

# Transfer Factors of Radioiodine from Volcanic-ash Soil (Andosol) to Crops

TADAALKI BAN-NAI<sup>1\*</sup> and YASUYUKI MURAMATSU<sup>1</sup>

## Transfer factor/Radioiodine/Vegetable/Wheat/Andosol.

In order to obtain soil-to-plant transfer factors (TFs) of radioiodine from volcanic-ash soil to agricultural crops, we carried out radiotracer experiments. The mean values of TFs (on a wet weight basis) of radioiodine from Andosol to edible parts of crops were as follows: water dropwort, 0.24; lettuce, 0.00098; onion, 0.0011; radish, 0.0044; turnip, 0.0013 and eggplant, 0.00010. The mean value of the TFs of radioiodine for edible parts of wheat (on a dry weight basis) was 0.00015. We also studied the distributions of iodine in crops. There was a tendency for the TFs of leaves to be higher than those of tubers, fruits and grains. A very high TF was found for water dropwort, because this plant was cultivated under a waterlogged condition, in which iodine desorbed from soil into soil solution with a drop in the Eh value. The data obtained in this study should be helpful to assess the long-lived <sup>129</sup>I (half life:  $1.57 \times 10^7$  yr) pathway related to the fuel cycle.

## INTRODUCTION

Iodine is known to be concentrated in the thyroid gland and to be a constituent of thyroxin, a hormone produced by the thyroid gland that stimulates the consumption of oxygen, and thus the metabolism of all cells and tissue in the body<sup>1)</sup>. Therefore, radioiodine is one of the notable radionuclides<sup>2)</sup> from the viewpoint of radiation protection, and there is a strong need to study its migration in the environment.

Short-lived radioiodine isotopes, such as <sup>131</sup>I (half-life: 8.02 days) and <sup>133</sup>I (half-life: 20.8 h), were released into the atmosphere from nuclear explosion tests as well as nuclear accidents (Three Mile Island<sup>3)</sup>, Chernobyl<sup>4)</sup>, JCO<sup>5,6)</sup>. There are already many data on radioiodine transfer parameters, such as the deposition velocities from air to vegetation (e.g. Nakamura and Ohmomo<sup>7,8)</sup>, Hoffman<sup>9)</sup>, Heinemann<sup>10)</sup>, Muramatsu *et al.*<sup>11)</sup> Uchida *et al.*<sup>12)</sup>), although the crop types are limited. For these short-lived radioiodine isotopes it is not necessary to consider the pathway from soil to agricultural crops (root-uptake), because these nuclides, if they enter the soil, decay out during cultivation. However, in the case of <sup>129</sup>I (half-life:  $1.57 \times 10^7$  yr) it is important to take account of its root-uptake pathway in the environmental assessment<sup>13,14)</sup> due to the long half-life.

Iodine-129 is known to be released into the environment from nuclear facilities, such as spent fuel reprocessing plants. In Japan, a new reprocessing plant is now under construction in

Rokkasho-mura (Aomori Prefecture). Therefore, it is important to assess the environmental transfer of this nuclide. Additionally, it is planned in Japan to start the disposition of high-level radioactive wastes into deep stable geological formations by the mid 2030's<sup>15)</sup>. Due to the long half-life of <sup>129</sup>I and to the high affinity of iodine to soil, the level of this nuclide in soil is expected to increase. Therefore, there is a need to study the soil-plant (agricultural products) pathway for people living in the area. However, the transfer factor (TF) values for I are lacking. In the IAEA Technical Report 364<sup>16)</sup> there are only two values for I (grass,  $3.4 \times 10^{-3}$ ; not specified,  $2.0 \times 10^{-2}$ ). The number of TF values for I is significantly fewer than that for other elements, such as Cs, Sr and so on.

We have already performed radiotracer experiments on the root uptake of radioiodine by rice plants<sup>17)</sup> and some vegetables<sup>18,19)</sup>. However, data are still limited for certain crops. It is therefore necessary to obtain more data on the TF of radioiodine from soil to various kinds of agricultural crops. In addition to the TF values for the edible parts of crops, we also studied the distribution of radioiodine in other organs. This is due to the following two reasons: (i) It is worthwhile to understand the distribution of iodine in crops from the viewpoint of plant physiology. (ii) Sometimes non-edible parts (e.g. leaves of radish) are also eaten.

In this study, we carried out radiotracer experiments using volcanic-ash soil (Andosol or Kuroboku soil), the most common of Japanese arable soils, to determine the TFs of radioiodine from soil to crops usually consumed in Japan.

## MATERIAL AND METHODS

Andosol samples collected in Tokai-mura (Andosol-1) and Tsukuba (Andosol-2), Ibaraki Prefecture, Japan, were used in

\*Corresponding author: Phone: +81-43-206-3255,  
Fax: +81-43-206-3267,  
E-mail: t\_bannai@nirs.go.jp

<sup>1</sup>Environmental and Toxicological Sciences Research Group, National Institute of Radiological Sciences, 4-9-1 Anagawa, Inage-ku, Chiba 263-8555, Japan

**Table 1.** The properties of soils.

	Andosol-1	Andosol-2
Sampling area	Ibaraki Prefecture, Tokai mura	Ibaraki Prefecture, Tsukuba
pH	5.3	5.5
Cultivation vegetables	water dropwort	lettuce, onion, radish, turnip, eggplant and wheat
CEC (cmol kg <sup>-1</sup> )	17	37
AEC (cmol kg <sup>-1</sup> )	6.7	0.4
Organic carbon (dry soil, %)	4.3	9.0
Iodine concentration (ppm)	38.5	32.6

**Table 2.** Cultivation design for the experiments.

Common name	Botanical name	Precultivation period	Cultivation period
water dropwort (Seri)	<i>Oenanthe javanica</i> (Blume) DC	none (cattage)	40–60 d
lettuce	<i>Lactuca sativa</i> L.	none (seed)	90 d
onion	<i>Allium cepa</i>	none (seed)	220 d
radish	<i>Raphanus sativus</i>	none (seed)	25 d
turnip	<i>Brassica campestris</i>	none (seed)	100–190 d
eggplant	<i>Solanum melongena</i>	—	70–120 d
wheat	<i>Triticum aestivum</i>	20 d	210 d

—: not measured.

the present experiments. The properties of the soil are shown in Table 1. Iodine-125 (half life: 59.4 d) was used as a radiotracer (KI in 10<sup>-5</sup> M NaOH solution) in the experiments. Iodine-125 (2–4 MBq pot<sup>-1</sup>) and a mixed chemical fertilizer (5 g pot<sup>-1</sup>, N : P : K = 14 : 10 : 13) were thoroughly mixed with 3.0 kg of the soil and used to fill Wagner pots (surface area, 200 cm<sup>2</sup>; volume, 3 liters). Some details of plant samples concerning the cultivation conditions are given in Table 2. One week after mixing on <sup>125</sup>I tracer into soil, young seedlings (or seeds) were planted in pots.

The pots were placed in plant growth chambers (Puffer-Hubbard CEC38-15HLE or Koito Koitoron) and cultivated until harvest. The chamber was operated at 24–30°C during the daytime (12 h) and at 20–25°C during the night (12 h). The light intensity at the plant level in the chamber was about 50,000–70,000 lx. The cultivation periods of each crop are also given in Table 2. After the plants were harvested, they were divided into organ parts (e.g. leaf, stem, root). To check for differences in the <sup>125</sup>I concentration of various parts, the crops were cut into small pieces. For example, in order to examine the differences in the transfer factors (TFs) by leaf age, lettuce leaves of various positions (from inner to outer) were also taken.

The samples were placed in polyethylene vials and the concentrations of <sup>125</sup>I were determined by means of a NaI scintillation counter (Aloka ARS-380). If the errors were more than 30%, these peaks were considered to be under the limits of detection, and were not used to calculate the mean value. The radioiodine concentrations in the soil samples used in the experiments were also measured to examine the homogeneity. Decay corrections were made at the beginning of the experiment. The soil-to-plant transfer factor (TF) of radioiodine is defined as the “concentration of the nuclide per unit fresh or dry weight of the plant

or plant organ at the time of harvest (Bq g<sup>-1</sup>)” divided by the “concentration of the nuclide per unit dry soil at the time of planting (Bq g<sup>-1</sup>)”.

After measuring the radioactivity, the samples were freeze-dried to calculate their dry/wet ratios, and then the TFs were estimated on a dry-weight basis.

The weighted mean values of TFs of radioiodine for edible parts of each leaf vegetable were calculated from the individual values as well as the weight of each part. For example, in calculating the TF for leaves, the weights of the leaf parts together with their TF values were considered. The number of samples used in the calculations for each crop was 4–18.

## RESULTS

In selecting the crops we considered those which had not been determined in our previous studies<sup>17–19</sup> (except for radish) using Andosol. The TFs of radioiodine obtained for various organs of the crops (water dropwort, lettuce, onion, radish, turnip, eggplant and wheat) in this study are given in Table 3.

As shown in this table, the TFs of I for the edible parts of the crops were as follows (on a wet weight basis): water dropwort (range, 0.14–0.39; mean, 0.24), lettuce (range, 0.00070–0.0011; mean, 0.00098), onion (range, 0.00094–0.0014; mean, 0.0011), radish (range, 0.0034–0.0059; mean, 0.0044), turnip (range, 0.0007–0.0023; mean, 0.0013), and eggplant (range, 0.00007–0.00014; mean, 0.00010). The TFs (on a dry-weight basis) of radioiodine for wheat grain (with bran) were in the range of <0.00004–0.00031 (mean, 0.00015).

The TF values for water dropwort were more than 50-times higher than those of other crops. A markedly low TF value (on a wet weight basis) was found in eggplant. On a dry-weight basis,

**Table 3.** Transfer factors (TFs) of the radioiodine for various organs of crops.

Organ	Sample number	TF (Wet)		TF (Dry)
a) Water dropwort				
leaf (edible)	6	0.24	$\pm 0.03$	—
stem	6	0.13	$\pm 0.01$	—
b) Lettuce				
leaf (edible)	6	0.00098	$\pm 0.00017^*$	0.0067 $\pm 0.0032$
stem	6	0.00050	$\pm 0.00019$	0.0030 $\pm 0.0015$
c) Onion				
bulb (edible)	4	0.0011	$\pm 0.0002$	0.011 $\pm 0.004$
leaf	4	0.0053	$\pm 0.0046$	0.045 $\pm 0.029$
d) Radish				
root (edible)	6	0.0044	$\pm 0.0011$	0.051 $\pm 0.013$
peel	6	0.0066	$\pm 0.0014$	0.061 $\pm 0.016$
leaf	6	0.013	$\pm 0.003$	0.12 $\pm 0.02$
e) Turnip				
root (edible)	6	0.0013	$\pm 0.0006$	0.013 $\pm 0.006$
f) Eggplant				
fruit (edible)	18	0.000098	$\pm 0.000024$	0.00095 $\pm 0.00029$
calyx	5	0.00099	$\pm 0.00061$	0.0070 $\pm 0.0035$
peel	12	0.000080	$\pm 0.000021$	0.00065 $\pm 0.00020$
leaf	18	0.0075	$\pm 0.0028$	—
branch	6	0.0024	$\pm 0.0013$	—
g) Wheat				
grain (edible)	10	—		0.00015 $\pm 0.00006$
chaff	10	—		0.0041 $\pm 0.0026$
rich	10	—		0.0046 $\pm 0.0023$
leaf	10	—		0.020 $\pm 0.006$
stalk	10	—		0.058 $\pm 0.015$

\*The wet weight of old withered leaves were calculated by using wet/dry ratio of no-withered leaves and TF (wet weight basis) was estimated.

the transfer factors of I for wheat were the lowest among the TFs estimated in this study.

We also determined the distribution of radioiodine in various organs of the crops; the results are given in Table 4. Details of the distribution patterns are discussed below.

## DISCUSSION

### Transfer factors for water dropwort

The high transfer factors of I for the water dropwort could be caused by cultivation of this crop under a flooded condition. We found in our previous study that the TFs (on a dry-weight basis) for the leaves of rice plants cultivated under a waterlogged condition were markedly high (1st leaf, 3.3; 2nd leaf, 6.0)<sup>20</sup>. Furthermore, radioiodine adsorbed on soil desorbs into a soil solution with time when the soil has been flooded with water<sup>21</sup>. This phenomenon could be explained as follows. After the water was logged on the soil, the number of aerobic microorganisms increased in the soil, and they consumed oxygen and organic matter. As a result, under a flooded condition, the soil became anaerobic. Under an anaerobic condition, iodine adsorbed on soil is known to be desorbed from the soil into solution as iodide

(I<sup>-</sup>)<sup>18</sup>. Since I is plant available<sup>13</sup>, the plants readily adsorbed it. This may have caused the higher TF values for water dropwort than other crops cultivated in non-flooded soil.

### Transfer factors for lettuce

The transfer factors of I for lettuce obtained in this study were lower than those for three vegetables in our previous studies<sup>18,19</sup>, i.e. spinach (0.0031), komatsuna (0.016) and cabbage (0.0016). According to Robens and Aumann<sup>22</sup>, the <sup>129</sup>I concentration of "lettuce and endive" is lower than that of white cabbage and spinach. Cline and Klepper<sup>23</sup> reported that the concentration factors of lettuce were in general lower than those of spinach. These results agreed with our findings. The low TFs for lettuce might partly have been caused by the cultivation condition, in which the soil was kept relatively dry. Under this condition, <sup>125</sup>I was strongly fixed on soil particles and hardly any iodine would be taken up by the lettuce. This contrasted sharply with the case of water dropwort, which was cultivated under the flooded condition, as mentioned above.

Figure 1 shows the relationship between the TFs of I and the leaf positions (i.e. leaf age) for lettuce. The concentrations of I in the outer leaves (old leaves) of the lettuce were higher than those

**Table 4.** Distribution of radioiodine in various organs of the crops.

## a) Radish (leaf part)

Leaf position from outside	Sample number	concentration ratio* (Wet)
1	6	1.00
2	6	1.08
3	6	1.00
4	6	1.11
5	6	0.96
6	6	0.79
7	6	0.61

\*Values were normalized by the concentration of the leaf at first position from outside.

## b) Wheat (leaf part)

Leaf position from top	Sample number	concentration ratio* (Dry)
1	10	0.74
2	10	0.93
3	10	1.10
4	7	1.46

\*Values were normalized by the mean value of concentration in leaf.

## c) Wheat (stalk)

Stalk position from top	Sample number	concentration ratio* (Dry)
1	10	0.41
2	10	0.65
3	10	1.17
4	10	1.78

\*Values were normalized by the mean value of concentration in stalk.

in the inner ones (young leaves), except for the innermost leaf. Cline and Klepper<sup>23)</sup> showed that the concentration factors of lettuce were in the order oldest leaves > middle leaves > youngest leaves. This result supported our findings. In our previous study<sup>18)</sup>, we found that the transfer value was higher in larger (older) leaves (e.g. spinach and komatsuna) than smaller (younger) ones. These trends suggested that I was taken up through the roots and transported into the leaves, where it accumulated, whereas water was transpired from the leaves.

As mentioned above, we also found a higher value of I in the innermost leaf (see Fig. 2). It is not easy to explain the reason for I. In the case of Cl, Kumazawa<sup>24)</sup> studied the Cl<sup>-</sup> distributions in rice plants using the <sup>36</sup>Cl tracer, and described that <sup>36</sup>Cl moved first to the innermost leaf, and subsequently to lower (older) leaves. It is possible that iodine behaves similar to Cl in the transportation in plants.

*Transfer factors for onion*

The transfer factors of I for the edible part (bulb, containing stem) of onion were higher than those for lettuce. Robens and Aumann<sup>21)</sup> also reported that the <sup>129</sup>I concentration in “onion” was higher than that of “lettuce and endive”. The TF of 0.0053 for leaf of onion (green parts above edible part) was higher than those for the bulb. This value was as high as the spinach value.

The relatively low TF value observed in the edible part of

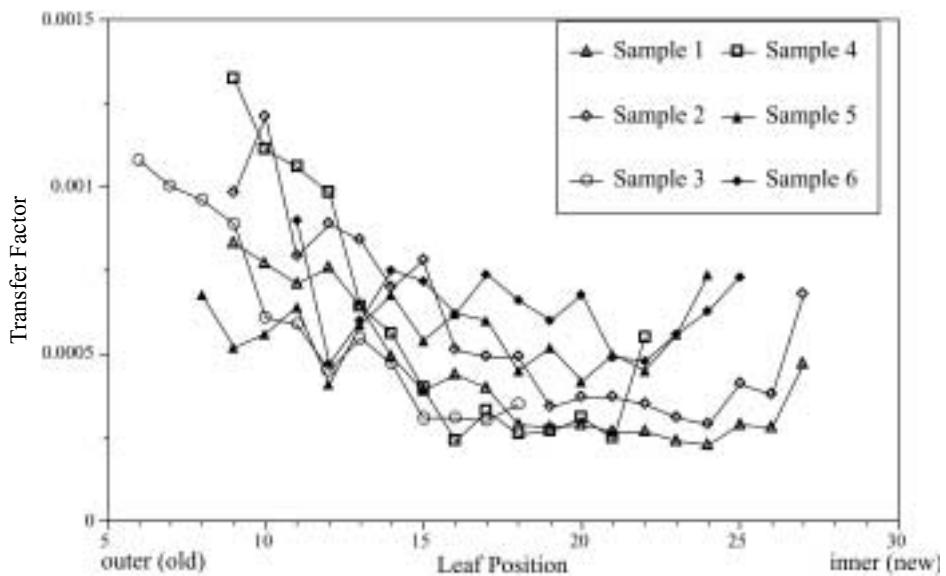
onion should be attributable to the role of the bulb as a storage organ. This means that proportions of organic substances, such as protein and carbohydrate, are high and those of inorganic compounds, such as ash, are very low. According to the STA (Science and Technology Agency) Standard Tables of Food Composition in Japan<sup>25)</sup>, the rate of ash for onion (0.4%) was much lower than the rates for spinach (1.7%), komatsuna (1.6%), water dropwort (1.1%) and lettuce (0.9%). Therefore, TF for onion (bulb as fresh weight) appeared to be low due to this dilution effect.

We divided an onion into various parts and measured the <sup>125</sup>I concentrations (Fig. 2). The <sup>125</sup>I concentrations in various parts of onion were in the order stem >> outer parts > inner parts. The higher TF values in the outer parts (older parts) might be explained in the same way as mentioned for lettuce.

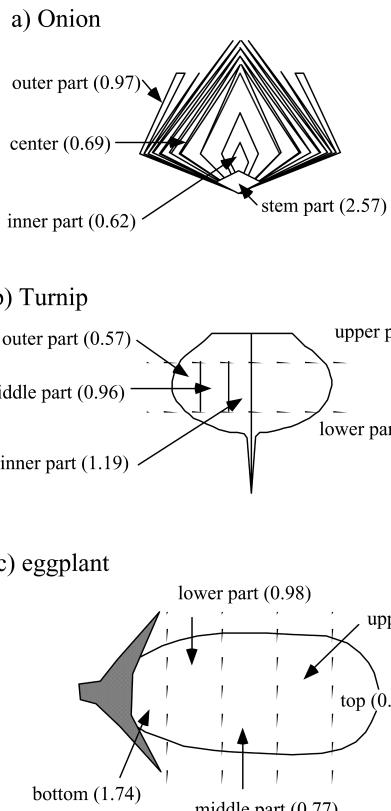
*Transfer factors for radish*

The transfer factors to the radish from Andosol-2 (Tsukuba) given in Result were estimated in this experiment. In previous work<sup>18)</sup>, we obtained a TF value of 0.0015 for the edible parts of radish which was cultivated in Andosol of Tokai-mura (Andosol-1 in Table 1). The values obtained from the two experiments using different Andosols were in a similar order of magnitude, if we considered the ranges and errors of the values.

Tsukada and Nakamura<sup>26)</sup> analyzed the concentrations of 31



**Fig. 1.** Transfer factors of radioiodine for different leaf positions of lettuce.



**Fig. 2.** Distribution of radioiodine in various organs of the crops.  
(Note) Values in parenthesis are normalized by the mean value of the whole part.

elements (including iodine) in several agricultural plants cultivated at 150 farm fields in Aomori Prefecture, and estimated the TF values based on the stable elements' data. The geometric mean of the TF value (fresh-weight basis) of I for Japanese rad-

ish determined by them was  $5.5 \times 10^{-4}$ . This value was about 8-times lower than our TF value for radish ( $4.4 \times 10^{-3}$ ) by radiotracer experiments. The lower TF value obtained from the stable element analysis should be related to the insoluble iodine in the soil solid phase as well as an aging effect.

The TF for leaf (0.013) was markedly higher than that for tuber (edible parts) of radish. Sheppard *et al.*<sup>27)</sup> reported that the concentration ratio of I in leafy materials was highest. These results agreed with our findings. The TF for leaf of radish was as high as that for komatsuna. This was attributable to both komatsuna and radish belonging to the same family. In comparison to the concentrations of I in the outer (old) and inner (young) leaves of radish, the former concentrations were higher than the latter ones (Table 4). This agreed with the findings for lettuce.

#### Transfer factors for turnip

In order to obtain the distribution of radioiodine for tuber of turnip, we divided a tuber into various parts (upper, lower, outer, inner parts) and estimated their concentrations. The boundary between the stele and cortex was clear. The outer part consists of only cortex, while the upper part is mostly of stele. The other parts consist of cortex and stele. As shown in Fig. 2, the concentrations for various parts of turnip were in the order upper part >> middle, inner and lower parts > outer part. This observation suggested that stele contained more iodine than cortex. The nutrition and water absorbed by the epidermal cells of the root passed through the cortex, and gathered and concentrated in the stele.

#### Transfer factors for eggplant

The transfer factors of I for eggplant were similar to that of another fruit vegetable, i.e. edible parts of tomato (0.00026). The TFs for leaves of eggplants were higher than that for lettuces, though the TFs for edible parts of eggplant were much lower than that for lettuces. This result showed that the ability on

uptake iodine of eggplant was not as low as those of other vegetable, such as lettuce.

The TF for leaf of eggplant was 0.0075, which was about 80-times higher than that for the edible parts. This result was similar to our previous observation<sup>18)</sup> for tomato plants, i.e. the TF for the leaf of tomato was about 100-times higher than that for the edible part. We divided an eggplant fruit into various parts (top, upper, lower, bottom parts) and measured their distributions (Fig. 2). We found that the TF of the bottom part (near the calyx) was the highest.

The concentrations in leaves near the origin (lower part) were higher than those near the tip (higher part). This might be related to the age of the leaves, i.e. higher concentrations in older leaves. However, the concentration in the branch near the origin was lower than that near the tip. This trend should be related to the transport of <sup>125</sup>I from the roots by water through the vascular system, followed by accumulation in leaves with time. The TFs for various parts of eggplant were in the order leaf > branch > calyx > fruit and peel. The TF for calyx was about ten-times higher than those for fruit and peel.

#### Transfer factors for wheat

The TF of wheat grain was lower than that of other parts in wheat. We reported that the TF of brown rice (hulled rice) with bran was 0.0063<sup>20)</sup>. This value was much higher than that of wheat grain. Rice was cultivated in a water-flooded soil condition, and the wheat was cultivated in an upland soil condition. As mentioned for water dropwort, radioiodine adsorbed on soil would desorb under a flooded condition, resulting in a higher availability of iodine for the rice<sup>20)</sup> and the other plants.

Shinonaga *et al.*<sup>28)</sup> determined the concentration of iodine in cereal grain cultivated at 38 location (24 locations for wheat, 14 for rye) in Austria, and reported that the TF values for cereal grains (without rye) were in the range 0.00047–0.017 (median, 0.0016). Their mean value was 10-times higher than our value. Among others, it was possible that the low TF value were caused by the high adsorption capacity or high  $K_d$  value (soil/water distribution coefficient) of iodine by Andosol, which we observed previously<sup>19,29)</sup>. Low stable iodine concentrations in Austrian soil<sup>28)</sup> might also result in high TF values.

The TFs for various parts of wheat were in the order stalk > leaf > rich and chaff > grain (Table 3). According to Robens and Aumann<sup>22)</sup>, the <sup>129</sup>I distribution in wheat was in the following order: stalk > chaff > grain. This result agreed with our data. The concentrations of leaf and stalk of higher positions (new leaves) were lower than those of lower positions (older leaves), and the same as those observed in lettuce and some other vegetables.

#### Comparison with literature values

The TF values obtained in this study are summarized in Fig. 3 together with literature values (i.e. Muramatsu: our previous studies using Japanese Andosol<sup>17–19)</sup>, Ng: soils in North America<sup>30)</sup>, Luo: sandy soil<sup>31)</sup> by radiotracer experiments for different crops. In this figure, the TFs are categorized in groups

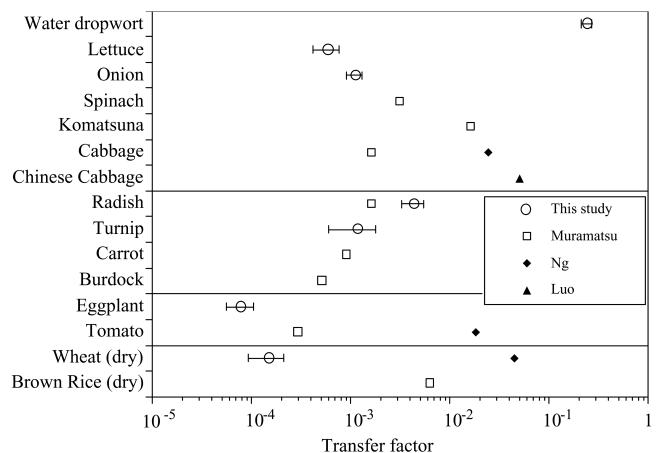


Fig. 3. Comparison of the TFs (on a wet weight basis) for edible parts obtained in this study with those in references (Muramatsu<sup>17–19)</sup>; Ng<sup>30)</sup>; Luo<sup>31)</sup>.

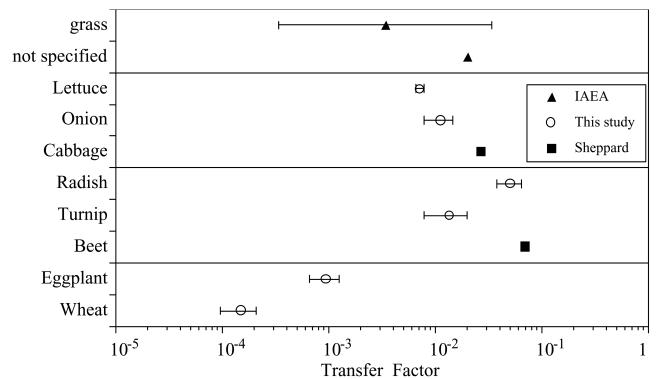


Fig. 4. Comparison of the TFs (on a dry weight basis) obtained in this study with those in references (IAEA<sup>16)</sup>; Sheppard<sup>27)</sup>.

based on the edible parts.

For cabbage, tomato and wheat, there are both Japanese and North American values. The Japanese values were considerably lower than those obtained in North America. The character of Andosol might cause this. Because the iodine sorption on Andosol is very high (i.e. high  $K_d$  value), added radioiodine is fixed on soil particles. Therefore, plants grown on Andosol showed lower TF values.

The tendency of TFs (on wet weight basis) for various groups were in the order stem and leaf vegetables  $\geq$  root vegetables > fruit vegetables > grain. No significant difference for I TF between leaf vegetables and root vegetables was observed in this experiment, though the TFs of several elements (e.g. Sr, Zn, Mn) in leaf vegetables were markedly higher than those in root vegetables in our previous paper<sup>32)</sup>. This was partly due to the low TFs of lettuce and onion as leaf vegetables.

Two types of reference values for TFs were reported in IAEA Reports in 1982<sup>33)</sup> and in 1994<sup>16)</sup>. The former were default values of the concentration factors, which were identical with the TFs (on a wet weight basis) for common (unidentified) food crops. The value for iodine was set at 0.02. As discussed above, this

value is too high for fruit vegetables (such as eggplant and tomato) and grain.

Our results obtained in the present experiments on a dry-weight basis were compared with the respective values listed in TR-364 for grass and other crops (not specified) and Sheppard *et al.*<sup>27)</sup> for cabbage and beet, and are shown in Fig. 3.

The TF (on a dry weight basis) of I for “not specified” (TR-364) was also much higher than those of eggplant and wheat. This value would cause an overestimation of the I intake from fruit vegetables and grains. On the other hand, this value was much lower than that of water dropwort using the common dry/wet ratio. Therefore, it is necessary to pay attention to plants growing in wet lands because of their possible high TF value for I.

Since the TF values for iodine vary very widely, it is necessary to consider the values according to the types of crops and edible parts as well as the soil type and/or crop cultivation practices.

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