Transfer of Zirconium, Niobium and Molybdenum from Japanese Agricultural Fields to Edible Parts of Crops -11254

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ABSTRACT

Soil-to-plant transfer factors (TFs) of stable zirconium (Zr), niobium (Nb) and molybdenum (Mo) for 148 crop samples were determined with the intention to apply these values for those of Zr-93, Nb-94 and Mo-93 under physico-chemical equilibrium conditions. The TF of an element is defined as the concentration ratio of the element in a crop to the element in soil. The geometric means of the TFs for seven crop types (leafy vegetables, tubers, root crops, fruit vegetables, legumes, wheat and barley, and brown rice) had the following ranges: Zr, 4.3×10^{-5} (brown rice) - 5.8×10^{-4} (leafy vegetables): Nb, 1.5×10^{-4} (brown rice) - 1.0×10^{-3} (leafy vegetables) and Mo, 1.8×10^{-1} (tubers) - 7.8×10^{-1} (brown rice and legumes). The TFs of Zr and Nb were higher than the values reported in IAEA-TRS-472, however, almost the same values were observed for Mo. Additionally, the TF ranges of Zr, Nb and Mo for each crop grown in different sampling fields usually varied by one or two orders of magnitude. These TF values will be informative to understand the behaviors of the radionuclides in agricultural fields.

INTRODUCTION

There are many important radionuclides for radiation dose assessment from high-level radioactive waste disposal sites [1]; among them, soil-to-plant transfer factors (TFs) of zirconium-93 (Zr-93; half life: 1.5×10^{6} y), niobium-94 (Nb-94; half life: 2.03×10^{4} y), and molybdenum-93 (Mo-93; half life: 4×10^{3} y) were selected for the present study. TF data can be obtained using radiotracer experiments; the IAEA has recently compiled peer-reviewed published data and provided Technical Report Series No.472 (IAEA-TRS-472) [2] on such experiments. Only 1-3 data for each crop type were found for Zr, Nb and Mo, while there are hundreds of data for Cs and Sr. The limited numbers of data are possibly because of the difficulty in obtaining data using radiotracers of Zr, Nb and Mo or in the difficulty of handling and using these long-lived radionuclides themselves. With these small numbers of data, it is difficult to provide a potential TF range for various crop types grown on various kinds of soils so that uncertainty for radiation dose assessment increases. On the other hand, the long-half lives of Zr-93, Nb-94 and Mo-93 mean that their behaviors become almost the same as those of stable elements.

Since Zr and Nb are non-essential elements to plants and of less interest to biologists, their soil-to-plant uptake behaviors are unclear. In the natural environment, most soils contain stable Zr at several hundred mg/kg dry level, but its limited plant availability results in low Zr concentrations in crops [3], and thus TF data of Zr are not available. The situation for Nb is much the same as that for Zr. On the other hand, Mo is an essential element and its soil-to-plant uptake behavior has been well studied. Kabata-Pendias [3] has reviewed Mo uptake behavior by plants in which it was noted that although there were some reports of a linear relationship for Mo content in herbs and its total concentration in soil, there was no clear and simple relationship between the total Mo content in soil and plants. It was also noted that soil pH affected Mo availability to plants. Thus, it is difficult to obtain TF values using limited literature data for Zr, Nb and Mo. Additionally, because soils in Japan are generally acidic Mo data obtained in other countries is often not be applicable. In this study, therefore, TFs of stable Zr, Nb and Mo were measured under agricultural conditions, and their ranges were provided.

EXPERIMENTAL

Sample Collection and Pretreatment

Seventy-nine upland field and 63 paddy field soil samples (plowed soil layer: up to ca. 20 cm depth), were collected nationwide from 2002 to 2006. From each sampling field, 5 sub-samples, approximately 1 kg on fresh weight basis each, were collected in the harvesting season and these sub-samples were mixed well. About 15 kg amounts (on a fresh weight basis) of edible parts of crops were also collected. Crops were classified into 7 crop types, and the numbers of samples were: 26 green vegetable samples (cabbage, Chinese cabbage, spinach, lettuce, and so on); 11 tuber samples (potato, sweet potato and taro); 14 root crop samples (carrot, onion, and Japanese radish); 18 fruit vegetable samples (cucumber, tomato, sweet pepper, and so on); 7 legume samples (bean and pea); 9 cereal samples (wheat and barley); and 63 brown rice samples (hulled rice).

Three kg from ca. 5 kg of wet weight soil sample 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 to complete the removal of dust and soil particles. The washed parts were dried with paper towels, chopped and freeze-dried. Four leek samples were separated into green and white parts. Carrot and Japanese radish leaves are also edible, so that the roots and leaves were separated for one each of the carrot and Japanese radish samples. In all, 142 soil samples and 148 crop parts were obtained and they were separately and thoroughly ground into fine powders. The powders were transferred into glass vials and stored at room temperature until analyzed.

Measurements

Water content for air-dried soil was calculated by measuring weights before and after oven-drying (110ºC) the air-dried soil. The pH (H_2O) of the soils was measured at a soil:water ratio of 1:2.5. Cation exchange capacity (CEC), exchangeable Ca (ex.-Ca) and K (ex.-K) concentrations were determined by the semi-micro Schollenberger's method [4] using 1 mol/L of neutral ammonium acetate. Acid oxalate extractable Al (act.-Al) and Fe (act.-Fe) content were measured by the literature method [5]. The contents of total carbon and nitrogen were analyzed with a CHN analyzer (Euro Vector, EuroEA3000) using 10 mg of soil samples. Geometric mean values of these soil properties, except pH, are listed in Table I. For pH, average values are listed for different soil uses. For upland fields and paddy fields, ranges are also listed. No big difference was observed among the 142 agricultural fields in these soil properties.

To measure Zr, Nb and Mo concentrations, 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 crop samples, 500 mg amounts were used. The digestion samples were made in duplicate. All the acids used were ultra-pure analytical grade (Tama Chemicals, AA-100). Water (>18.1MΩ) which was treated using a Milli-Q water system (Millipore Co.) was used throughout the work. Standard reference materials, such as SRM-1573a (NIST, 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.

After diluting the acid solutions to a suitable concentration, Zr, Nb and Mo in crop samples were measured by inductively coupled plasma mass spectrometry (ICP-MS). For soil samples, Nb and Mo were measured by ICP-MS, but Zr was measured by energy dispersive X-ray fluorescence (EDXRF), which has recently been proposed as a non-destructive method for Zr in soil. Since Zr is contained in soil particles as zircon which is almost non-destructible using mineral acids, EDXRF and neutron activation analysis are the only two methods available to measure total Zr.

Table I. Soil properties (air-dried mass basis). Except for pH, the values are geometric mean values.

Soil use	Water content (%)	pH (H_2O)	CEC (cmol/kg)	ex.-Ca, (g/kg)	ex.-K (g/kg)	act.-Al (g/kg)	act.-Fe (g/kg)	Total C (g/kg)	Total N (g/kg)
Upland fields	4.9 (0.3-16.1)	6.3 (4.3-8.1)	14.1 (6-30)	2.5 (0.4-8.4)	0.44 (0.08-3.2)	6.2 (0.5-96)	7.6 (1.1-33)	28 (2-100)	2.7 (0.6-7.5)
Paddy fields	4.1 (0.9-18.8)	5.7 (4.8-6.9)	13 (5-27)	1.7 (0.6-7.9)	0.19 (0.06-0.59)	3.5 (0.7-55)	6.6 (1.5-19)	24 (14-125)	2.5 (1.4-10)

RESULTS AND DISCUSSION

Concentrations of Zr, Nb and Mo in Soil and Crops

Concentrations of Zr, Nb and Mo in upland field and paddy field soil samples showed a log-normal distribution (Fig.1) so that geometric means for different soil uses were calculated (Table II). When the results of upland field and paddy field soil samples were compared, no statistical difference was observed for Zr, but differences were observed for Mo and Nb, although the difference was small. The obtained results were within the ranges of world soil values [3].

The concentration results of Zr, Nb and Mo for crops are shown in box-and whisker plots (Fig.2). Among the crop types, leafy vegetables showed the highest value for Zr and Nb, and compared to these values, brown rice (Zr, Nb), root crops (Zr), fruit vegetables (Zr), and wheat and barley (Zr) showed significant differences. For Mo, legumes had the highest concentration, while tubers and root vegetables had significantly low values. Detailed concentrations are listed in Table III; when these concentrations were compared with world food plants values [3] (no data for Nb), the present data were almost within the range of the reported values.

Table II. Concentrations as geometric means of Zr, Nb and Mo in the soil samples.

Soil use	N	Zr, mg/kg-dry Geometric mean (Min.-Max.)	Nb, mg/kg-dry Geometric mean (Min.-Max.)	Mo, mg/kg-dry Geometric mean (Min.-Max.)
Upland fields	79	152 (33-247)	9.0 (2.7-18.6)	1.2 (0.3-3.4)
Paddy fields	63	167 (84-266)	10.9 (3.0-31.4)	1.0 (0.4-2.4)
World soil [3]		32-850	<4-300	0.013-17

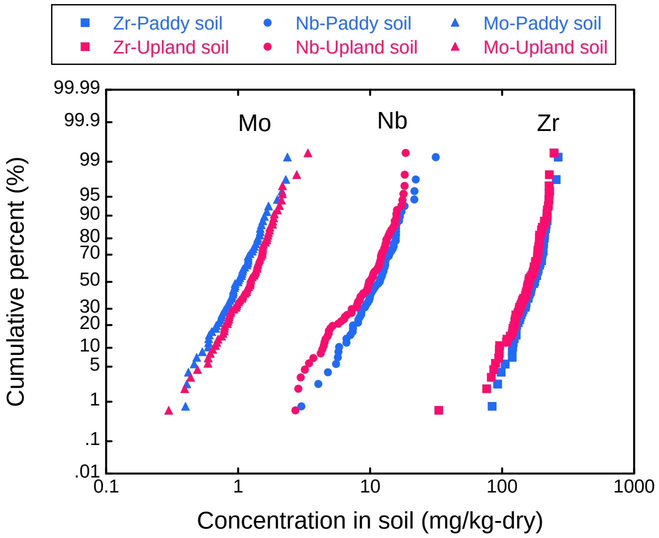

Fig.1. Probability distributions of Zr, Nb and Mo concentrations in upland and paddy field soil samples.

The concentration results of Zr, Nb and Mo for crops are shown in box-and whisker plot (Fig.2). Among the crop types, leafy vegetables showed the highest value for Zr and Nb, and compared to these values, brown rice (Zr, Nb), root crops (Zr), fruit vegetables (Zr), and wheat and barley (Zr) showed significant difference. For Mo, legumes showed highest concentration, while tubers and root vegetables showed significantly low values. Detailed concentrations are listed in Table III; when we compared these concentrations with world food plants values (no data for Nb) [3], the present data were almost within the range of the reported values.

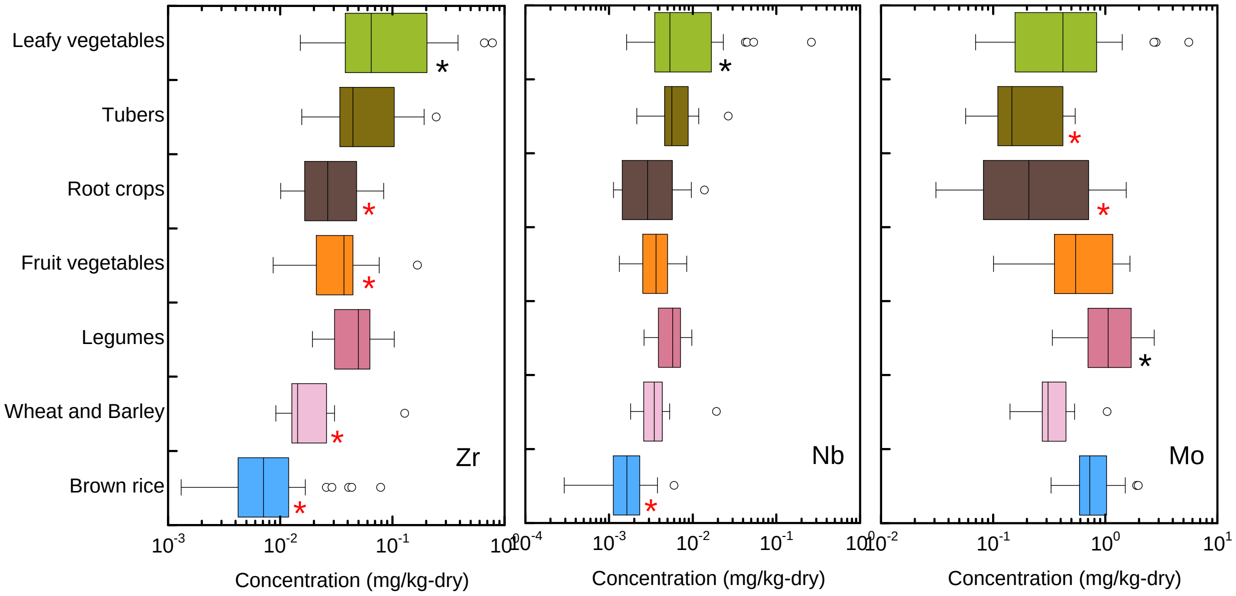

Fig.2. Box-and-whisker plots of Zr, Nb and Mo concentrations in crop samples. In each figure, the data followed by an asterisk (black or red) showed significant difference by the ANOVA test ($p<0.05$).

Table III. Concentrations of Zr, Nb and Mo in the soil samples.

Crop type	N	Zr, mg/kg-dry Geometric mean (Min.-Max.)	Nb, mg/kg-dry Geometric mean (Min.-Max.)	Mo, mg/kg-dry Geometric mean (Min.-Max.)
Leafy vegetables	26	$8.2x10^{-2}$ $(1.5x10^{-2}-7.8x10^{-1})$	$7.9x10^{-3}$ $(1.6x10^{-3}-2.6x10^{-1})$	$4.2x10^{-1}$ $(7.0x10^{-2}-5.6\ x10^{0})$
Tubers	11	$6.0x10^{-2}$ $(1.6x10^{-2}-2.5x10^{-1})$	$6.3x10^{-3}$ $(2.1x10^{-3}-2.7x10^{-2})$	$1.9x10^{-1}$ $(5.7x10^{-2}-5.4x10^{-1})$
Root crops	14	$3.0x10^{-2}$ $(1.0x10^{-2}-8.4x10^{-2})$	$3.3x10^{-3}$ $(1.1x10^{-3}-1.4x10^{-2})$	$2.1x10^{-1}$ $(3.1x10^{-2}-1.5x10^{0})$
Fruit vegetables	18	$3.3x10^{-2}$ $(8.7x10^{-2}-1.7x10^{-1})$	$3.5x10^{-3}$ $(1.3x10^{-3}-8.5x10^{-3})$	$5.7x10^{-1}$ $(1.0x10^{-1}-1.7x10^{0})$
Legumes	7	$4.5x10^{-2}$ $(1.9x10^{-2}-1.0x10^{-1})$	$5.3x10^{-3}$ $(2.6x10^{-3}-9.8x10^{-3})$	$1.0x10^{0}$ $(3.4x10^{-1}-2.7x10^{0})$
Wheat and Barley	9	$2.0x10^{-2}$ $(9.0x10^{-3}-1.3x10^{-1})$	$3.9x10^{-3}$ $(1.8x10^{-3}-1.9x10^{-2})$	$3.5x10^{-1}$ $(1.4x10^{-1}-1.0x10^{0})$
Brown rice	63	$7.1x10^{-3}$ $(1.3x10^{-3}-7.9x10^{-2})$	$1.6x10^{-3}$ $(2.9x10^{-4}-6.0x10^{-3})$	$7.6x10^{-1}$ $(3.3x10^{-1}-2.0x10^{0})$
World food plants [5]		$5x10^{-3}-2.6x10^{0}$		$7x10^{-2}-2.1x10^{0}$

Soil-to-plant Transfer Factors of Zr, Nb and Mo

Soil-to-plant transfer factor (TF) of an element is defined as follows.

TF = element concentration in plant (mg/kg-dry) / element concentration in soil (mg/kg-dry)

The TF values in 7 crop types are plotted in Fig.3. For Zr and Nb, the highest TFs were observed in leafy vegetables, and compared to them, root crops, fruit vegetables, wheat and barley, and brown rice showed significant differences. No differences were observed for TFs of Mo (TFs-Mo) in the crop types. From the figure, similar distribution tendencies for TFs of Zr (TFs-Zr) and Nb (TFs-Nb) were found. Thus they were compared and the results are shown in Fig.4. Values of TF-Zr and TF-Nb were well correlated with a correlation factor of 0.86 ($p<0.001$) by Student's t-test. Thus TF value could be estimated if one of them has been measured, but the other has not. Indeed, Coughtrey and Thorne [6] reported the TF of Nb based on the assumption of its similar behavior with Zr.

The TF values of Zr, Nb and Mo were distributed as a log-normal type (data are not shown) so that a geometric mean for each crop type was calculated. The results, including ranges, are listed in Table IV. For Zr, geometric means of TF ranged from $4.3x10^{-5}$ (brown rice) to $5.8x10^{-4}$ (leafy vegetables) and these were lower than the values reported in IAEA-TRS-472 (range: $1.0x10^{-3}$ to $1.0x10^{-2}$). Since the non-destructive fraction in the soil particles was also measured, the TF values would be lower than those from radiotracer experiments: freshly added radiotracer and native stable element are not at equilibrium so that the TF of a radiotracer obtained under greenhouse conditions is usually higher than the TF observed in the environment. However, Tsukada and Nakamura [7] measured stable Zr using neutron activation analysis; interestingly, their results were one to two orders of magnitude higher than the present results although they also measured non-destructive fraction in the soil particles. When the number of crop samples they measured in total and the number of detectable samples were compared, it was seen that the former was less than 30% of the total number of samples. Because the detectable samples would have high Zr concentrations, the TF-Zr value by Tsukada and Nakamura was slightly higher than the present results.

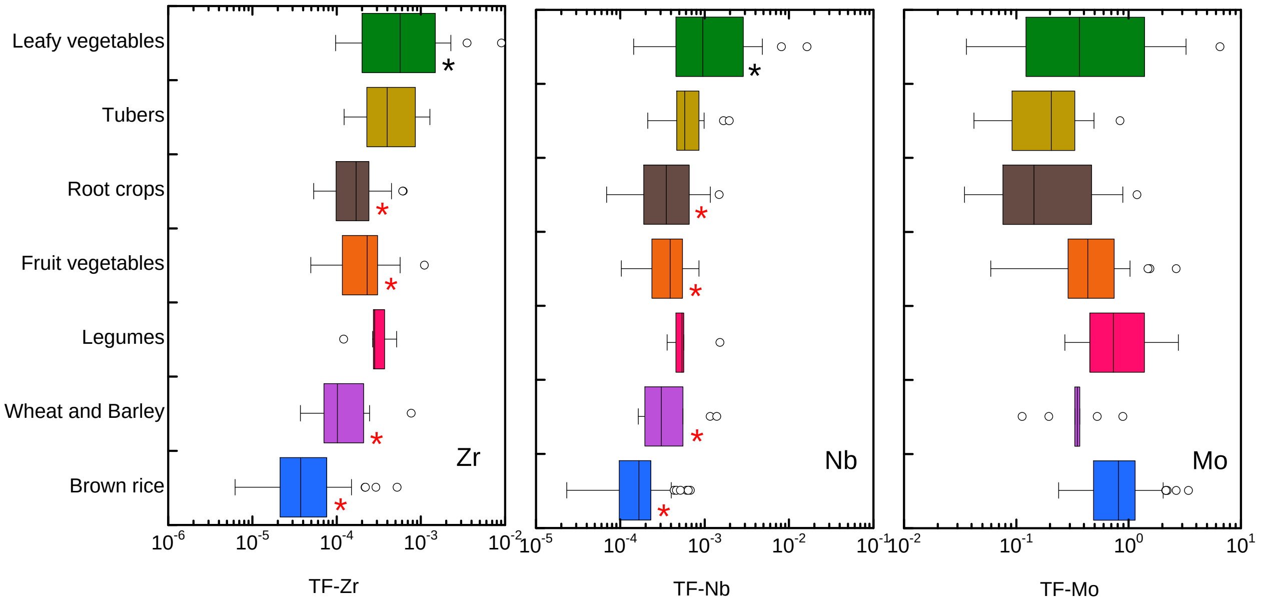

Fig.3. Box-and-whisker plots of TFs of Zr, Nb and Mo in crop samples. In each figure, the data followed by an asterisk (black or red) showed significant difference by the ANOVA test ($p<0.05$).

Fig.4. Correlations between TF-Zr and TF-Nb (in logarithm values).

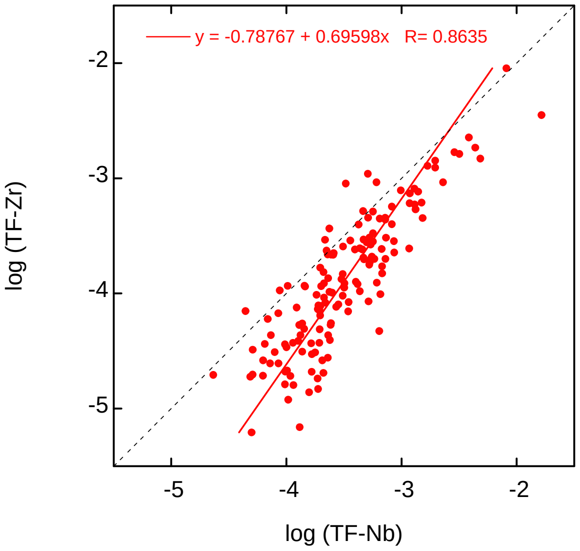

Table IV. Soil-to-crop transfer factors of Zr, Nb, and Mo on dry mass basis.

Crop type	TF-Zr Geometric mean (Min.-Max.)	TF-Nb Geometric mean (Min.-Max.)	TF-Mo Geometric mean (Min.-Max.)	Ref.
Leafy vegetables	5.8x10^{-4}, N=26 (9.7x10^{-5}-9.0x10^{-3})	1.0x10^{-3}, N=26 (1.4x10^{-4}-1.6x10^{-2})	3.6x10^{-1}, N=26 (3.6x10^{-2}-6.5x10^{0})	This study
	4.0x10^{-3}, N=1	1.7x10^{-2}, N=2	5.1 x10^{-1}, N=1	[2]
Cabbage	2.7x10^{-4}, N=6 (9.7x10^{-5}-7.4x10^{-4})	4.6 x10^{-4}, N=6 (1.8x10^{-4}-1.3x10^{-3})	3.9x10^{-1}, N=6 (3.6x10^{-2}-3.2x10^{0})	This study
Cabbage	1.5x10^{-2}, N=2		1.1x10^{-1}, N=2	[7]
Tubers	4.2x10^{-4}, N=11 (1.2x10^{-4}-1.3x10^{-3})	6.3x10^{-4}, N=11 (2.1x10^{-4}-2.0x10^{-3})	1.8x10^{-1}, N=11 (4.2x10^{-2}-8.4x10^{-1})	This study
	2.0x10^{-3}, N=1	4.0x10^{-3}, N=2	3.2 x10^{-1}, N=3	[2]
Potato	4.2x10^{-4}, N=6 (1.2x10^{-4}-1.3x10^{-3})	5.7x10^{-4}, N=6 (2.1x10^{-4}-1.3x10^{-3})	1.6x10^{-1}, N=6 (6.5x10^{-2}-3.5x10^{-1})	This study
Potato	8.0x10^{-3}, N=6		3.3x10^{-2}, N=16	[7]
Root crops	1.7x10^{-4}, N=14 (5.3x10^{-5}-6.2x10^{-4})	3.6x10^{-4}, N=14 (6.9x10^{-5}-1.5x10^{-3})	1.9x10^{-1}, N=14 (3.5x10^{-2}-1.2x10^{0})	This study
	4.0x10^{-3}, N=1	1.7x10^{-2}, N=2		[2]
Japanese radish	2.1x10^{-4}, N=7 (6.0x10^{-5}-6.2x10^{-4})	4.2x10^{-4}, N=7 (6.9x10^{-5}-1.5x10^{-3})	3.2x10^{-1}, N=7 (8.4x10^{-2}-1.2x10^{0})	This study
Japanese radish	3.4x10^{-2}, N=4		5.0x10^{-2}, N=1	[7]
Fruit vegetables	2.1x10^{-4}, N=18 (4.9x10^{-5}-1.1x10^{-3})	3.7x10^{-4}, N=18 (1.0x10^{-4}-8.6x10^{-4})	4.5x10^{-1}, N=18 (5.9x10^{-2}-2.7x10^{0})	This study
Tomato	1.5x10^{-4}, N=6 (4.9x10^{-5}-4.0x10^{-4})	2.5x10^{-4}, N=6 (1.0x10^{-4}-8.2x10^{-4})	2.3x10^{-1}, N=6 (8.2x10^{-2}-5.0x10^{-1})	This study
Tomato	2.0x10^{-2}, N=5		6.2x10^{-2}, N=15	[7]
Legumes	2.9x10^{-4}, N=7 (1.2x10^{-4}-5.2x10^{-4})	5.7x10^{-4}, N=7 (3.6x10^{-4}-1.5x10^{-3})	7.8x10^{-1}, N=7 (2.7x10^{-1}-2.8x10^{0})	This study
Wheat and Barley	1.3x10^{-4}, N=9 (3.7x10^{-5}-7.7x10^{-4})	3.9x10^{-4}, N=9 (1.6x10^{-4}-1.4x10^{-3})	3.4x10^{-1}, N=9 (1.1x10^{-1}-8.9x10^{-1})	This study
Brown rice	4.3x10^{-5}, N=63 (6.2x10^{-6}-5.2x10^{-4})	1.5x10^{-4}, N=63 (2.3x10^{-5}-6.8x10^{-4})	7.8x10^{-1}, N=63 (2.4x10^{-1}-3.4x10^{0})	This study
Cereals (excl. rice)	1.0x10^{-3}, N=1	1.4x10^{-2}, N=2	8.0 x10^{-1}, N=1	[2]

For Nb, geometric means of TF ranged from $1.5x10^{-4}$ (brown rice) to $1.0x10^{-3}$ (leafy vegetables), while those listed in TRS-472 range from $4.0x10^{-3}$ to $1.7x10^{-2}$. When geometric means of TFs complied by IAEA and the present study were compared, the differences were one or two orders of magnitude, and the latter results were usually lower, possibly because the non-destructive portion in soil was included in the present results. Coughtrey and Thorne [6] estimated TF-Nb for unspecified crops as $2.5x10^{-2}$ using TF-Zr values as mentioned above. Other literature values for TF-Nb are $1.4x10^{-3}$ to $2.5x10^{-3}$ for rape, $6.9x10^{-3}$ to $1.3x10^{-2}$ for bean shoot and $<1.2x10^{-3}$ to $1.7x10^{-3}$ for bean pod [8]. When the bean pod data were compared with the present legume data ($3.6x10^{-4}$ -$1.5x10^{-3}$), they are almost the same.

Molybdenum is an essential element for plants; plants selectively absorb Mo from soil and thus TFs will be higher than those of Zr or Nb. The TF-Mo data range was smaller than the ranges for Zr and Nb, i.e. geometric means of TF in the present study ranged from $1.8x10^{-1}$ (tubers) to $7.8x10^{-1}$ (brown rice and legumes), and recommended values listed in TRS-472 range from $3.2x10^{-1}$ to $8.0\ x10^{-1}$. There is no big difference between TF-Mo values of the stable element observed in this study and those of radiotracer experiments compiled in IAEA-TRS-472. Thus it was concluded that TFs-Mo observed in the field using the stable element as an analogue can be applied for radioactive Mo. Other stable element measurement results by Tsukada and Nakamura [7] agreed well with the present results.

The TF data used in IAEA-TRS-472 for a temperate area were compiled from radiotracer experiments; thus, the observation period would be short and in soil these radionuclides may not have reached a physico-chemical equilibrium. However, for radiation dose assessment from high-level radioactive waste disposal sites, data taken under physico-chemical equilibrium conditions are more realistic; it is expected that radionuclides released from the sites and stable elements initially present in the environment will show the same behavior in the far future. Thus TFs obtained from stable elements will be useful for dose assessment, although more data collection on stable elements and data comparisons with radiotracer studies are needed.

CONCLUSION

Among important radionuclides for radiation dose assessment from high-level radioactive waste disposal sites, Zr-93, Nb-94, and Mo-93 were studied because TF data for these nuclides, based on radiotracer experiments, are very limited and TF ranges for various crops are unclear. Stable Zr, Nb and Mo were used as analogues of the radionuclides of interest and 148 samples of edible crop parts were analyzed to get TFs from which potential TF values and ranges of these radionuclides in agricultural fields could be determined. For Zr, the TFs ranged from $4.3x10^{-5}$ to $5.8x10^{-4}$, and the values were one or two orders of magnitude lower than values in IAEA-TRS-472 [2] because Zr contents in the non-destructive soil part were included in the present study. For Nb, the TFs ranged from $1.5x10^{-4}$ to $1.0x10^{-3}$. Although the values were also smaller than those in IAEA-TRS-472, legumes had values similar to previously reported values for bean pod as obtained by radiotracer experiments [8]. There is a high correlation between TFs of Zr and Nb, thus TF of one of them could be assumes as the TF of the other. For Mo, the TFs ranged from $1.8x10^{-1}$ to $7.8x10^{-1}$ and the range was the same as of IAEA-TRS-472 and Tsukada and Nakamura [7]. TFs of stable Mo can be used for those of radioactive Mo without any additional considerations; however, for stable Zr and Nb, it is necessary to provide a method to get their readily plant-available fractions in soil to use the TFs of stable Zr and Nb as analogues of their radionuclides.

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