Measurement of Stable Iodine in Crops and Soils as an Analogue of Iodine-129 -10347

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ABSTRACT

Long-life iodine-129 (half-life: 1.57×10^7 y) is the most critical radionuclide to be managed for the safe disposal of nuclear fuel waste. Its fate in the environment would be the same as that of stable iodine (I). Thus we measured stable I as an analogue of I-129 especially focusing on the soil-to-plant transfer in agricultural systems. Stable I concentrations were measured in 142 crop samples and associated agricultural field soil samples collected throughout Japan. We also measured bromine (Br) for comparison. The average I/Br concentration ratio in agricultural soil samples was about 0.5, while that in crop samples was about 0.01; the difference was possibly due to the different mobilities of these elements in soil as well as the different plant root uptake behaviors. We also calculated soil-to-crop transfer factors (TFs) of 25 crop species/parts. The highest TF for I, 0.48, was obtained for lettuce, and the lowest TF was 4.5×10^{-4} for brown rice. The TF data from stable I observation for crops collected in agricultural fields were close to or within those of radiotracer experiments so that we judged the data would be applicable for dose estimation from I-129.

INTRODUCTION

Iodine-129 (half-life: 1.57×10^7 y) is the most limiting and the most critical radionuclide in both transuranic and high level radioactive waste management. Since stable iodine (I) can be used as a natural analogue of I-129, information about the distribution and cycling of stable I in the environment is highly useful for estimating the dose from I-129. Stable I behavior itself is also interesting because it is an essential element for humans and other animals, and its suitable levels in food are necessary for the former to avoid I deficiency disorders and excess intake.

In this study, we focused on the behavior of I in soil-to-crop systems using bromine (Br) for comparison. The soil-to-plant transfer factor (TF) is an important parameter because a significant exposure pathway to humans is ingestion of contaminated food. The obtained TF values were compared with previously reported values such as in the recently published IAEA TECDOC-1616 [1].

MATERIALS AND METHODS

Surface layer (0-20 cm) soil samples from 79 upland fields and 63 paddy fields were collected nationwide in Japan from 2002 to 2006 during the harvesting season. Edible parts from crops grown in these agricultural fields were also collected at the same time. For 4 leek plants, green and white parts were separated as individual samples. For Japanese radish and carrot samples, leaves are also edible parts so that the roots and leaves were separated and elemental compositions were measured for both parts in one sample of each crop. Table I lists the crop groups and parts. In total, 148 crop part samples together with the associated soil samples were used in this analysis.

Table I. Concentrations of Br and I in Soil and Crop Samples Collected in Japan.

Crop name		N	Soil		Crop	
			Br, mg/kg-dry	I, mg/kg-dry	Br, mg/kg-dry	I, mg/kg-dry
			GM/AM (Min. – Max)	GM/AM (Min. – Max)	GM/AM (Min. – Max)	GM/AM (Min. – Max)
Leafy vegetables	Cabbage	7	7.4 (1.6 – 43.6)	4.8 (1.3 – 37.4)	7.0 (2.1 – 26.6)	0.030 (0.012 – 0.115)
	Chinese cabbage	4	12.1 (4.9 – 45.8)	6.9 (3.0 – 15.4)	11.0 (6.8 – 19.6)	0.067 (0.033 – 0.182)
	Leek (green part)	4	21.9 (12.1 – 60.0)	5.0 (2.6 – 9.5)	8.8 (5.7 – 16.1)	0.094 (0.032 – 0.224)
	Spinach	2	14.2 (4.8 – 23.6)	8.0 (2.5 – 13.5)	26.1 (18.4 – 33.8)	0.122 (0.109 - 0.135)
	Lettuce	2	19.6 (5.2 – 34.0)	11.0 (1.7 – 20.3)	19.0 (9.9 – 28.2)	0.546 (0.272 – 0.820)
	Nozawana	1	5.6	5.3	20.0	0.123
	Carrot, leaves	1	6.2	4.4	85.9	0.362
	Japanese radish, leaves	1	74.8	29.6	44.4	0.371
Root crops	Japanese radish	7	33.6 (4.7 – 159)	15.0 (2.2 – 65.5)	21.1 (7.6 – 72.4)	0.082 (0.022 – 1.54)
	Onion	4	4.3 (2.7 – 7.4)	2.2 (1.5 – 6.1)	3.3 (1.8 – 5.2)	0.026 (0.014 – 0.053)
	Carrot	3	22.4 (6.2 -54.3)	9.3 (4.4 – 16.7)	15.4 (10.2 – 22.4)	0.032 (0.015 – 0.068)
Tubers	Potato	6	12.2 (2.0 – 49.8)	8.1 (2.7 – 37.1)	11.6 (2.8 – 44.1)	0.039 (0.013 – 0.089)
	Sweet potato	3	9.0 (0.3 – 74.7)	3.5 (0.2 – 21.8)	5.7 (3.3 – 14.6)	0.027 (0.015 – 0.062)
	Taro	2	9.5 (4.2 – 14.7)	2.2 (1.7 – 2.7)	5.4 (2.1 – 8.6)	0.063 (0.013 – 0.113)
Fruit vegetables	Tomato	6	10.0 (4.2 – 44.2)	3.8 (2.4 – 13.9)	10.0 (4.8 – 15.2)	0.037 (0.016 – 0.079)
	Egg plant	5	7.8 (2.1 – 46.6)	2.8 (0.8 – 19.9)	32.9 (11.3 – 1410)	0.054 (0.039 – 0.108)
	Cucumber	4	18.9 (10.8 – 35.2)	6.8 (2.1 – 22.8)	32.6 (22.5 – 47.2)	0.127 (0.079 – 0.263)
	Sweet pepper	2	88.1 (57.0 – 119)	24.9 (10.5 – 39.3)	24.5 (4.4 – 44.6)	0.107 (0.064 – 0.150)
	Bitter cucumber	1	2.8	1.8	7.0	0.109
Legumes	Soybean	6	8.3 (2.2 – 27.5)	4.6 (1.6 – 11.6)	3.7 (2.8 – 5.3)	0.030 (0.013 – 0.085)
	Peanut	1	80.7	31.0	1.6	0.020
Cereals	Brown rice	63	4.4 (1.1 – 42.7)	1.9 (0.8 – 10.1)	0.9 (0.4 – 2.7)	0.008 (0.003 – 0.019)
	Wheat	7	8.0 (2.6 – 103)	3.8 (1.4 – 41.9)	1.1 (0.7 – 3.2)	0.022 (0.010 – 0.046)
	Barely	2	11.8 (4.1 – 19.4)	6.5 (2.6 – 10.4)	3.0 (2.3 – 3.7)	0.015 (0.014 – 0.015)
	Leek (white part)	4	21.9 (12.1 – 60.0)	5.0 (2.6 – 9.5)	4.1 (3.6 – 4.6)	0.020 (0.013 – 0.041)

A simple extraction method was applied to measure stable I and Br in soil and plant samples by inductively coupled plasma mass spectrometry [2]. The same method is also applicable to Br concentration measurements. For soil samples, energy dispersive X-ray fluorescence (Epsilon 5, PANalytical B. V.) was used as described before [3].

RESULTS AND DISCUSSION

Total Br and I concentrations in crop and associated soil samples are shown in Table I. Geometric mean (GM) was calculated if the sample numbers for the crop was more than 3 and arithmetic mean (AM) was calculated for the crops having two samples. A single value was also listed when only one sample per crop was obtained. The Br and I concentration ranges were 0.32-159 and 0.16-65 mg/kg-dry. If we compared the Br and I concentrations in soil for different agricultural uses, that is, paddy fields and upland fields, upland fields soil samples were significantly higher for both elements.

Since the main source of Br and I in the terrestrial environment is seawater, I/Br concentration ratios were compared in soil and crop samples (Fig.1A) to elucidate how these elements went from the sea through the terrestrial environment. We also plotted I/Br in 45 major rivers in Japan [4] as well as that in rainwater samples collected in Japan [5, 6]. The I/Br ratios in soil samples for different agricultural uses showed no difference, and those in crops grown on the paddy fields (brown rice) and upland field crops were the same, however, the I/Br ratios were significantly different in soil and crop; the average I/Br ratio in soil samples was about 0.5 and that in crop samples was about 0.01. Among the crop samples, I/Br ratios were significantly different by ANOVA test ($p<0.001$) as shown in Fig.1B. I/Br ratios for wheat samples were significantly higher than other crops except Chinese cabbage, green part of leek, and onion samples, although except wheat samples, no difference was observed. Thus I/Br ratios were almost the same for most crops.

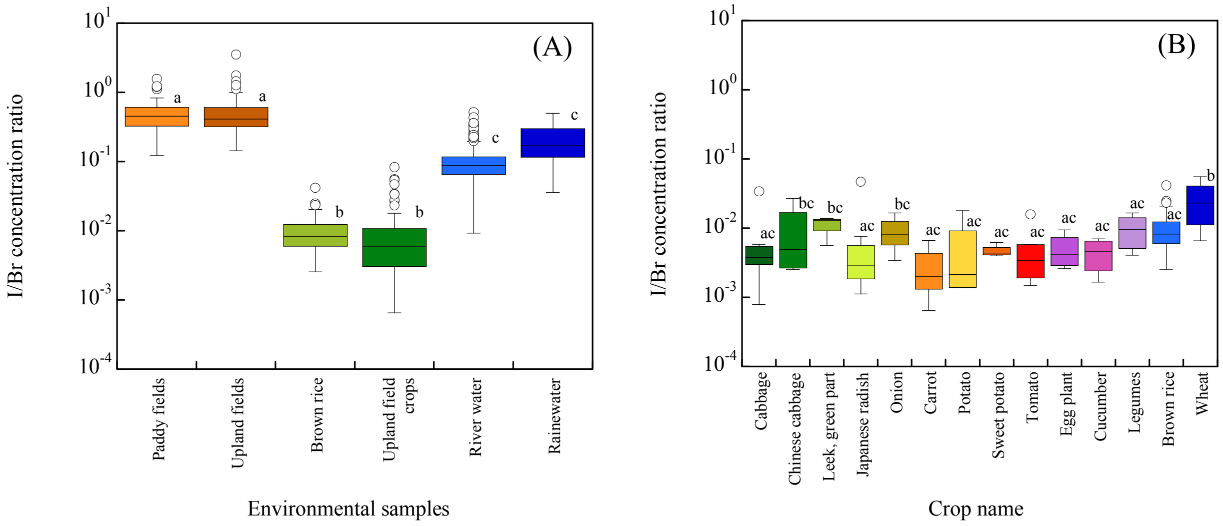

Fig.1. Box-and-whisker plot of I/Br concentration ratios in (A) environmental samples and (B) crop samples. In each figure, the data followed by the same letter are not significantly different ($p<0.05$).

Rainwater could be one source for I and Br in agricultural soils and the rainwater samples collected in Japan had a lower I/Br ratios than that in the soil samples. Also, the I/Br ratios in river water samples were slightly lower than those in the rainwater samples, although no statistical difference was observed by t-test. Since the water path is thought to be rain-soil-river water, the I/Br increase in soil and decrease in river water as compared to the rainwater ratio means longer I retention in soil. Thus the mobilities of Br and I would be different in the Japanese agricultural soils. Moreover, we found that Br and I uptake mechanisms by plant roots would be different because Br and I concentrations were well correlated in the soil samples but no correlation was observed in the samples of cereals. These factors would affect the 50-fold differences in I/Br ratios between soil and crop samples.

The observed Br and I concentrations in crop and soil samples were used for TF calculations. The TF is defined as the plant/soil concentration ratio (on dry weight basis). The results are listed in Tables II to IV with comparison to previously reported values [7-13]. Table II shows TFs for leafy vegetables. The TFs of I observed in this study were similar to those of previously reported values for stable I in the agricultural field observations [7] and radiotracer experiments [8, 9]. The GM of TFs of I compiled by IAEA [1] was slightly lower than the reported values, though the difference was small.

Table II. Transfer Factors of Br and I in Leafy Vegetable Samples.

Crop name	N	Br	I	Ref.
		GM/AM (Min. – Max)	GM/AM (Min. – Max)	
Cabbage	Br: 15 I: 19	3.6E-1 [a]	5.0E-3 [a]	[7]
Cabbage	7	7.7E-1 (4.8E-1 – 1.1E+0)	3.6E-3 (9.8E-4 – 8.1E-3)	This study
Chinese cabbage	Br: 7 I: 11	1.7E+0 [a]	1.5E-2 [a]	[7]
Chinese cabbage	4	9.1E-1 (2.1E-1 – 4.0E+0)	9.7E-3 (4.3E-3 – 2.7E-2)	This study
Onion, leaves	6		4.5E-2±2.9E-2 [b, c]	[8]
Leek (green part)	4	4.0E-1 (9.5E-2 – 8.9E-1)	1.9E-2 (3.4E-3 – 4.8E-2)	This study
Spinach	2	2.3E+0 (1.4E+0 – 3.8E+0)	2.1E-2 (8.1E-3 – 5.5E-2)	This study
Lettuce	6		6.7E-3±3.2E-3 [b]	[8]
Lettuce	14		9.4E-2 [a, b] (2.1E-2 – 7.6E-1)	[9]
Lettuce	2	1.3E+0 (8.3E-1 – 1.9E+0)	8.1E-2 (1.3E-2 – 4.9E-1)	This study
Nozawana	1	3.6E+0	2.3E-2	This study
Carrot, leaves	1	1.4E+1	8.2E-2	This study
Radish, leaves	6		1.2E-1±0.2E-1 [b, c]	[8]
Japanese radish, leaves	1	5.9E-1	1.3E-2	This study
Japanese parsley	6		2.4E-1±0.3E-1 [b, c, d]	[8]
Leafy vegetables	12		6.5E-3 [b] (1.1E-3 – 1.0E-1)	[1]

[a] Converted from wet weight basis to dry weight basis using wet/dry ratios. [b] Radiotracer experiments. [c] Arithmetic mean. [d] Wet weight basis.

The results of TFs for root crops and tubers are listed in Table III. The same trend was observed in root crops and tubers, however, the TFs of I for tubers by IAEA was 1-2 orders of magnitude higher than the reported values but there was only one compiled datum. It has been reported that TF for stable I was lower than that for ^{129}I due to different sources and mobilities in soil [14]. This might affect the differences, however, the TF data for stable and radioactive I were the same as mentioned above except for this datum by IAEA. More data would be necessary for tubers to obtain suitable data.

Table III. Transfer Factors of Br and I in Root Crop and Tuber Samples.

Crop name	N	Br	I	Ref.
		GM/AM (Min. – Max)	GM/AM (Min. – Max)	
Root crops				
Japanese radish	Br: 12 I: 13	1.1E+0 [a]	1.1E-2 [a]	[7]
Radish	6		5.1E-2±1.3E-2 [b, c]	[8]
Turnip	6		1.3E-2±0.6E-2 [b, c]	[8]
Radish	33		1.3E-1 [a, b] (2.8E-2 – 9.4E-1)	[9]
Japanese radish	7	4.5E-1 (1.1E-1 – 8.6E-1)	3.5E-3 (2.0E-3 – 6.1E-3)	This study
Garlic	Br: 8 I: 13	3.6E-2 [a]	5.3E-3 [a]	[7]
Onion	4		1.1E-2±0.4E-2 [b, c]	[8]
Onion	4	7.6E-1 (5.4E-1 – 1.5E+0)	1.2E-2 (5.4E-3 – 2.9E-2)	This study
Carrot	Br: 9 I: 9	5.7E-1 [a]	5.1E-3 [a]	[7]
Carrot	3	6.9E-1 (3.1E-1 – 2.6E+0)	3.4E-3 (8.7E-4 – 7.2E-3)	This study
Root crops	28		7.7E-3 [b] (1.4E-3 – 4.7E-2)	[1]
Tubers				
Potato	Br: 20 I: 26	1.3E-1 [a]	3.5E-3 [a]	[7]
Potato	6	9.5E-1 (3.6E-1 – 2.5E+0)	4.8E-3 (1.6E-3 – 1.3E-2)	This study
Sweet potato	3	6.3E-1 (1.1E-1 – 1.2E+1)	7.6E-3 (9.4E-4 – 9.4E-2)	This study
Yam	Br: 4 I: 4	2.2E-1 [a]	9.3E-4 [a]	[7]
Taro	2	5.4E-1 (5.0E-1 – 5.9E-1)	1.8E-2 (7.8E-3 – 4.2E-2)	This study
Tubers	1		1.0E-1 [b]	[1]

[a] Converted from wet weight basis to dry weight basis using wet/dry ratios. [b] Radiotracer experiments. [c] Arithmetic mean.

Table IV lists fruit vegetable and leguminous group crops. Although previously reported TFs of I for leguminous crops were almost the same as those of the present study, for fruit vegetables, the TF of I by IAEA [1] was 1-2 orders of magnitude of higher and the same trend as observed for tubers. At least, however, the TFs of I obtained in this study was within the error of previously reported values [7, 8].

Table IV. Transfer Factors of Br and I in Fruit Vegetables and Leguminous Samples.

Crop name	N	Br	I	Ref.
		GM/AM (Min. – Max)	GM/AM (Min. – Max)	
Fruit vegetables				
Tomato	Br: 15 I: 19	2.0E-1 [a]	1.4E-2 [a]	[7]
Tomato	6	1.0E+0 (3.4E-1 – 2.1E+0)	9.7E-3 (2.3E-3 – 3.2E-2)	This study
Egg plant	18		9.5E-4±2.9E-4 [b, c]	[8]
Egg plant	5	4.2E+0 (5.0E-1 – 3.0E+1)	1.9E-2 (2.2E-3 – 1.4E-1)	This study
Cucumber	Br: 7 I: 11	3.6E-1 [a]	2.6E-2 [a]	[7]
Cucumber	4	1.7E+0 (1.1E+0 – 3.4E+0)	1.9E-2 (8.7E-3 – 3.7E-2)	This study
Sweet pepper	2	1.7E-1 (7.8E-2 – 3.7E-1)	4.8E-3 (1.6E-3 – 1.4E-2)	This study
Bitter cucumber	1	2.5E+0	5.9E-2	This study
Fruit vegetables	1		1.0E-1 [b]	[1]
Legumes				
Green asparagus string beans - pods	8		3.0E-3 [a, b] (0.1E-2 – 1.9E-2)	[9]
Soybean	6	4.5E-1 (1.8E-1 – 1.6E+0)	6.5E-3 (1.3E-3 – 4.2E-2)	This study
Peanut	1	2.0E-2	6.3E-4	This study
Legume			8.5E-3, n=23 [b] (2.0E-4 – 1.4E-1)	[1]

[a] Converted from wet weight basis to dry weight basis using wet/dry ratios. [b] Radiotracer experiments. [c] Arithmetic mean.

The TF data for cereals are shown in Table V. The GM/AM data of TFs of I for white rice and brown rice were almost the same order of magnitude for all the data including ours. For wheat samples, our data were about one order of magnitude higher than those from radiotracer experiments, although TF data for Br were almost the same as those for white rice and brown rice. One possible explanation for this would be the I variation in the upland field soils due to different soil characteristics. Indeed, Kashparov et al. [9] found transfer differences in different soil types and pointed out the soil had various sorption abilities of the soils relative to I. Thus I mobility in soil was judged the key parameter for I uptake by plants and it could be expressed by soil-soil solution distribution coefficients, K_d and other indices such as water soluble I concentrations in soil samples. It would be desirable to collect not only soil characteristics but also K_d values for further explanation of the slightly higher TFs of I for wheat and barley grains in Japanese agricultural fields than their TFs obtained from radiotracer experiments.

Table V. Transfer Factors of Br and I in Cereal Samples.

Crop name	N	Br	I	Ref.
		GM/AM (Min. – Max)	GM/AM (Min. – Max)	
White rice	27		2.0E-3 (1.3E-4 – 7.8E-3)	[10]
White rice	1		7.1E-4	[11]
White rice	Br: 63 I: 45	1.5E-1 (1.5E-2 – 1.0E+0)	2.1E-3 (3.5E-4 – 4.9E-3)	This study
Brown rice	13		5.0E-3 (1.5E-4 – 2.0E-2)	[10]
Brown rice	2	6.7E-1 [a] (3.3E-1 – 1.0E+0)	2.9E-2 [a] (1.8E-2 – 3.9E-2)	[12]
Brown rice	4		2.5E-3 (1.1E-3 – 7.3E-3)	[13]
Brown rice	Br: 63 I: 59	2.0E-1 (1.4E-2 – 1.3E+0)	3.9E-3 (4.5E-4 – 1.8E-2)	This study
Brown rice	8		3.8E-3 (1.1E-3 – 7.6E-3)	[1]
Wheat	10		1.5E-4 ± 0.6E-4 [a, b]	[8]
Wheat	4		2.9E-4 [b] (1E-4 – 1E-3)	[9]
Wheat	7	9.4E-2 (3.1E-2 – 2.3E-1)	3.8E-3 (5.1E-4 – 3.0E-2)	This study
Barely	2	3.3E-1 (1.9E-1 – 5.7E-1)	2.8E-3 [a] (1.4E-3 – 5.4E-3)	This study
Cereals	13		6.3E-4 [b] (1.0E-4 – 1.1E-2)	[1]

[a] Arithmetic mean. [b] Radiotracer experiments.

The TFs of Br were 1-3 orders of magnitude higher than the TFs of I for all crops. The correlation between GM/AM of TFs of Br and I in crops is shown in Fig.2. They were well correlated so that they might have some similarity in their uptake by plant roots; however the slope was less than 1 so that I would not be taken up by plants as well as Br would be.

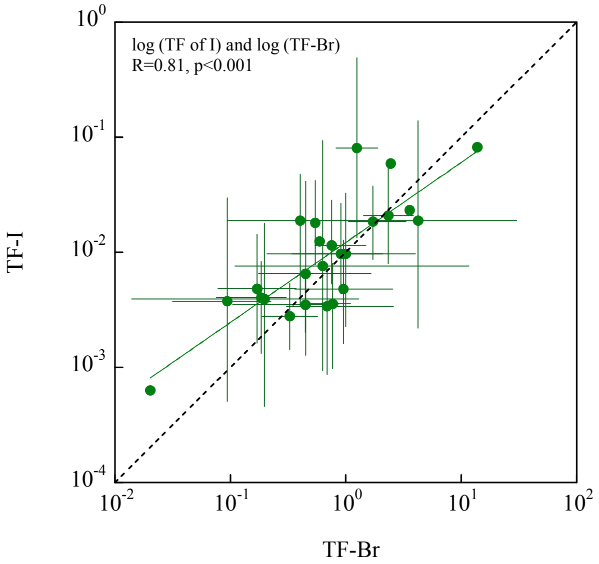

Fig.2. Correlation between TFs of Br and I in crop samples collected in Japan. Respective ends of the error bars show minimum and maximum TF values of crops.

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REFERENCES

1. International Atomic Energy Agency, “Quantification of Radionuclide Transfer in Terrestrial and Freshwater Environments for Radiological Assessments”, Technical Document Series No. 1616, IAEA, Vienna (2009).
2. K. TAGAMI, S. UCHIDA, I. HIRAI, H. TSUKADA, and H. TAKEDA, “Concentrations of chlorine, bromine and iodine in Japanese rivers”, Anal. Chim. Acta 570, 88-92 (2006).
3. K. TAGAMI, S. UCHIDA, A. TAKEDA, S. YAMASAKI, and N. TSUCHIYA, “Estimation of plant unavailable iodine concentrations in agricultural fields”, manuscript submitted for publication.
4. K. TAGAMI, and S. UCHIDA, “Concentrations of chlorine, bromine and iodine in Japanese rivers”, Chemosphere 65, 2358-2365 (2006).
5. K. YUITA, “Application of radioisotope on the dynamics of halogen elements in the soil-plant system –radioisotope tracer-“, Radioisotopes 26, 736-742 (1977).

6. S. YOSHIDA, and Y, MURAMATSU, “Determination of organic, inorganic and particulate iodine in the coastal atmosphere of Japan”,J. Radioanal. Nucl. Chem. 196, 295-302 (1995).
7. H. TSUKADA, and Y. NAKAMURA, “Transfer factors of 31 elements in several agricultural plants collected from 150 farm fields in Aomori, Japan”, J. Radioanal. Nucl. Chem. 236, 123-131 (1998).
8. T. BAN-NAI, and Y. MURAMATSU, “Transfer factors of radioiodine from volcanic-ash soil (Andosol) to crops”, J. Radiat. Res. 44, 23-30 (2003).
9. V. KASHPAROV, C. COLLE, S. ZVARICH, V. YOSCHENKO, S. LEVCHUK, and S. LUNDIN, “Soil-to-plant halogen transfer studies 1. Root uptake of radioiodine by plants”, J. Environ. Radioactiv. 79, 187-204 (2005).
10. H. TAKAGI, T. KIMURA, H. KOBAYASHI, K. IWASHIMA, and N. YAMAGATA, *Hokenbutsuri* (Japanese Journal of Health Physics) 20, 251-257 (1985) Abstract in English.
11. H. TSUKADA, A. TAKEDA, K. TAGAMI, and S. UCHIDA, “Uptake and distribution of iodine in rice plants”, J. Environ. Qual. 37, 2243-2247 (2008).
12. K. YUITA, Y. NOBUSAWA, M. SHIBUYA, and S. ASO, “Iodine, bromine and chlorine contents in soils and plants of Japan. 1. Iodine, bromine and chlorine contents in soils and plants of the basin of the Mimote river”. Soil. Sci. Plant Nutr. 28, 315-336 (1982).
13. Y. MURAMATSU, S. UCHIDA, Y. OHMOMO, “Root-uptake of radioiodine by rice plants”, J. Radiat. Res. 34, 214-220 (1993).
14. K. SCHMITZ, and D.C. AUMANN, “Why are the soil-to-plant transfer factors, as determined by field measurements, for ^{127}I lower than for ^{129}I?”, J. Environ. Radioactiv. 24, 91-100 (1994).