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Articles

Uptake of ^{131}I, ^{134}Cs and ^{137}Cs in tulip (*Tulipa gesneriana* L.) after the Fukushima Daiichi nuclear accident and their translocation from its above ground parts to the bulb

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Radioiodine (^{131}I) and radiocesium (^{134}Cs and ^{137}Cs) uptake through the above ground parts and transfer to the bulb of tulip (*Tulipa gesneriana* L.) was measured in April 2011, one month after the heavy depositions of radioactive materials. Plant samples were collected from a flower garden of J Village on April 20 and 28, 2011. Each sampled above ground parts was equally divided into two portions and one portion was washed with detergent including surfactant to remove dust from the surface and the other portion was unwashed as a control. The average retention factor after washing (activity concentration ratios after and before washing) for radioiodine and radiocesium were 0.97 and 0.58, which means more radioiodine was fixed on the plant surface. Activity of one sampled tulip bulb was measured and the concentration ratios between the bulb to the directly contaminated above ground parts were 0.03 for ^{131}I and 0.47 for radiocesium. It was clear that radiocesium was more mobile than iodine in the plant.

1. Introduction

Translocation of radiocesium in plants is well understood because Cs behaves analogously to K, an essential element for plants.[1, 2] In the March 2011 accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP), large amounts of radionuclides were released to the environment, with the major nuclides being radioiodine (^{131}I, $T_{1/2}$=8.02 d) and radiocesium (^{134}Cs, $T_{1/2}$=2.06 y; ^{137}Cs, $T_{1/2}$=30.2 y). These radionuclides were found for leafy vegetable samples which had directly received radioactive fallout from the FDNPP accident; the concentrations exceeded the guidance levels at that time, that is, 2000 Bq kg^{-1} for ^{131}I and 500 Bq kg^{-1} for total radiocesium ($^{134+137}Cs$).[3]

In June 2011, about 3 months after the heavy depositions, ^{131}I contamination was no longer being reported in agricultural products mainly due to the short physical decay of ^{131}I. However, radiocesium concentrations remained high in some agricultural foods, such as fruits, green tea leaves and shiitake mushrooms,[4,5] which drew public concern as people wanted to avoid an internal dose by ingestion of contaminated foods. Because radiocesium can translocate to non-directly contaminated edible parts of plants from directly contaminated parts or roots, how much radiocesium can be translocated from contaminated parts and taken up by roots is of interest. The root uptake ability has been studied for rice and other crops and results were reported.[6-9] From these results, it was deduced that the uptake of radiocesium through roots was usually very low for the soils found in Japan, so that the root uptake pathway effect was limited to enhancement of the radiocesium concentration in edible parts of crops. Radiocesium translocation from above ground parts has been studied for orchard trees and some forest trees[10-13]; it is now considered that uptake processes through the bark and leaves are the major transfer pathways of radiocesium in trees.

Translocation is an important factor affecting radiocesium concentration and effective half-life in plants[14]; however, the translocation of radiocesium from the above ground parts to the root or bulb has not been reported yet after the FDNPP accident. Moreover, it would be interesting to identify the translocation difference between radioiodine and radiocesium since these are the major radionuclides in accidental releases from nuclear power plants. In April 2011, we collected tulip (*Tulipa gesneriana* L.) samples at J Village, about 20 km south of the FDNPP; they consisted of both above ground and below ground (bulb) parts. Here, we report the translocation factors (F_{tr}) from the above ground parts to the bulb for the radioactive FDNPP accident fallout. Although we could obtain only a few samples for analysis, particularly because of the relatively short half-life of ^{131}I, the data will be useful to understand radionuclide fates in the environment.

2. Materials and Methods

Two tulip samples were collected from a flower garden at J Village on April 20 and 28, 2011 (one per sampling); the flowering time had almost finished on the former date. On April 20, we also collected a bulb with roots but without above ground parts. The bulb was not dead but no shoot had emerged. These tulip bulbs had been planted in soil just below the surface, at 0-5 cm depths. The root part was distributed deeper than 5 cm and the respective ^{131}I and ^{137}Cs concentrations in the soil 5-7 cm were 1850 and 630 Bq kg^{-1} dry soil on April 20, and 590 and 330 Bq kg^{-1} dry soil on April 28. The vertical distribution of the radionuclides in this garden soil has been reported elsewhere.[15]

The samples were transferred into our laboratory in Chiba within 2 days, and after the following treatments, their activity concentrations were measured with a Ge detecting system (Seiko EG&G) for 2000 – 3600 s. A mixed gamma standard solution (Amersham, QCY-46) was used for an efficiency correction and three reference standard materials IAEA-156, 373 and 375 were used for an accuracy check.

The weight of the above ground parts was 27.1 g-wet for April 20 sample, and 41.3 g-wet for April 28 sample. Each above ground parts was divided into two as shown in Fig. 1. One portion was not treated (control) and the other portion was washed carefully by hand with liquid detergent including a

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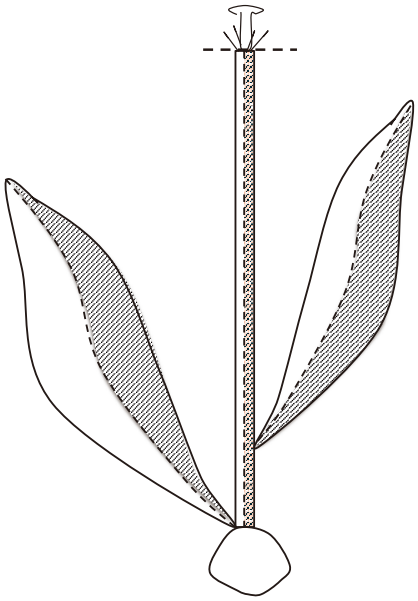

Figure 1. Schematic showing division of the tulip above ground parts into two portions.

surfactant to remove dust from the surface. The detergent on the washed sample portion was then removed using reverse osmosis (RO) water to avoid adding possible radioactive contamination from tap water. Both control portions were cut into pieces and put into separate 100-mL plastic vessels for measurement of their radioactivity.

The two bulbs collected on April 20 were not cut for measurement. Each bulb sample was simply treated as follows: soil was removed with a paper towel and then the activity concentration was measured for 3600 s with the Ge detecting system (control). After the measurement, this control sample was washed with detergent as described above to remove contaminated soil, and then rinsed with RO water. Water was removed with a paper towel and then the sample was set in the same place in the Ge detecting system as before washing, and its activity concentration was measured to obtain the washing effect. The bulb without the above ground parts was similarly treated: that is, soil was removed and the activity concentration was measured; then it was washed and the activity concentration was measured again. Unfortunately, we did not weigh these bulbs; but based on the April 28 bulb sample weight, we assumed that these two bulbs had a similar weight of 30 g-wet. The bulb collected on April 28 was washed as mentioned above and then cut into pieces for quantitative radioactivity analysis. The weight of this bulb was 33.3 g-wet.

Radioactivity correction dates for all samples were the date of sampling. Quantitative data were available for above ground part samples for both dates and for the bulb collected on April 28. Data for ^{131}I (365 keV), ^{134}Cs (605 keV and 796 keV), ^{137}Cs (662 keV), and ^{129m}Te (696 keV) were measured when possible.

3. Results and Discussion

3.1. Activity ratios between ^{131}I/^{137}Cs in above ground parts of tulip. Activity concentrations of ^{131}I and ^{137}Cs in above ground parts of tulip samples before washing are shown in Table 1. To compare the concentration data with other plant species, concentration data were further corrected to March 11, 2011. The concentrations of ^{131}I were similar on both sampling dates but the ^{137}Cs concentration was lower in the April 28 sample than that in the April 20 sample. The activity ratios of ^{131}I/^{137}Cs were 200-260. When the activity concentration ratio data were compared with our previously reported values in soil 0-2 cm depth samples[15] (data were corrected to March 11, 2011), it was clear that the ratios in soil were 3-4 times lower than those observed on plants. Ratios of their amounts of gaseous forms to the total released were reported to be 0.5-0.7 for ^{131}I and 0.02-0.07 for radiocesium.[16] Unfortunately, the chemical forms of ^{131}I and radiocesium were not known. Because most depositions of the nuclides were observed into be wet forms, chemical forms of ^{131}I and radiocesium would likely be I^- or IO_3^- and Cs^+. At least, the present results showed that radioiodine and radiocesium were both retained on above ground parts of tulip plants.

Radioiodine retention on plants was observed for several plants collected in March to April 2011 in Chiba City, ca. 220 km south from the FDNPP. We previously reported washing effect for the above ground parts of Japanese dock (*Rumex japonicas*), mugwort (*Artemisia indica* var *maximowiczii*), dandelion (*Taraxacum officinale*), wild onion (*Allium macrostemon*), butterbur (leaf blades) (*Petasites japonicus*)[17]; thus these measured activity concentration data are listed in Table 1. Further to compare, activity concentration data in leaves of two tree species, that is, red robin (mature leaves) (*Photinia glabra*) and sweet viburnum (*Vibrnum odoratissimum*), are also listed in Table 1 together with soil sample data collected from the same sampling site. It was clear that activity ratios of ^{131}I/^{137}Cs in plant samples collected in Chiba ranged from 15-60, and these values were similar to those in soil. According to model simulation results,[18] heavy radioactive deposition occurred during March 21-22, 2011 at J Village and in Chiba areas. The ratios of ^{131}I/^{137}Cs in two soil samples in J Village (ratio=50-61), one soil sample in Chiba (ratio=26), and 36 soil samples collected at the coastal areas of three municipalities in Fukushima Prefecture south of the FDNPP from open data,[19] are also listed in Table 1. Because these data were in the same order of magnitude and considering the analytical results of radioactive material distribution and time of discharges from the FDNPP by Nagai et al.,[20] it was assumed that the ^{131}I and ^{137}C sources were similar in these areas.

3.2. Effect of Washing. To obtain washing effect, the retention factor after washing ($F_{r\_washing}$) was calculated. The factor is defined as the activity concentration ratio between before and after washing, and the results are shown in Fig. 2 for above ground parts. Because only two samples were available, it is difficult to compare the $F_{r\_washing}$ data difference between radionuclides statistically; however, averages of the $F_{r\_washing}$ of the samples collected on April 20 and 28, 2011 was calculated to be 0.97 for ^{131}I, 0.58 for radiocesium (^{134}Cs and ^{137}Cs), and 0.71 for ^{129m}Te. The results showed ^{131}I was less mobile on the plant surface than the other nuclides. Probably because one month had already passed since the heaviest direct deposition in March 2011, the mobile portion on the plant surface had already been removed by weathering.

We previously reported the washing effect for these three elements for some wild plants collected on March 28, 2011 and found similar $F_{r\_washing}$ with the average values being 0.79 for ^{131}I, 0.79 for radiocesium and 0.71 for ^{132}Te.[17] It is highly probable that differences in leaf surface structures, plant shapes of the species and chemical forms of radionuclides could influence the washing effect, and we also cannot ignore the samples were collected at different times. However, it was clear that iodine, which was thought to be in anionic forms when it was deposited,[18] was not mobile on the plant surface. The IAEA[21] noted that the key parameters of plant surface retention of radionuclides are valence state of the radionuclide and particle size. For radiocesium, if the size of deposited material was very small such as Cs micro-particles in several μm size, then it could retain on the plant surface physically, and if ionic form radiocesium was deposited then it could bound weakly with

TABLE 1: Activity Concentrations (Bq g^{-1}) of ^{131}I and ^{137}Cs and the Ratios of ^{131}I/^{137}Cs in Soil Samples and Tulip Above Ground Samples (control) Collected in J Village, One Soil Sample and Seven Plant Species Collected in Chiba, and Soil Data Collected from Coastal Area of Naraha, Hirono and Iwaki in Fukushima Prefecture Reported by MEXT.[19] Activity Concentrations was Decay Corrected to March 11, 2011. Units for Soil were on Dry Mass Basis and Those for Plants were on Wet Mass Basis Unless Otherwise Indicated.

Sample	Collection date	^{131}I, Bq g^{-1} [c]	^{137}Cs, Bq g^{-1} [c]	^{131}I/^{137}Cs
Tulip, above ground	April 20, 2011	225±2	1.11±0.03	202
Soil (J Village), 0-2 cm[a]	April 20, 2011	699±41	11.5±0.6	61
Tulip, above ground	April 28, 201	229±4	0.88±0.04	261
Soil (J Village), 0-2 cm[a]	April 28, 2011	587±27	11.8±0.2	50
Soil (Naraha), 0-5 cm[b] (N=1)	June 7, 2011	1.18×10^7 (Bq m^{-2})	1.31×10^5 (Bq m^{-2})	90
Soil (Hirono), 0-5 cm[b] (Geometric mean, N=9)	June 7, 2011	5.97×10^6 (Bq m^{-2})	8.52×10^4 (Bq m^{-2})	70 (Range: 31-113)
Soil (Iwaki), 0-5 cm[b] (Geometric mean, N=26)	June 5-6, 2011	3.34×10^6 (Bq m^{-2})	4.97×10^4 (Bq m^{-2})	67 (Range: 37-127)
Japanese dock (*Rumex japonicas*)	March 28, 2011	41.8±0.8	2.49±0.13	17
Mugwort (*Artemisia indica* var *maximowiczii*)	March 28, 2011	37.3±0.5	2.32±0.07	16
Dandelion (*Taraxacum officinale*)	March 28, 2011	28.9±0.3	1.88±0.05	15
Butterbur, leaf blade (*Petasites japonicus*)	March 28, 2011	37.7±0.3	1.51±0.04	25
Wild onion (Allium macrostemon)	March 28, 2011	22.0±0.4	0.63±0.04	32
Red robin (*Photinia glabra*)	April 26, 2011	18.6±0.7	1.01±0.02	18
Sweet viburnum (*Vibrnum odoratissimum*)	April 28, 2011	12.0±1.2	0.20±0.02	60
Soil (Chiba), 0-5 cm (N=1)	April 25, 2011 (Major deposition observed on March 20-21, 2011)	2.2×10^5 (Bq m^{-2})	8.3×10^3 (Bq m^{-2})	26

[a]Data from Tagami et al., 2011.[15] [b]Data from MEXT.[19] [c]±: Counting error

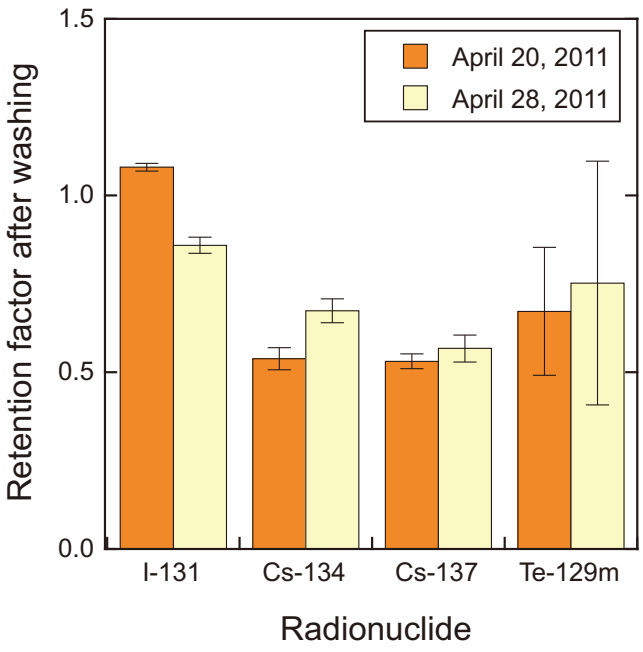

Figure 2. Retention factor after washing (activity concentration ratios between after and before washing) of above ground parts of tulip samples collected on April 20 and 28, 2011 for ^{131}I, ^{134}Cs, ^{137}Cs and ^{129m}Te. Bar shows one sigma error from counting.

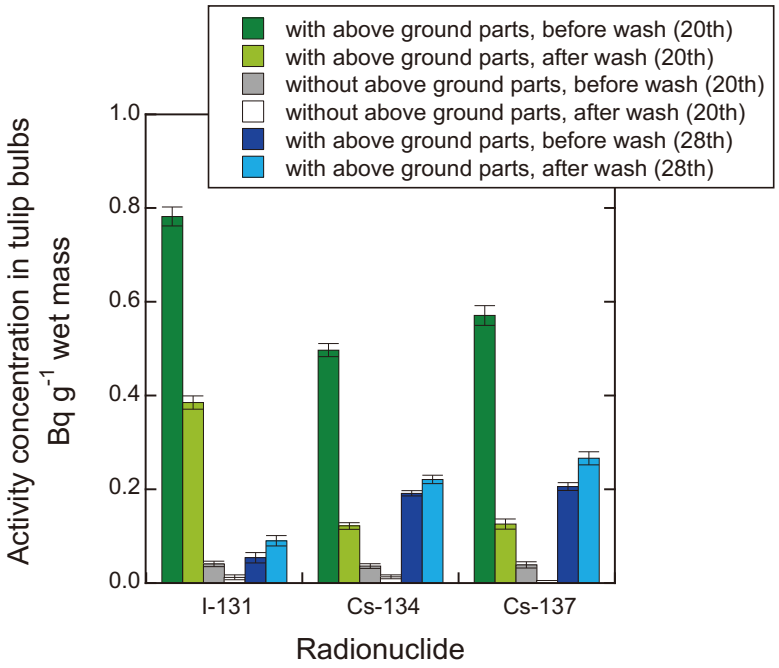

Figure 3. Activity concentration of ^{131}I, ^{134}Cs and ^{137}Cs in before and after washing tulip bulb samples with the above ground parts collected on April 20 and 28, 2011 and without the above ground parts collected on April 20, 2011. Bar shows one sigma error from counting.

organic matter on the plant surface. It was highly probable that radiocesium in these forms are not readily mobile by weathering, but some could be removed by washing. However, further study is needed to identify the mechanism.

For the two tulip bulbs collected on April 20 and one bulb collected on April 28, activity concentrations of ^{131}I and radiocesium before and after washing are shown in Fig. 3. ^{129m}Te was not detected in any of these bulb samples. As explained in the Materials and method section, we assumed that the bulbs with and without the above ground parts had the same weight; yet we compared the concentration between bulbs with and without the above ground parts. When the bulb with the above ground parts was washed, ^{131}I and radiocesium activity concentration ratios decreased to 0.49 and 0.30, respectively. Thus, washing effect was higher for both elements just as we found for the above ground parts, probably due to contaminated soil particles that were retained on the samples before wash for the April 20 sample. Interestingly, for the April 28 sample, no further washing effect was observed for ^{131}I and radiocesium, although it was difficult to explain this discrepancy.

When the activity concentrations of ^{131}I and radiocesium for the bulb without the above ground parts was compared to those for the bulb with the above ground parts for April 20 samples, the level was even smaller for the samples before wash. When the bulb without above ground parts was washed, all the radionuclide concentrations were near the detection limit. The root uptake pathway of these radionuclides may not be the major contributor to the radioactivity in the tulip bulbs; however, it is also true that only a small root uptake could be expected for the bulb sample without the above ground parts.

3.3. Translocation Factor from Above Bround Parts to Bulb. Using the concentration data for ^{131}I and radiocesium, we calculated the translocation factor (F_{tr}, dimensionless), which is defined as follows,

$$F_{tr} = A_{bulb} / A_{above}$$

A_{bulb} is activity concentration in the bulb (Bq g^{-1}) and A_{above} is activity concentration in the above ground parts of the same tulip (Bq g^{-1}). This is similar to the definition used in the IAEA Technical Report Series No. 472.[21] For this calculation, we only used the washed part data to avoid any effect from contaminants attached on the plant above ground part surface. The data for April 20, 2011 in the washed bulb were roughly estimated assuming that the weight was approximately 30 g, as the sample, which had not been destroyed (i.e., cut into pieces) but measured in its original form, gave the concentrations of radionuclides as just information values.

The results are listed in Table 2. Considering the ^{131}I half-life, the ^{131}I concentration in the above ground parts was almost the same for both sampling dates and the radiocesium concentration was also similar. For the sample collected on April 28, 2011, the F_{tr} for ^{131}I was 0.03 while that for radiocesium was 0.47. Thus more Cs was translocated from the above ground parts to the bulb. Oestling et al.[1] reported similar results from leaves to edible part of crops in radiotracer experiments.

Hurtevent et al.[22] investigated translocation of ^{125}I$^-$ in wheat by changing contamination timing; they reported the translocation factor of less than 0.01, which was similar to the present result. It should be noted that the iodine F_{tr} was higher from the flowering stage to the stage closest to grain ripening. We can apply this condition to the tulip samples in this study; it was highly probable that most of the ^{131}I was deposited on the tulips just before they flowered in March and the time of collection was at the bulb growing stage (the end of April), which corresponds to grain ripening. Thus the F_{tr} observed in this study was similar to that of the previous work.[22]

As we already noted, the data for April 20 were just information values; therefore, it would be invalid to compare these values with those observed on April 28, 2011. At least, however, we can conclude that the tendency was the same, that is, the F_{tr} for ^{131}I was much lower than that for radiocesium. This is a rare finding from actual field observations after the FDNPP accident. The Cs F_{tr} for root crops and tubers have been compiled in the IAEA publication,[21] and for root crops are 0.007-0.13 (mean = 0.046) and for tubers are 0.013 to 0.46 (mean=0.12). Thus what we observed in the present study is

TABLE 2: Activity Concentrations of ^{131}I, ^{134}Cs and ^{137}Cs in Washed Tulip Parts and Translocation Factor (F_{tr}) from Above Ground Parts to Bulb on the Sampling Date. The Data for the Bulb Collected on April 20, 2011 were Roughly Estimated, and Consequently, the F_{tr} in Italics are Roughly Estimated Values.

Sampling date	Plant part	Wet mass g	Activity concentration, Bq g^{-1}wet mass			
			^{131}I	^{134}Cs	^{137}Cs	^{129m}Te
April 20, 2011	Above ground	27.1	7.66±0.05	0.61±0.03	0.59±0.02	0.80±0.17
	Bulb	(ca. 30)	*0.38±0.01*	*0.12±0.01*	*0.13±0.01*	*Not detected*
	F_{tr}		*0.05±0.00*	*0.20±0.01*	*0.21±0.02*	-
April 28, 2011	Above ground	41.3	3.11±0.06	0.54±0.02	0.50±0.03	0.73±0.25
	Bulb	33.3	0.09±0.01	0.22±0.01	0.27±0.01	*Not detected*
	F_{tr}		0.03±0.00	0.41±0.02	0.53±0.04	-

±: Counting error

among the highest values. This high F_{tr} can be explained by the plant physiological condition at the time. After flowering, photosynthesized materials in the above ground parts of the tulip plants translocate mainly to the bulb part to produce bulblets.[23] The bulblets are highly active parts in the plant at that time, therefore potassium and its analogue, radiocesium, translocate to the tissue. Our results indicate that plant physiological conditions affect the radiocesium translocation.

4. Conclusions

^{131}I and radiocesium attachment by plant surface structures and then translocation of these radionuclides from the above ground parts to the bulbs were observed in tulip samples collected at J Village in April 2011. It was clear that ^{131}I and radiocesium were attached to the tulip plant surface; similar results were found in several plant species collected in Chiba Prefecture. However, their F_{tr} were completely different for ^{131}I and radiocesium. Much more radiocesium was taken up by plants than ^{131}I from the above ground parts and transferred to the bulbs.

These findings will help us to identify the chemical forms of ^{131}I released at the time of the accident and to understand the importance of the Cs supply timing, including when it is taken up through the plant surface and its translocation. Such data are important to explain the effect of radiation on plants that has been reported.[24] Further studies are necessary to clarify the fates of ^{131}I and radiocesium in plants after the FDNPP accident.

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