Mass Interception Fractions and Weathering Half-lives of Iodine-131 and Radiocesium in Leafy Vegetables Observed after the Fukushima Daiichi Nuclear Power Plant Accident

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ABSTRACT

Background: This study was carried out to provide environmental transfer parameter values to estimate activity concentrations of these radionuclides in agricultural crops when direct contamination occurred.

Materials and Methods: Mass interception fractions (F_Bs) and weathering half-lives (T_ws) of ^{131}I and radiocesium were calculated using openly available monitoring data obtained after the Fukushima Daiichi Nuclear Power Plant accident. F_B is the ratio between the initial radioactivity concentration of a radionuclide retained by the edible part of the plant (Bq$\cdot$kg^{-1} fresh weight [FW]) and the amount of deposited radionuclide in that area (Bq$\cdot$m^{-2}). T_w values can be calculated using activity concentrations of crops decreased with time after the initial contamination.

Results and Discussion: Calculated F_B and T_w values for ^{131}I and radiocesium were mostly obtained for leafy vegetables. The analytical results showed that there was no difference of F_Bs between ^{131}I and radiocesium by t-test; geometric mean values for leafy vegetables cultivated under outdoor conditions were 0.058 and 0.12 m$^2\cdot$kg^{-1} FW, respectively. Geometric mean T_w value of ^{131}I in leafy vegetables grown under outdoor conditions was 8.6 days, and that of radiocesium was 6.6 days; there was no significant difference between T_w values of these radionuclides by Wilcoxon rank sum test.

Conclusion: There was no difference between ^{131}I and radiocesium for F_Bs and T_ws. By using these factors, we would be able to carry out a rough estimation of the activity concentrations of ^{131}I and radiocesium in the edible part of leafy crops when a nuclear accident occurred.

Keywords: Mass Interception Fraction, Weathering Half-Life, Leafy Vegetable, Iodine-131, Radiocesium

Original Research

Introduction

Agricultural crops were directly contaminated with radioactive fallout due to the TEPCO's Fukushima Daiichi Nuclear Power Plant (FDNPP) accident that occurred on March 11, 2011. In affected areas, such as Fukushima Prefecture and its surrounding prefectures, activity concentrations of ^{131}I (T$_{1/2}$ = 8.02 days) and radiocesium (^{134}Cs: T$_{1/2}$ = 2.065 years+^{137}Cs: T$_{1/2}$ = 30.2 years) in some leafy vegetable samples exceeded the provisional limits at that time, 2,000 Bq$\cdot$kg^{-1} and 500 Bq$\cdot$kg^{-1}, respectively (from April 1, 2012, the standard limit for total radiocesium was set to 100 Bq$\cdot$kg^{-1}). When the radioactivity

concentration(s) in a crop sample in a certain municipality exceeded the limit, the municipality was not allowed to sell their crop products. Then, once a week, radioactivity monitoring needed to be carried out to confirm that the activity concentration data obtained in the following continuous 3 weeks were lower than the limit values, which were the required conditions to sell and to eat the products again.

When a new nuclear accident happens, we can use the monitoring data obtained in 2011 to estimate the initial radioactivity levels in leafy vegetables and how fast the radioactivity in the crops would decrease with time. For that purpose, it is better to parameterize the related factors, that is, mass interception fractions (F_Bs) and weathering half-lives (T_ws). Following deposition of radionuclides on vegetables, the initial radionuclide activity concentrations of the vegetables can be estimated by F_B, which is the ratio between the initial radioactivity concentration of a radionuclide retained by the edible part of the plant (Bq·kg⁻¹ fresh weight [FW]) and the amount of deposited radionuclide in that area (Bq·m⁻²). T_w is the time taken for the quantity of radionuclides on and in plants to be reduced by half due to weathering (rain and window), tissue ageing, mass increase, and other processes. Thus, following activity concentrations in the vegetable can be estimated using T_w.

These parameter data were available in the International Atomic Energy Agency (IAEA) Technical Report Series No. 472 (TRS 472) [1]; the values were originally from laboratory experiments and observation results after the Chernobyl accident. Recently published technical document of the IAEA No. 1927 (TECDOC 1927) [2] summarized the F_B and T_w observed after the FDNPP accident, but no detailed analysis was reported yet. Therefore, in this study, we have re-collated these values and checked the differences among crop species.

Materials and Methods

Because the radioactivity monitoring of fresh vegetables was not carried out immediately after the FDNPP accident started, it was difficult to obtain the initial activity concentrations in those vegetables. According to the TECDOC 1927 [2], we can back-calculate the initial activity concentrations from continuous monitoring data, which were available from the food monitoring results in March to April 2011 reported by the Ministry of Health, Labour and Welfare (MHLW), Japan [3]. The image of the calculation is shown in Fig. 1.

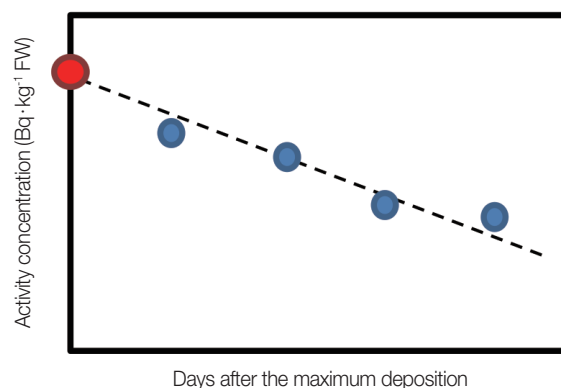The exponential decline in radioactivity concentration in

Fig. 1. An image of back-calculating method to obtain initial radioactivity concentration (red) of a radionuclide in leafy vegetable. The dotted line (fitting result of the continuous monitoring data) was used to calculate weathering half-life. FW, fresh weight.

the edible part of the crop over time is explained as follows.

$$A_t = A_0 \exp(-\lambda_{eff} t) \qquad (1)$$

where A_t is activity concentration (Bq·kg⁻¹ FW) of ¹³¹I, ¹³⁷Cs or total radioactive Cs (¹³⁴Cs+¹³⁷Cs) at time t (day), A_0 is the expected initial activity concentration (Bq·kg⁻¹ FW), and λ_{eff} is the radionuclide loss rate constant. Using the concentration data, a best fit exponential line for each leafy vegetable sample set collected in a certain municipality was calculated using KaleidaGraph software version 4.5 (Synergy Software, Reading, PA, USA).

Using A_0 and total deposition amounts in the area A_s (Bq·m⁻²), mass interception fraction F_B is calculated as follows.

$$F_B = \frac{A_0}{A_s} \qquad (2)$$

For radiocesium, A_s was calculated using the second soil monitoring results among nine sampling campaigns carried out in 2011–2016 by *in situ* Ge gamma-ray spectrometry, which was conducted for ¹³⁴Cs and ¹³⁷Cs (decay corrected to March 1, 2012). Details of the soil data are found in our report [4]. For ¹³¹I, we used activity concentrations of measured ¹³¹I in soil and estimated ¹³¹I from ¹²⁹I in soil carried out by the Ministry of Education, Culture, Sports, Science and Technology and the Nuclear Regulation Authority, Japan [5]. All the data for ¹³¹I, ¹³⁴Cs, and ¹³⁷Cs were corrected to March 20, 2011 because the heaviest radioactivity deposition was observed in large areas in eastern Japan on March 20–22 [6]. Corresponding soil data were based on data in each municipality

in Fukushima Prefecture and the geometric mean value was calculated to obtain A_s values. For other areas, if there were no corresponding data for ^{131}I and radiocesium in a municipality, then the data in each prefecture's deposition monitoring site were applied.

To calculate T_w, effective half-life (T_{eff}, in day) needs to be calculated first using λ_{eff} as follows.

$$T_{eff} = \frac{\ln 2}{\lambda_{eff}} \tag{3}$$

The T_{eff} is expressed as follows.

$$\frac{1}{T_{eff}} = \frac{1}{T_w} + \frac{1}{T_p} \tag{4}$$

where T_p is the physical half-life of the radionuclide. Using these equations, T_w is then calculated as follows.

$$T_w = T_{eff} \times \frac{T_p}{T_p - T_{eff}} \tag{5}$$

We checked the correlation factor between time (day) and the logarithm of the activity concentrations by Student t-test to accept the data set for F_B and T_w calculations or not. Firstly based on p-values, we considered that if $p < 0.05$ and the coefficient of determination (R^2) value was higher than 0.6, then the data set was used for T_{eff} calculation. Next, if R^2 was higher than 0.9, then we included the data set to calculate T_{eff} even if p-values were more than 0.05.

Results and Discussion

After examination of the food monitoring results by MHLW [3], leafy vegetable data obtained in Fukushima Pre-fecture and surrounding five prefectures (Chiba, Gunma, Ibaraki, Saitama, and Tochigi) were found. Numbers of data sets used for decline fittings by Equation (1) were 15 for radiocesium (eight datasets were from Fukushima Prefecture), and 42 for ^{131}I (19 datasets were from Fukushima Prefecture). Crops were celery, crown daisy, Japanese honeywort, kakina, komatsuna, kukitachina, mizuna, parsley, shinobufuyuna, shiso, and spinach. For comparison, we included heads of cabbage and flowering shoots of *Brassica* species (e.g., broccoli). Additional data from our experiments for giant butterbur [6] were also included in the analysis. Summary of the data analysis carried out using JMP 9.0 (SAS Institute Inc., Cary, NC, USA) is shown in Supplementary Table S1.

1. Mass Interception Fractions of ^{131}I and Radiocesium

By applying Equation (1), we successfully obtained A_0 values for crops mentioned above with good fitting results. However, because some areas were difficult to find corresponding total deposition data, A_s, thus, the numbers of F_B values were smaller compared to those listed above. In addition to the F_B calculation using openly available data sets, because we measured edible wild plants on March 28, 2011 and deposition samples [7], the data were also included for analysis.

F_B values should be affected by the plant shape (e.g., cabbage, spinach) and the cultivation conditions, i.e., outside and indoor (greenhouse and tunnel) cultivations, thus F_B data were summarized in the leafy vegetables and other shape green vegetables (head and flowering shoots), and also classified into outdoor and indoor cultivations, if possible. The F_B data are summarized in Table 1; because we lacked knowledge about the distribution characteristics of the data for all groups,

Table 1. Mass Interception Fractions (F_B) of Radiocesium and ^{131}I and for Leafy Vegetables Observed after the Fukushima Daiichi Nuclear Power Plant Accident in 2011

Radionuclide	Cultivation condition	Crop type	n	F_B (m$^2 \cdot$kg^{-1} FW)				
				AM	SD	GM	GSD	Range
Cs	Outdoor	Leafy vegetables	15	0.210	0.220	0.130	2,8	0.020–0.720
		(Spinach)	6	0.250	0.270	0.140	3.4	0.026–0.720
		Other green vegetables	2	0.082	0.060	0.070	2.2	0.040–0.120
	Indoor	Leafy vegetables	4	0.014	0.010	0.011	2.2	0.005–0.028
I	Outdoor	Leafy vegetables	28	0.092	0.091	0.057	2.8	0.012–0.300
		(Spinach)	15	0.063	0.048	0.048	2.2	0.012–0.200
		Other green vegetables	9	0.008	0.007	0.005	3.9	0.001–0.022
	Indoor	Leafy vegetables	11	0.039	0.052	0.015	5.7	0.001–0.140
		(Spinach)	4	0.046	0.061	0.025	3.2	0.010–0.140

FW, fresh weight; n, sample size; AM, arithmetic mean; SD, standard deviation; GM, geometric mean; GSD, geometric standard deviation.

arithmetic mean (AM), standard deviation (SD), geometric mean (GM), and geometric standard deviation (GSD) are provided with ranges of the F_B data. Using the sample groups having the largest n for ^{131}I and radiocesium were examined for their data distribution. Based on the Shapiro-Wilk normality test result, the F_B of leafy vegetables cultivated under outdoor conditions did not follow a normal distribution for each radionuclide. When the logarithms of the F_B data were calculated, the data agreed to a log-normal distribution for each radionuclide. We assumed that log-normal distributions were suitable for other groups, thus, data comparisons were carried out using the logarithm of the F_B data.

From the results for F_B of Cs, leafy vegetables cultivated under outdoor conditions tended to show higher values (GM = 0.13 $m^2 \cdot kg^{-1}$ FW) than that for other green vegetables (GM = 0.07 $m^2 \cdot kg^{-1}$ FW) but the numbers of data were too small for statistical analysis. For F_B of ^{131}I, the logarithm of the data for leafy vegetables cultivated under outdoor conditions (GM = 0.057 $m^2 \cdot kg^{-1}$ FW) was significantly higher than those from indoor cultivation (GM = 0.015 $m^2 \cdot kg^{-1}$ FW) by Wilcoxon rank sum test ($p = 0.018$). However, when we used the data only for spinach, no difference was found between these different cultivation conditions. When the data of leafy vegetables were compared to those of other green vegetables both grown under outdoor conditions, a significant difference was observed by the same test ($p < 0.001$). From these results, we assumed that crop type (shape) and/or species would affect the F_B value rather than cultivation conditions.

For leafy vegetables grown under outdoor conditions, the F_B data for radiocesium were significantly higher than those for ^{131}I by Wilcoxon rank sum test ($p = 0.023$). According to IAEA TRS 472 [5], F_B values for grass observed after the Chernobyl accident were 1.1 $m^2 \cdot kg^{-1}$ dry weight for Cs and 0.7 $m^2 \cdot kg^{-1}$ dry weight for I, thus F_B of Cs was slightly higher. Our present results showed a similar tendency; however, more specifically, we compared the F_B values for spinach. The result showed no difference between ^{131}I and radiocesium by Wilcoxon rank sum test ($p > 0.05$). From these results, although the chemical forms of ^{131}I and radiocesium were different, F_B values were not much different. Yet, further studies are needed to clarify the chemical form effect on F_B values by plants.

Values in IAEA TRS 472 were collated on a dry weight basis; thus, it was difficult to compare the data directly with our data. Fresh mass to dry mass conversion factors are needed for this comparison; according to the TRS 472, dry matter contents for grass species (e.g., Timothy grass, Standard crested grass, Meadow fescue) were 20%–39%. When the median value, ca. 30% was used to calculate FW basis F_B for grass in IAEA TRS 472, the calculated values were 0.33 $m^2 \cdot kg^{-1}$ FW for Cs, and for 0.21 $m^2 \cdot kg^{-1}$ FW for I. The GM of F_B values for radiocesium and ^{131}I in the present study was about one-third of the TRS 472 values, respectively, probably because only edible parts were considered in this study.

2. Weathering Half-lives of ^{131}I and Radiocesium

T_w data were calculated using Equations (3)–(5), and the summaries of the results are shown in Table 2. Similar to the F_B data analysis, using the sample group having the largest n was examined for its data distribution, and we found that the data distributed log-normally. Thus for T_w data, we also assumed that all data would distribute log-normally thus logarithm of the T_w data were used for analysis.

T_w values of ^{131}I in leafy vegetables obtained under outdoor

Table 2. Weathering Half-life (T_w) of Radiocesium and ^{131}I and for Leafy Vegetables Observed after the Fukushima Daiichi Nuclear Power Plant Accident in 2011

Radionuclide	Cultivation condition	Crop type	n	T_w (d)				
				AM	SD	GM	GSD	Range
Cs	Outdoor	Leafy vegetables	11	6.5	1.9	6.2	1.4	3.1–8.9
		(Spinach)	6	6.7	2.0	6.3	1.5	3.1–8.9
		Other green vegetables	2	6.8	1.4	6.7	1.2	5.8–7.8
	Indoor	Leafy vegetables	5	13.0	5.0	12.0	1.4	8.6–21.0
I	Outdoor	Leafy vegetables	26	9.7	5.3	8.4	1.7	3.0–23.0
		(Spinach)	17	11.0	5.7	9.3	1.8	3.0–23.0
		Other green vegetables	9	9.0	7.1	7.3	1.9	2.2–27.0
	Indoor	Leafy vegetables	14	11.0	5.8	9.9	1.7	4.2–24.0
		(Spinach)	4	7.7	2.3	7.4	1.4	5.1–10.0

n, sample size; AM, arithmetic mean; SD, standard deviation; GM, geometric mean; GSD, geometric standard deviation.

cultivation conditions ranged from 3.0–23 days (GM = 8.4 days), and that for radiocesium ranged 3.1–8.9 days (GM = 6.2 days). There were no significant differences between T_w values of these radionuclides by t-test. In our recent publication, we carefully examined the fate of ^{131}I and radiocesium on plants, and we concluded that these similar T_w values between ^{131}I and radiocesium may be affected by the crop mass increase (dilution effect) rather than decrease by weathering (e.g., wash-out with rain) [6, 8].

For T_w values of Cs, leafy vegetables cultivated under outdoor and indoor conditions were different by Wilcoxon rank sum test ($p < 0.01$). Because the GM of T_w of vegetables cultivated under indoor conditions was longer, it was expected that washing effect by rain caused this difference; however, the crop types were also different in these datasets, thus the analysis results may also be affected by the crop types/species as we found for F_B of Cs.

Further comparison was carried using T_w values for ^{131}I; however, no difference was observed between T_ws for outdoor (GM = 8.4 days) and those for indoor (GM = 9.9 days) cultivation conditions by Wilcoxon rank sum test. More specific data comparison was carried out using spinach T_w data of [131]I. Because the number of data was small for indoor cultivation ($n = 4$), the result was not reliable enough, but we did not find a difference between the conditions by Wilcoxon rank sum test. Moreover, we compared the T_w of ^{131}I data between leafy vegetables and other green vegetables grown under outdoor conditions, but we did not find a significant difference between these vegetable types as well.

From these results, it was difficult to explain why T_w of Cs between crops grown under outdoor and indoor conditions was different, and vice versa, it was not clear why T_w of ^{131}I between crops grown under outdoor and indoor conditions and different crop types was not different. Further studies are needed to clarify the weathering effect for these radionuclides.

Conclusion

In the present study, we reported F_B and T_w values of ^{131}I and radiocesium using openly available monitoring data obtained after the FDNPP accident. The purpose of this study was to provide environmental transfer parameter values to estimate activity concentrations of these radionuclides in agricultural crops when direct contamination occurred. The study results showed that there was no difference between

[131]I and radiocesium for both parameter values. The results showed that the F_B values may be affected by crop types (shapes) and/or species but the difference was not clear for T_ws. By using these values presented in this study, we would be able to carry out a rough estimation of the activity concentrations of ^{131}I and radiocesium in the edible part of leafy crops when these radionuclides were released into the atmosphere. However, because the effect of mass dilution by crop growth may lead to overestimation or underestimation, sufficient attention should be paid to apply these factors.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

Author Contribution

Conceptualization: Tagami K, Uchida S. Writing - original draft: Tagami K. Writing - review & editing: Uchida S.

Supplementary Materials

Supplementary materials can be found via https://doi.org/10.14407/jrpr.2021.00164.

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