	4.2E-3	4.4E-3	4.2E-3	5.8E-3	5.1E-3	6.1E-3
19	4.7E-3	1.4E-3	1.5E-4	1.1E-4	n.d.	1.3E-4	n.d.	n.d.	1.2E-4	n.d.	n.d.	n.d.	4.1E-4	2.5E-4	1.8E-4
20	2.0E-3	2.8E-3	5.5E-4	4.4E-4	5.0E-4	5.1E-4	5.1E-4	5.2E-4	5.3E-4	4.1E-4	3.2E-4	4.6E-4	5.1E-4	3.5E-4	4.4E-4
21	2.3E-3	9.4E-3	3.0E-3	2.3E-3	2.7E-3	2.7E-3	2.6E-3	3.0E-3	2.8E-3	2.6E-3	2.5E-3	2.6E-3	1.8E-3	2.4E-3	2.3E-3
22	7.7E-3	6.5E-3	1.0E-3	8.3E-4	8.7E-4	8.2E-4	8.2E-4	1.4E-3	8.1E-4	7.6E-4	8.5E-4	7.6E-4	6.7E-4	7.4E-4	6.6E-4
23	5.7E-2	1.3E-2	2.5E-3	3.0E-3	2.7E-3	3.0E-3	3.0E-3	4.0E-3	3.5E-3	3.4E-3	3.8E-3	3.6E-3	1.4E-3	1.5E-3	1.5E-3
24	8.3E-4	3.8E-3	1.0E-3	7.8E-4	8.5E-4	8.6E-4	7.8E-4	1.1E-3	8.1E-4	7.6E-4	7.7E-4	7.4E-4	6.7E-4	6.1E-4	6.1E-4
25	5.3E-3	9.1E-3	7.2E-4	4.5E-4	4.5E-4	5.0E-4	4.4E-4	7.1E-4	4.0E-4	3.4E-4	3.4E-4	4.3E-4	2.6E-4	3.0E-4	1.8E-4
26	9.3E-3	1.1E-3	7.5E-4	8.1E-4	8.5E-4	1.1E-3	1.6E-3	2.3E-3	2.1E-3	2.7E-3	3.1E-3	2.8E-3	1.7E-3	7.0E-4	3.7E-3
27	6.1E-3	4.6E-3	6.0E-4	4.6E-4	4.5E-4	4.7E-4	5.0E-4	8.1E-4	4.8E-4	4.8E-4	5.9E-4	4.7E-4	7.5E-4	4.0E-4	3.5E-4
28	8.1E-4	1.8E-3	4.8E-4	4.0E-4	4.8E-4	4.9E-4	5.2E-4	7.1E-4	5.6E-4	5.9E-4	6.4E-4	5.5E-4	4.7E-4	5.2E-4	1.7E-3
29	1.0E-2	1.1E-2	8.5E-4	7.0E-4	6.4E-4	6.5E-4	n.d.	5.2E-4	5.6E-4	5.2E-4	4.6E-4	5.5E-4	5.7E-3	8.1E-4	8.8E-4
30	4.6E-3	1.7E-2	2.3E-4	1.5E-4	n.d.	1.5E-4	6.0E-5	n.d.	n.d.	n.d.	n.d.	9.3E-5	4.7E-3	n.d.	n.d.
31	1.0E-2	4.5E-2	1.5E-2	1.1E-2	1.0E-2	1.0E-2	9.1E-3	9.8E-3	1.0E-2	9.3E-3	9.3E-3	9.3E-3	1.2E-2	7.1E-3	8.5E-3
32	1.8E-3	2.3E-2	1.1E-3	7.7E-4	6.9E-4	7.8E-4	6.3E-4	1.0E-3	8.2E-4	7.5E-4	n.d.	7.9E-4	1.8E-3	3.4E-4	6.2E-4
33	1.3E-2	6.7E-2	8.7E-3	7.6E-3	8.0E-3	8.2E-3	8.2E-3	9.8E-3	8.3E-3	8.0E-3	8.0E-3	8.2E-3	1.2E-2	9.6E-3	1.0E-2
34	2.8E-3	2.0E-2	1.0E-3	9.0E-4	8.7E-4	8.5E-4	8.7E-4	1.4E-3	8.5E-4	8.7E-4	9.4E-4	9.1E-4	1.7E-3	1.5E-3	1.0E-3
35	5.4E-3	1.2E-1	4.4E-3	2.7E-3	2.5E-3	2.4E-3	2.2E-3	6.6E-3	2.5E-3	2.1E-3	2.5E-3	2.1E-3	2.9E-3	1.4E-3	1.2E-3
36	1.9E-3	8.9E-2	1.5E-3	7.8E-4	6.9E-4	6.4E-4	6.3E-4	4.0E-3	7.0E-4	5.1E-4	4.1E-4	4.4E-4	1.6E-3	4.1E-4	3.2E-4
37	1.6E-4	3.1E-3	1.3E-4	8.7E-5	5.2E-5	6.5E-5	n.d.	3.7E-4	n.d.	n.d.	n.d.	6.4E-5	9.4E-4	3.7E-5	n.d.
38	9.9E-5	7.6E-4	8.6E-5	7.3E-5	1.0E-4	7.2E-5	n.d.	n.d.	1.8E-4	6.9E-5	2.6E-4	1.4E-4	5.3E-4	5.9E-5	n.d.
39	2.5E-4	9.8E-3	2.9E-4	1.4E-4	2.1E-4	1.9E-4	2.5E-4	1.5E-3	2.6E-4	2.7E-4	3.9E-4	2.5E-4	8.6E-4	2.0E-4	2.0E-4
40	3.9E-4	1.3E-2	1.0E-4	6.7E-5	6.5E-5	5.5E-5	n.d.	6.0E-4	1.6E-4	n.d.	n.d.	1.5E-4	3.0E-4	6.0E-5	1.9E-4
41	2.2E-3	1.5E-2	2.4E-4	2.3E-4	2.0E-4	1.8E-4	2.1E-4	3.8E-4	2.0E-4	1.3E-4	2.3E-4	1.4E-4	2.5E-3	2.0E-4	n.d.
42	1.2E-2	1.8E-2	2.8E-3	1.9E-3	1.6E-3	1.6E-3	1.2E-3	1.7E-3	1.4E-3	8.1E-4	8.1E-4	7.6E-4	4.3E-3	1.3E-4	1.6E-4
43	5.4E-3	1.7E-2	1.4E-3	3.2E-4	6.6E-4	5.8E-4	4.4E-4	1.0E-3	5.1E-4	3.9E-4	6.1E-4	3.9E-4	6.7E-4	2.7E-4	2.3E-4
44	3.4E-2	5.5E-2	9.9E-3	2.6E-3	4.7E-3	4.4E-3	3.3E-3	6.0E-3	4.0E-3	2.5E-3	2.5E-3	2.0E-3	7.8E-4	1.2E-4	1.3E-4
45	1.3E-2	5.4E-4	1.4E-4	2.2E-4	5.9E-5	7.6E-5	n.d.	1.3E-4	5.3E-4	3.4E-5	3.1E-4	9.3E-5	2.2E-3	2.1E-5	n.d.
46	5.3E-3	3.0E-3	3.7E-4	2.7E-4	1.8E-4	2.2E-4	1.6E-4	4.1E-4	2.0E-4	2.1E-4	5.8E-4	3.5E-4	2.2E-4	9.4E-6	n.d.
47	2.7E-3	3.2E-3	2.1E-3	1.5E-3	1.3E-3	1.3E-3	1.2E-3	1.3E-3	1.3E-3	1.3E-3	1.5E-3	1.4E-3	1.3E-3	1.4E-3	7.3E-4
48	6.2E-3	8.6E-3	1.3E-3	3.4E-4	6.8E-4	6.4E-4	4.9E-4	9.3E-4	6.0E-4	5.8E-4	6.5E-4	5.3E-4	1.6E-3	6.7E-4	2.3E-4
49	4.8E-3	3.2E-3	6.4E-4	2.6E-4	4.4E-4	3.7E-4	4.6E-4	9.6E-4	5.3E-4	5.9E-4	9.5E-4	5.3E-4	6.7E-4	1.8E-4	1.7E-4
50	1.6E-3	1.2E-2	5.2E-4	2.1E-4	2.9E-4	2.8E-4	2.2E-4	9.6E-4	3.0E-4	2.6E-4	2.3E-4	2.1E-4	3.9E-4	1.4E-4	7.5E-5
51	3.5E-2	9.8E-4	2.2E-4	9.8E-5	1.3E-4	1.2E-4	1.3E-4	1.6E-4	1.5E-4	1.1E-4	1.5E-4	9.1E-5	3.0E-3	1.6E-4	4.5E-5
52	3.1E-3	5.4E-3	1.4E-3	1.0E-3	1.0E-3	9.7E-4	9.3E-4	9.4E-4	9.7E-4	1.1E-3	1.1E-3	1.1E-3	2.4E-3	9.7E-4	4.6E-4
53	3.9E-4	2.9E-3	7.3E-5	4.7E-5	5.3E-5	5.4E-5	4.6E-5	2.1E-4	3.1E-4	8.2E-5	7.3E-5	7.9E-5	8.5E-4	5.2E-5	3.4E-4
54	5.4E-3	4.5E-2	2.0E-3	1.5E-3	1.5E-3	1.5E-3	1.4E-3	1.9E-3	1.4E-3	1.3E-3	1.1E-3	1.2E-3	2.7E-3	1.3E-3	1.9E-3
55	4.0E-3	5.8E-2	2.9E-3	7.3E-4	1.1E-3	8.8E-4	7.6E-4	2.6E-3	7.5E-4	5.1E-4	6.1E-4	5.1E-4	1.9E-3	1.0E-3	7.1E-4
56	2.7E-3	6.8E-3	4.6E-4	9.2E-5	1.8E-4	1.5E-4	n.d.	2.9E-4	n.d.	1.2E-4	2.3E-4	1.3E-4	1.7E-3	7.9E-5	9.4E-5
57	2.3E-3	4.5E-2	1.2E-2	8.9E-4	5.1E-3	4.0E-3	3.4E-3	6.0E-3	3.7E-3	2.4E-3	3.3E-3	1.9E-3	1.6E-3	5.0E-4	4.3E-4
58	2.9E-3	2.6E-1	2.8E-2	1.0E-2	8.2E-3	6.6E-3	4.3E-3	2.1E-2	5.6E-3	2.9E-3	2.9E-3	2.0E-3	2.5E-3	7.9E-4	6.2E-4
59	1.3E-2	5.4E-3	9.4E-4	7.7E-4	6.1E-4	5.3E-4	4.5E-4	6.2E-4	3.8E-4	4.1E-4	3.6E-4	3.8E-4	9.3E-3	1.1E-3	1.2E-3
60	2.1E-3	4.8E-2	3.9E-2	1.1E-2	1.1E-2	9.2E-3	5.0E-3	7.8E-3	6.4E-3	2.4E-3	1.8E-3	1.4E-3	2.7E-3	6.0E-4	6.6E-4
61	9.7E-4	6.1E-3	1.0E-3	5.9E-4	4.6E-4	4.1E-4	3.3E-4	8.1E-4	3.9E-4	3.8E-4	4.5E-4	3.4E-4	9.0E-4	2.9E-4	5.0E-4
62	2.0E-3	6.9E-3	2.6E-4	2.5E-4	2.5E-4	2.0E-4	2.0E-4	2.9E-4	1.7E-4	1.4E-4	2.6E-4	2.0E-4	2.3E-3	1.8E-4	3.6E-4
63	7.7E-4	4.3E-2	1.5E-4	7.9E-5	6.6E-5	6.5E-5	8.3E-5	1.0E-3	8.7E-5	7.8E-5	9.4E-5	6.5E-5	1.2E-3	1.0E-4	5.9E-5
64	3.4E-4	2.5E-2	1.4E-4	2.8E-5	8.6E-5	6.5E-5	1.2E-4	9.5E-4	8.5E-5	9.2E-5	2.1E-4	5.6E-5	3.4E-4	1.7E-4	2.6E-5
65	2.4E-4	1.0E-2	7.3E-5	3.7E-5	4.6E-5	4.5E-5	7.4E-5	6.8E-4	3.9E-5	2.2E-5	n.d.	2.4E-5	4.9E-4	7.1E-5	3.6E-5
66	7.9E-5	2.7E-3	1.7E-4	1.4E-4	1.2E-4	1.2E-4	n.d.	1.7E-4	6.4E-5	9.0E-5	7.9E-5	1.0E-4	2.6E-4	1.3E-4	6.8E-5
67	6.9E-4	1.5E-3	2.0E-4	2.2E-4	1.5E-4	1.3E-4	1.5E-4	4.3E-4	1.5E-4	6.1E-5	2.4E-4	1.9E-4	9.3E-3	1.5E-4	6.5E-5
68	2.0E-3	8.4E-3	1.4E-3	1.2E-3	1.5E-3	1.4E-3	1.3E-3	1.4E-3	1.4E-3	1.2E-3	1.2E-3	1.2E-3	2.5E-3	1.4E-3	9.8E-4

n.d.: Not detected

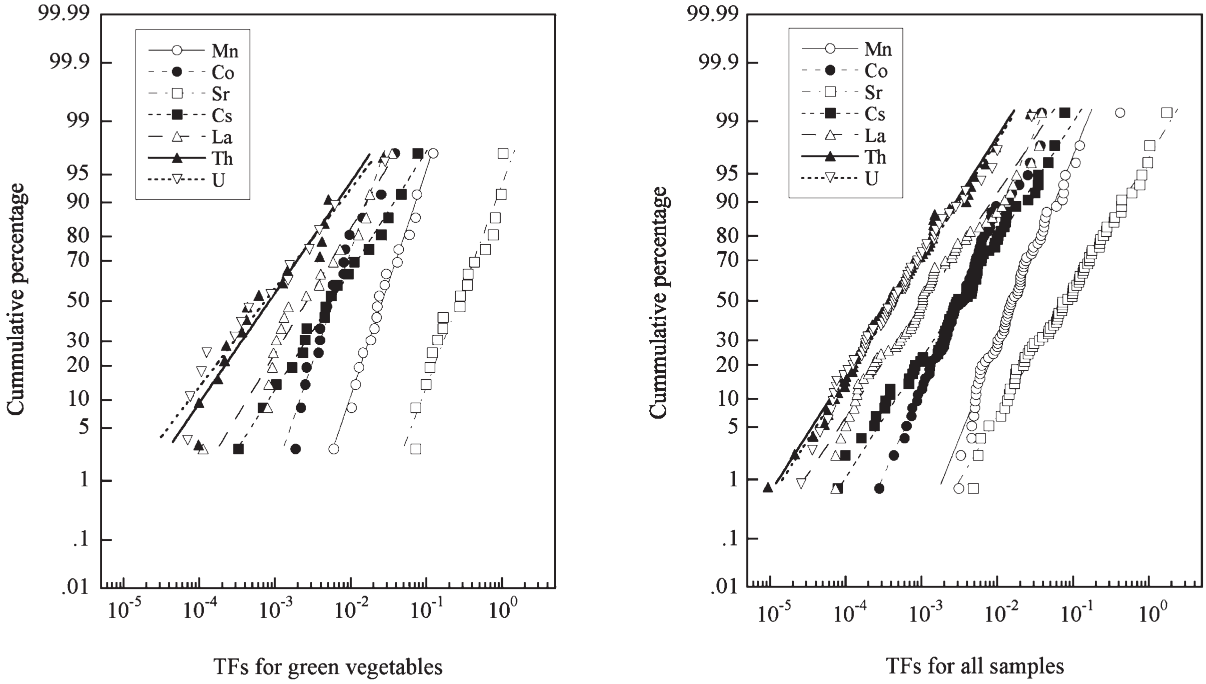

Fig. 4 Probability distributions of TFs (dry weight basis) of Mn, Co, Sr, Cs, La, Th and U for green vegetables (left) and all samples (right)

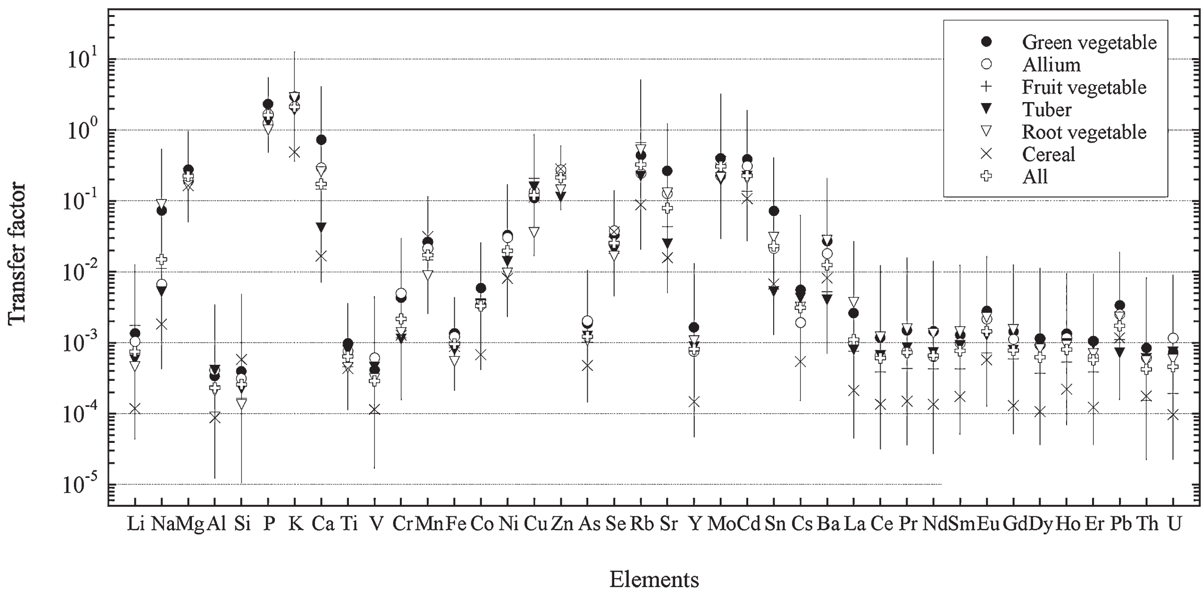

Fig. 5 Geometric means of TFs for 6 crop types and all samples (dry weight basis)
Bars show 95% upper and lower confidence limits for all samples.

The TF-GMs of green vegetables were usually the highest among the crops in the present study. However, when Co, Sr, Cs, and U were compared with TRS-364, the values were 1-2 orders of magnitude lower than the expected values and lower than the 95% confidence range. The TF-GMs of La, Pb, and Th were within the 95% confidence limits of TRS-364. For potatoes, the TF-GMs of Mn, Co, Sr, Cs, and U were, by about one order of magnitude, lower than the expected values. However, the TF-GMs of La, Pb and Th, 9.3E-04, 7.9E-04 and 7.2E-04, respectively, were close to or slightly higher than the expected values, 2.9E-04, 1.3E-03 and 5.6E-05, respectively. Tsukada and Nakamura[24] also reported on the TF-GMs of La and Th for potatoes (on wet weight basis); using dry/wet ratio of the crop, the values were calculated on dry weight basis and the results were 2.3E-04 and 1.3E-03, respectively. From these results, it was probable that the TF-GM of La observed in Japan had almost the same value as that in TRS-364, however, the TF-GM of Th was higher than that in TRS-364. Since only 6 samples were used in this study, the range of TFs was

Table 4 Transfer factors for four crops collected in Japan (dry weight basis)

Crops	Elements					Reference
	Mn	Co	Zn	Sr	Cs	
Cabbage	1.4E+00	8.1E-02	6.9E-01	8.1E-01	8.1E-01	Ban-nai *et al.*[23),a),c)]
	2.5E-02	7.0E-03	1.9E-01	2.3E-01	6.7E-03	Tsukada and Nakamura[24),b),c)]
	2.8E-02	4.3E-03	2.8E-01	5.3E-01	8.8E-03	Tsukada and Hasegawa[25),b)]
	2.9E-02	6.0E-03	2.0E-01	1.7E-01	3.3E-03	This study[b)]
Chinese cabbage	1.7E+00	1.1E-01	2.1E+00	9.4E-01	9.4E-01	Ban-nai *et al.*[23),a),c)]
	3.7E-02	6.5E-03	9.1E-01	4.2E-01	3.0E-02	Tsukada and Nakamura[24),b),c)]
	1.2E-02	3.3E-03	2.8E-01	5.0E-01	5.9E-03	This study[b)]
Japanese radish	4.0E-01	8.0E-02	8.0E-01	1.0E+00	4.0E-01	Ban-nai *et al.*[26),a),c)]
	1.2E-02	6.2E-03	8.4E-01	1.6E-01	9.8E-03	Tsukada and Nakamura[24),b),c)]
	1.0E-02	4.6E-03	1.5E-01	1.5E-01	2.8E-03	This study[b)]
Carrot	1.5E+00	3.3E-02	1.4E+00	8.0E-01	9.3E-02	Ban-nai *et al.*[26),a),c)]
	1.8E-02	2.4E-03	5.3E-01	9.6E-02	1.1E-02	Tsukada and Nakamura[24),b),c)]
	6.0E-03	1.3E-03	1.1E-01	8.2E-02	4.7E-03	This study[b)]

[a)]radiotracer experiment, [b)]field observation, [c)]data (dry) were calculated using dry/wet ratio from TF (wet) values.

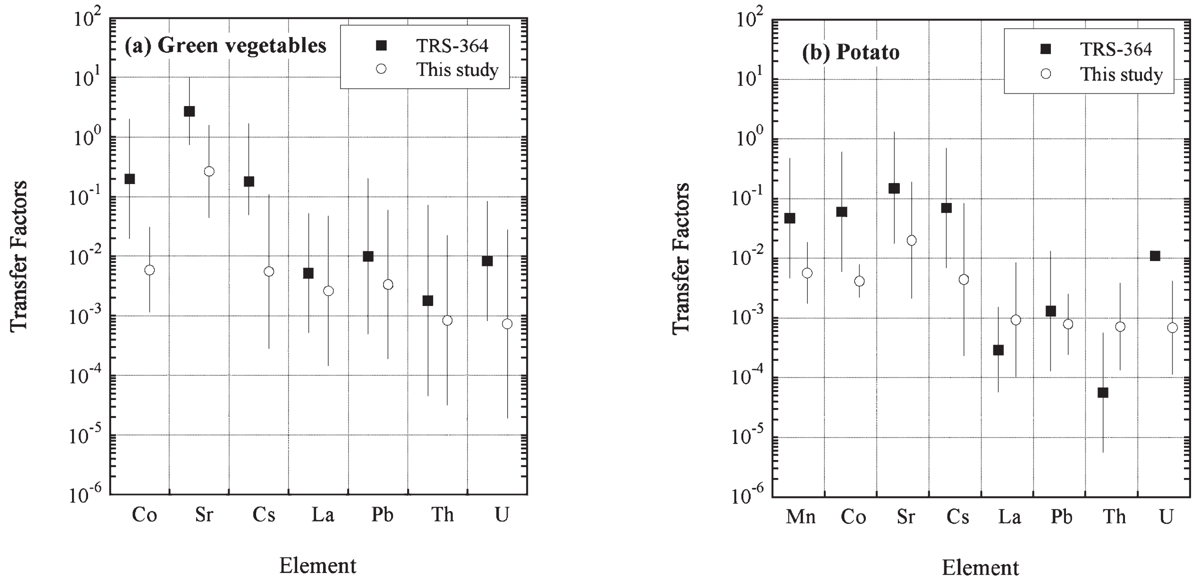

Fig. 6 Comparison of TFs of selected elements between compiled values in TRS-364[3)] and values obtained in this work for (a) green vegetables and (2) potatoes

The values from TRS-364 are the best estimates and the values from this study are geometric means of a maximum of 18 samples for green vegetables (cabbage, lettuce, *etc.*) and 6 samples for potatoes. Bars show 95% upper and lower confidence limits.

not clear yet; however, in TRS-364, La, Pb, Th, and U also have less than 10 data. Thus, more data are needed to understand the differences between climates in Japan and in the temperate areas like Europe.

IV. Conclusions

Crop and associated upland field soil samples were collected nationwide from 62 sampling sites in 2002–2004 in the harvesting season to obtain soil-to-plant transfer factors of stable and naturally occurring radionuclides under equilibrium conditions. The TF values (dry weight basis) of 40 elements in 68 edible parts of crops were presented in this study. Green vegetables showed the highest TFs for almost all the elements among the crop types studied. Then the TFs of selected elements for green vegetables and potato were compared with those of TRS-364. The TF-GMs of Co, Sr, Cs, Pb, and U were, by 1-2 orders of magnitude, lower than the expected values of TRS-364 for both green vegetables and potatoes, being lower than the 95% confidence range. The TF-GMs of La, Th for green vegetables were within the 95% confidence limit, however, those for potatoes were slightly higher than the expected values. Since numbers of samples were still small, it is necessary to collect more TF data in Japan. Further, there are radionuclides for which information on soil-to-plant transfer factors are still lacking due to analytical and methodological difficulties, *i.e.*, ^{3}H, ^{14}C, ^{36}Cl, ^{93}Zr, ^{93m,94}Nb, ^{99}Tc, ^{107}Pd, ^{108m}Ag, ^{129}I and ^{226}Ra. It is necessary to obtain the TF data for these nuclides or related elements as analogues in the future.

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