Radiocesium Distribution in Soil and *Brassica napus* Grown in Contaminated Soils

Yoshinori Kanayama^{1*,**}, Michiaki Omura^{1**}, Takeshi Goryo^{2***}, Tomohisa Shigeta^{2****},
Tomoki Shibuya¹ and Yutaka Nakai¹

¹Graduate School of Agricultural Science, Tohoku University, Sendai 980-0845, Japan

²euofins EAC Corporation, Edogawa-ku, Tokyo 132-0025, Japan

Brassica napus was cultivated in three test fields exposed to different radionuclide contamination levels 1.5 years after the Fukushima Daiichi nuclear power plant accident to investigate the correlation between soil contamination and radiocesium activity concentration (RCs conc.) in plants. The correlations between total and exchangeable RCs conc. were strong in the plow layer (L1) and the layer 5 cm below L1 (L2). Water-soluble radiocesium was not detected in either layer, and little radiocesium penetration was observed below L2. Correlation coefficients between RCs conc. in each plant organ and soil layers were calculated. Correlations between RCs conc. in each vegetative organ and total and exchangeable RCs conc. in L1 and correlations between vegetative organs were mostly strong during vegetative growth and flowering periods. Correlations between RCs conc. in flowers with that in L1, L2, and other vegetative organs were not significant. RCs conc. in L1 may more directly affect concentrations in roots and shoots. RCs conc. in oil extracted from seeds was measured, and the results suggested that radiocesium was not detectable in the oil fraction. Our data regarding radiocesium in soil and plants and their correlations obtained by extensive cultivation in contaminated soils could be useful to gain a better scientific understanding of radiocesium transport and accumulation and to provide useful information for the future utilization of areas with low contamination levels.

Key Words: contamination, exchangeable cesium, Fukushima Daiichi nuclear power plant, rape.

Introduction

The Fukushima Daiichi nuclear power plant accident was caused by the Great East Japan Earthquake and the resulting tsunami on March 11, 2011. The deposition of radionuclides from the accident spread over Fukushima and Miyagi Prefectures and to certain nearby prefectures (Hirose, 2016). Although short-lived radionuclides, such as ¹³²I, decayed in a short time, the presence of long-lived radionuclides, such as ¹³⁴Cs and

¹³⁷Cs, is resulting in long-term environmental effects. One of these long-term effects is the uptake of the radionuclides by plants.

There have been some studies on the radiocesium contamination of wild plants, such as grasses, trees, and bamboo, after the accident to monitor environmental radioactive contamination (Higaki et al., 2014; Kaunisto et al., 2002; Mori et al., 2012; Terashima et al., 2014). Moreover, studies on the radiocesium contamination of crops cultivated in fields near the nuclear power plant after the accident were mainly performed in rice. These studies focused on soil-to-rice transfer coefficients (Endo et al., 2013); the effects of applying potassium, zeolite, and vermiculite on radiocesium uptake (Fujimura et al., 2013) and the uptake and translocation of radiocesium (Kondo et al., 2015; Nobori et al., 2014).

Although model studies using artificially contaminated media have been performed on vegetables and oil crops (Bengtsson et al., 2013; Choi et al., 2008; Nisbet

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* Corresponding author (E-mail: yoshinori.kanayama.a7@tohoku.ac.jp).

** These authors equally contributed to the paper.

*** Present address: Euofins Nihon Kankyo K.K., Kanazawa-ku, Yokohama 236-0003, Japan

**** Retired

and Shaw, 1994; Yasutaka et al., 2014), studies on the effect of radiocesium exposure on vegetables and oil crops cultivated in fields near the nuclear power plant after the accident are difficult to perform, but are important. Radiocesium activity concentrations (RCs conc.; the concentrations of ^{134}Cs and ^{137}Cs) and transfer coefficients have been reported for some vegetables contaminated with radionuclides after the Chernobyl power plant accident and by surface nuclear tests (Kozhakhonov et al., 2014; Paasikallio et al., 1994; Penrose et al., 2016). Most recently, after the Fukushima Daiichi nuclear power plant accident, RCs concs. have been reported in leafy vegetables cultivated in the fields near the power plant (Aung et al., 2015; Djedidi et al., 2016; Ohse et al., 2015). These studies simply determined radiocesium activity and transfer coefficients in various species and cultivars, and further detailed scientific research regarding the relationship between soil contamination and RCs conc. in plants is still required in various crop species cultivated in the contaminated areas.

Crops that are industrially useful, show potential for phytoremediation, and are suitable for extensive cultivation in emergency situations are expected to be grown in contaminated as well as neighboring areas. The present study focused on *Brassica napus* because its flowers have ornamental value and this plant is a valuable vegetable and seed oil crop. In addition, *B. napus* is suitable for extensive cultivation and is a member of the Brassicaceae family, which is known to include crop species with comparatively higher radiocesium transfer coefficients; thus, this species shows potential for phytoremediation (Choi et al., 2008; Fuhrmann et al., 2002; Lasat et al., 1997). *B. napus* was in fact previously studied for its potential use in environmental recovery from the tsunami disaster caused by the Great East Japan Earthquake (Yong et al., 2015). We cultivated *B. napus* in three fields with different radiocesium contamination levels and investigated correlations between soil contamination and plant radiocesium accumulation. We further determined the RCs conc. in seed oil. Although direct deposition of radiocesium onto plants by fallout should be considered in addition to soil uptake during the year of the accident (Ohse et al., 2015; Sato et al., 2015), we analyzed radiocesium soil uptake assuming no direct deposition onto plants by fallout. Since the effect of radiocesium contamination including direct deposition on RCs conc. in seed oil has been reported in *B. napus* (Hirayama et al., 2011), the effect of soil uptake without direct deposition was investigated in this study. In addition, Brassicaceae includes many important crop species; different organs of these species, such as radish roots, cabbage leaves, and rapeseed flower buds and seeds are used as vegetables and to extract oil. Therefore, examining the RCs conc. and transfer coefficients of each organ at different developmental phases is important.

Our data using multiple plant organs, developmental stages, states of radiocesium, and soil layers will contribute valuable information.

Materials and Methods

Test fields and plant materials

Test fields are indicated as FLD1, FLD2, and FLD3, which have been used as upland fields; the distances of each field from the Fukushima Daiichi nuclear power plant and their soil types are presented in Figure 1. The areas of FLD1, FLD2, and FLD3 are approximately 3000, 7000, and 3500 m², respectively. *B. napus* ‘Kizakinonatanne’, which can grow in the climate of the test fields, was cultivated by broadcast seeding during October 2012, 1.5 years after the accident. Plowing was carried out, and preplanting fertilizer was not applied to any test fields. Five g·m⁻² of supplemental composite fertilizer (N:P:K = 16%:10%:14%) was applied to FLD1 and FLD2. Although the average lengths of spikes were similar among the three test fields, the average plant heights (60, 75, and 30 cm in FLD1, FLD2, and FLD3, respectively) varied, which could be because of different conditions, including supplemental fertilizer.

Sampling, sample preparation, and measurement of radioactivity

Soil was sampled using a corer from the plow layer (L1; average 14 cm), a layer 5-cm below L1 (L2), and a layer 5-cm below L2 (L3) at three different plots in

Fig. 1. The location of test fields, FLD1, FLD2, and FLD3, including the distance from the Fukushima Daiichi nuclear power plant, air dose rates, and soil types. Air dose rates were measured at 1 m above the ground surface, and values indicate means with SD (n = 3). Soil types were determined according to the database of the National Institute for Agro-Environmental Sciences in Japan (http://agrimesh.dc.affrc.go.jp/soil_db/map_select_figure.shtml). A blank map from CraftMAP (<http://www.craftmap.box-i.net/>) was used.

each test field during November 2012 (FLD1 and FLD2) and February 2013 (FLD3). The three plots were located in the center of each field, 5 m from the opposite ends of each field. After total radiocesium activity was determined in the soil sampled from each layer, a volume of water 5-fold of that of the dry soil (60 g) was added to the soil; the sample was shaken for 1 h and filtered using a 0.45- μ m membrane filter to determine water-soluble radiocesium activity (Committee of Standard Methods for Soil Analysis, 1986). A volume of 1 N ammonium acetate (pH 7.0) 20-fold of that of the dry soil (60 g) was added to the soil after the water extraction. Then, the sample was shaken for 1 h and filtered using a 0.45- μ m membrane filter to determine the exchangeable fraction of radiocesium activity. The method of the Committee of Standard Methods for Soil Analysis (1986) was applied to prepare the exchangeable fractions. Leaves, stems, roots, and spikes were sampled from 1.7 m² of three different plots in each test field, washed thoroughly to avoid soil contamination, and shredded using a food processor to determine radiocesium activity. The plant samples were collected from the same sites as those of soil sampling. The seeds sampled at harvest were ground, extracted with n-hexane, dried over anhydrous sodium sulfate, and distilled to remove hexane. Radiocesium activity in the seeds was determined before extraction in the seed oil and after the extraction in the residue.

Gamma rays from the samples were measured for ¹³⁴Cs and ¹³⁷Cs using a germanium semiconductor detector (GEM20-70; SEIKO EG&G, Japan) with a resolution of 1.8 keV (for the 1.33 MeV line of ⁶⁰Co) and a relative efficiency of 23%. The detection time ranged from 2000 to 5000 s per sample. All values are shown after a decay correction for November 29, 2012. The average sample masses (kg) were 0.142 (soil), 0.284 (shoots in February), 0.067 (roots in February), 0.059 (flowers in April), 1.17 (shoots in April), and 0.271 (roots in April). The detectors were calibrated with standards, and the results were corrected for the summing effect. The detection limit for radiocesium activity was determined as three times the standard deviation (SD) using counting statistics based on the method described by Cooper (1970). Means were calculated when activity was detected in one or more plots in each field. In a few samples, the values measured were used only for the calculation when some activity was obtained but did not exceed the detection limit.

Statistics

Correlation coefficients were calculated using R version 3.1.1 (R Core Team, 2014).

Results and Discussion

Radiocesium activity concentrations in the soil

The total and exchangeable radiocesium fractions in L1, L2, and L3 of the three test fields are summarized

in Tables 1 and S1. A ¹³⁴Cs to ¹³⁷Cs activity ratio of 1 has been used for the Fukushima fallout (Ramzaev et al., 2013); a ratio of 0.59 was calculated in November 2012, whereas the ratios in the soils were between 0.44 and 0.79 in the present study. Although the range of the ratios was wide, the ratios of ¹³⁴Cs with short half-life were 0.44 or more, and the ratios in the layers where the values were obtained in all three plots were between 0.53 and 0.60 (Table S1); therefore, contamination was likely derived from the Fukushima fallout. RCs conc. in the fixed fraction [total radiocesium – (exchangeable and water-soluble radiocesium)], from which radiocesium accessibility by plants is considered to be low, was much higher than that in water-soluble and exchangeable fractions, from which radiocesium is accessible to plants. Water-soluble radiocesium was not detected in the three layers. RCs conc. in all fractions decreased from L1 to L3 and was very low in L3. Kusaba et al. (2016) reported much higher RCs conc. in the surface layer (0–5 cm) of an orchard compared to that at a 15–30 cm depth in several types of soil surface management. Correlations between the exchangeable and fixed fractions of radiocesium in L1 and L2 are summarized in Table 2. Correlation coefficients were calculated for RCs conc. at five plots in FLD2 and FLD3, where exchangeable radiocesium was detected. Correlations between the exchangeable and fixed fractions of radiocesium were strong in L1 and L2. Correlations of the two fractions between L1 and L2 were not significant.

Our results revealed that water-soluble radiocesium was not detected 1.5 years after the accident at our detection limit and that there was very little radiocesium penetration deeper than 5 cm below L1. Since most of the fallout radiocesium was deposited at less than 5 cm below the surface (Kusaba et al., 2016), the low RCs concs. detected in L2 and L3 were considered to be due to plowing. The ratios of the exchangeable radiocesium fraction to total radiocesium in L1 and L2 were almost identical, i.e., 0.14 ± 0.019 in L1 and 0.13 ± 0.017 in L2 (means $\pm$ SD) in FLD3 (fine-grained Yellow soil), and the correlations between exchangeable and total radiocesium were strong in both L1 and L2, suggesting that the exchangeable radiocesium fraction was present in a certain percentage of total radiocesium. If the clay minerals were the same between layers, the fixation ability of radiocesium would be similar; therefore, the ratios of the exchangeable RCs concs. were assumed to be almost the same. Although previous studies (Endo et al., 2013; Terashima et al., 2014) also indicated that the majority of radiocesium was present within 15 cm of the surface after the 2011 fallout, they did not measure exchangeable and fixed-radiocesium fractions. If the ratio of exchangeable to total radiocesium was variable, a reliable estimation of the transfer coefficient between soil and plants using the total RCs conc. in the soil would be a challenge. Paasikallio et al. (1994) reported

Table 1. RCs concs. in each soil layer in the test fields.

		$^{134}\text{Cs}+^{137}\text{Cs}$ (Bq·kg ⁻¹ DW)			Soil type
			Mean ^z	SD ^z	
FLD1	L1	tl ^y	117	63	Muck soil
		ex	ND ^x	—	
	L2	tl	41	31	
		ex	ND	—	
	L3	tl	7	—	
		ex	ND	—	
FLD2	L1	tl	727	740	Medium- and coarse-textured Gray Lowland soil
		ex	134	78	
	L2	tl	13	—	
		ex	ND	—	
	L3	tl	12	—	
		ex	ND	—	
FLD3	L1	tl	10604	4202	Fine-grained Yellow soils
		ex	1431	389	
	L2	tl	2330	2936	
		ex	329	426	
	L3	tl	73	101	
		ex	19	—	

Detection limit for ND: 3.34–6.08 Bq·g⁻¹ DW for tl-¹³⁴Cs and 3.60–5.50 Bq·g⁻¹ DW for tl-¹³⁷Cs; 11.9–19.7 Bq·kg⁻¹ DW for ex-¹³⁴Cs, 11.7–22.0 Bq·kg⁻¹ DW for ex-¹³⁷Cs; 14.2–42.8 Bq·kg⁻¹ DW for wt-¹³⁴Cs, and 15.4–44.2 Bq·kg⁻¹ DW for wt-¹³⁷Cs. Since some set times were used as measurement times, detection limits were different among measurements.

^z Mean values are shown with SD (n=3). The plot in which radiocesium was not detected was excluded from the calculation of mean value.

^y The abbreviations ex and tl indicate exchangeable and total RCs conc., respectively.

^x ND indicates not detected.

that transfer coefficients between soil and plants were variable immediately after the nuclear accident and gradually became constant. At the time of our experiment, reliable transfer coefficients between the soil and plants could be estimated using the total RCs conc. in the soil because the correlation between exchangeable and total radiocesium was strong. Because radiocesium is strongly fixed to a frayed edge site over time, which depends on the type of clay minerals, the radiocesium interception potential (RIP), an index of the fixation ability, was reported for the Tohoku region (MAFF and NARO, 2015). The RIP was 1440 mmol·kg⁻¹ for Muck soils (the soil type of FLD1), 1860 mmol·kg⁻¹ for Gray Lowland soils (FLD2), and 1720 mmol·kg⁻¹ for Yellow soils (FLD3). Although the RIP for Muck soils is low, the value is not markedly low, such as the 352 mmol·kg⁻¹ value for High-humic Andosols. The similar ratios of exchangeable to total radiocesium in FLD2 and FLD3 (0.13 ± 0.025 and 0.14 ± 0.019, respectively; mean ± SD) could be due to the similar RIP in FLD2 and FLD3.

Radiocesium activity concentrations in plants

Relatively higher radiocesium activity was measured in plants harvested from FLD3, and RCs concs. in certain *B. napus* organs are summarized in Table 3; RCs

Table 2. Correlation coefficients among exchangeable and total RCs concs. in two soil layers.

	L1 tl ^z	L2 ex	L2 tl
L1 ex	0.988***	0.355	0.372
L1 tl		0.259	0.283
L2 ex			0.999***

^z The abbreviations ex and tl indicate exchangeable and total RCs concs., respectively.

^y Correlation coefficients were calculated for RCs concs. (¹³⁷Cs) at five plots in FLD2 and FLD3, where radiocesium activity in the exchangeable fraction was detected in L1. L1, plow layer and L2, layer 5 cm below L1. Significant correlations are indicated with ** (*P*<0.01) and *** (*P*<0.001).

concs. in February, the vegetative growth period, and in April, the flowering period, are shown. The ¹³⁴Cs to ¹³⁷Cs activity ratio in plants was between 0.43 and 0.63. While radiocesium was undetectable in almost all the plant samples from the three plots in FLD1 and one plot in FLD2 (detection limit: 0.98–7.16 Bq·kg⁻¹FW for ¹³⁴Cs and 1.01–7.25 Bq·kg⁻¹FW for ¹³⁷Cs), where exchangeable radiocesium was not detected, radiocesium was detected in half of the plant samples from two plots in FLD2, where exchangeable radiocesium was detected. These results indicate that the radiocesium dis-

Table 3. RCs concs. and transfer coefficients (TF) in *Brassica napus* organs during the vegetative growth (February) and flowering (April) periods in FLD2 and FLD3.

				Bq·kg ⁻¹ FW		¹³⁴ Cs/ ¹³⁷ Cs	TF ^y
				Mean ^z	SD ^z		
February	FLD2	Shoot	¹³⁴ Cs	ND ^x	—	—	—
			¹³⁷ Cs	2.36	0.03	—	0.0035
		Root	¹³⁴ Cs	ND	—	—	—
			¹³⁷ Cs	ND	—	—	—
	FLD3	Shoot	¹³⁴ Cs	10.40	2.98	0.63	0.0027
			¹³⁷ Cs	16.63	1.68	—	0.0024
		Root	¹³⁴ Cs	16.52	0.46	0.43	0.0040
			¹³⁷ Cs	38.10	16.45	—	0.0056
April	FLD2	Flower	¹³⁴ Cs	ND	—	—	—
			¹³⁷ Cs	5.35	—	—	0.0053
		Shoot	¹³⁴ Cs	ND	—	—	—
			¹³⁷ Cs	ND	—	—	—
		Root	¹³⁴ Cs	ND	—	—	—
			¹³⁷ Cs	4.44	2.57	—	0.0044
	FLD3	Flower	¹³⁴ Cs	8.87	—	0.56	0.0016
			¹³⁷ Cs	15.86	10.43	—	0.0021
		Shoot	¹³⁴ Cs	8.68	3.72	0.51	0.0023
			¹³⁷ Cs	17.10	8.35	—	0.0025
		Root	¹³⁴ Cs	9.29	2.66	0.49	0.0024
			¹³⁷ Cs	18.86	3.82	—	0.0028

Detection limit (FLD2 and FLD3): 1.69–10.4 Bq·kg⁻¹ FW for ¹³⁴Cs and 2.11–10.1 Bq·kg⁻¹ FW for ¹³⁷Cs in February and 1.14–11.3 Bq·kg⁻¹ FW for ¹³⁴Cs and 1.40–8.24 Bq·kg⁻¹ FW for ¹³⁷Cs in April. Since some set times were used as measurement times, detection limits were different among measurements.

^z Mean values are shown with SD (n=3). The plot in which radiocesium was not detected was excluded from the calculation of mean value.

^y Transfer coefficients from the soil to plant organs were calculated as the ratio of the RCs conc. [¹³⁴Cs or ¹³⁷Cs, Bq·kg⁻¹ FW] in each plant organ to that [Bq·kg⁻¹ DW] in the soil (total) in L1. L1, plow layer.

^x ND indicates not detected.

tribution observed in the soil of FLD1 did not result in severe contamination of *B. napus* products. The transfer coefficients from soil to plant organs were calculated as the ratio of the RCs conc. in each plant organ [¹³⁴Cs or ¹³⁷Cs, Bq·kg⁻¹ FW] to that in the soil [Bq·kg⁻¹ DW]; the obtained ratios ranged between 0.0016 and 0.0056. Although the transfer coefficient is affected by exchangeable potassium concentration, it was not measured in this study.

RCs concs. in roots were found to be higher than that in shoots of rice grown in paddy fields (Endo et al., 2013), whereas the differences in RCs concs. between organs were small in cabbage and barley, and RCs concs. in roots were rather lower than those in the leaves of carrots (Nisbet and Shaw, 1994). In FLD3, it appears that the RCs conc. was high in roots during the vegetative growth period and was similar between shoots and roots during the reproductive growth period. Cesium is considered to be phloem-mobile, similar to potassium. However, large differences in cesium concentrations between organs were found in the eggplant, which was one of the four crop species tested during an ionic analysis of non-radioactive elements (Shibuya et al., 2015), whereas differences in potassium concen-

trations between organs were small in all four species. These results suggest that organ-specific cesium accumulation varies with crop species.

Certain Brassicaceae species have high radiocesium transfer coefficients (Lasat et al., 1997). Transfer coefficients in shoots have recently been reported in some *B. napus* cultivars cultivated in fields contaminated by the nuclear power plant accident (Djedidi et al., 2016). The values were calculated using radiocesium per g DW of plants, whereas our values were calculated using radiocesium per g FW of plants. If our values obtained from shoots are converted using 90% water content (Albert et al., 2012), the values are approximately fall in the range of the values obtained in the previous study. Brassicaceae crops have a high potential for radiocesium uptake from the soil, although this is not equivalent to *Amaranthus* (Chu et al., 2015), and are expected to be useful for the phytoremediation of radiocesium contamination (Lasat et al., 1997); however, the transfer coefficients in shoots and flowers described in this study and yield (1.3 t·ha⁻¹, 1999, Statistics from Ministry of Agriculture, Forestry and Fisheries, Japan) have revealed difficulties in using *B. napus* for phytoremediation.

Correlations between each plant organ and the soil layer

The correlation coefficients between RCs conc. in *B. napus* organs and soil layers (Table 4) are presented with scatter plots (Fig. S1), illustrating the effect of spatial distribution and the solubility of radiocesium on RCs concs. in plant organs. Correlation coefficients were calculated at five plots in FLD2 and FLD3 where exchangeable radiocesium was detected. While RCs concs. were very low in L3 and the water-soluble radiocesium fractions of L1 and L2 were low, exchangeable radiocesium fractions of L1 and L2 were comparatively high (Table 1; Table S1); therefore, total and exchangeable RCs concs. in L1 and L2 were used for the calculation. Correlations of each vegetative organ with fixed- and exchangeable fractions of L1 were mostly strong in both February (the vegetative growth period) and April (the flowering period). The correlations between flower RCs concs. and both fractions of L1 and L2 were not significant. Correlations between RCs concs. in the plant organs are also summarized in Table 4. Correlations between vegetative organs were mostly strong in February and April, whereas correlations between shoots and flowers and between roots and flowers were not significant. These results suggest that RCs concs. in the soil more directly affect root and shoot concentrations.

Radiocesium activity concentration in *B. napus* seed oil

Oil was extracted from *B. napus* seeds harvested from FLD2 and FLD3, and RCs concs. in seed oil and residue were determined (Table 5). Although the seeds from FLD2 and FLD3 contained 43 Bq·kg⁻¹ FW and 99 Bq·kg⁻¹ FW of total RCs concs., respectively, radiocesium was not detected in the oil. The detection limit for the measurement of RCs conc. in oil was <1

Bq·kg⁻¹. The recovery rate of radiocesium activity in the residue fractions was 85% and 83% in FLD2 and FLD3, respectively, and the remaining radiocesium activity was considered to be lost in the water fraction, which was removed during oil extraction. Few studies have previously described radiocesium transfer to the oil fraction of *B. napus* seeds; however, a review by Fesenko et al. (2007) and a report by Hirayama et al. (2011) covered this point. According to Hirayama et al. (2011), in plants containing radiocesium from fallout on leaves and absorption, RCs conc. in the oil fraction was very low, approximately 667 Bq·kg⁻¹ FW in seeds and 1.2 or 3.6 Bq·kg⁻¹ in oil. This report also mentions the need to measure radiocesium derived from root absorption without the influence of radioactive fallout on leaves, and this was performed in the present study. The result revealed that the RCs conc. in the oil fraction was similarly very low in the case of soil uptake alone.

Concluding remarks

Our results obtained 1.5 years after the nuclear accident revealed that the correlation between the exchangeable and fixed-radiocesium fractions was high, that water-soluble radiocesium was not detected, and that there was little penetration of radiocesium below L2. RCs concs. in soil showed a stronger correlation with those in roots and shoots compared to those in flowers. In fact, the distribution of radiocesium, such as that observed in FLD1, may not lead to severe contamination of Brassicaceae crops. The oil extraction experiment provided evidence that radiocesium did not transfer to the oil fraction. Our study using extensive cultivation in contaminated soils is useful to gain a better scientific understanding of radiocesium transport and accumulation and to provide useful information for the utilization of areas with low contamination levels because timely decontamination is difficult after radiocesium fallout over large areas.

Table 4. Correlation coefficients between RCs concs. in plant organs and soil layers.

	Shoot (Feb.)	Root (Feb.)	Shoot (Apr.)	Root (Apr.)	Flower (Apr.)
L1 ex ^z	0.902* ^y	0.943*	0.965**	0.978**	0.713
L1 tl	0.826	0.932*	0.975**	0.958*	0.792
L2 ex	0.648	0.573	0.418	0.359	0.229
L2 tl	0.649	0.598	0.446	0.375	0.274
Shoot (Feb.)		0.890*	0.839	0.905*	0.459
Root (Feb.)			0.984**	0.926*	0.801
Shoot (Apr.)				0.945*	0.846
Root (Apr.)					0.637

^z The abbreviations ex and tl indicate exchangeable and total radiocesium, respectively.

^y Correlation coefficients were calculated for RCs concs. (¹³⁷Cs) at five plots in FLD2 and FLD3, where radiocesium activity in the exchangeable fraction was detected in L1. L1, plow layer and L2, layer 5 cm below L1. Significant correlations are indicated with * ($P < 0.05$) and ** ($P < 0.01$).

Table 5. Summary of results from seed oil extraction experiments.

	FLD2 ^z	FLD3
Seed weight (kg FW)	135.2	135.3
Residue (kg FW)	72.9	74.8
Oil (kg)	61.6	58.2
Seeds		
¹³⁴ Cs (Bq·kg ⁻¹ FW)	18.7	26.4
¹³⁷ Cs	24.0	72.3
Oil		
¹³⁴ Cs (Bq·kg ⁻¹ FW)	<0.532 ^y	<0.664
¹³⁷ Cs	<0.507	<0.664
Residue		
¹³⁴ Cs (Bq·kg ⁻¹ FW)	28.3	53.9
¹³⁷ Cs	39.1	93.7

^z A single sample of seeds was used for extraction after harvesting from two test fields.

^y Radiocesium was not detected and the detection limits are indicated.

Literature Cited

- Albert, B., F. Le Cahérec, M. Niogret, P. Faes, J. Avice, L. Leport and A. Bouchereau. 2012. Nitrogen availability impacts oil-seed rape (*Brassica napus* L.) plant water status and proline production efficiency under water-limited conditions. *Planta* 236: 659–676.
- Aung, H. P., S. Djedidi, A. Z. Oo, Y. S. Aye, T. Yokoyama, S. Suzuki, H. Sekimoto and S. D. Bellingrath-Kimura. 2015. Growth and ^{137}Cs uptake of four *Brassica* species influenced by inoculation with a plant growth-promoting rhizobacterium *Bacillus pumilus* in three contaminated farmlands in Fukushima prefecture, Japan. *Sci. Total Environ.* 521–522: 261–269.
- Bengtsson, S. B., J. Eriksson, A. I. Gärdenäs, M. Vinichuk and K. Rosen. 2013. Accumulation of wet-deposited radiocaesium and radiostrontium by spring oilseed rape (*Brassica napus* L.) and spring wheat (*Triticum aestivum* L.). *Environ. Pollut.* 182: 335–342.
- Choi, Y.-H., K.-M. Lim, I. Jun, D.-K. Keum and C.-W. Lee. 2008. Effects of the simultaneous application of potassium and calcium on the soil-to-Chinese cabbage transfer of radiocesium and radiostrontium. *J. Environ. Radioact.* 99: 1853–1858.
- Chu, Q., T. Watanabe, Z. Sha, M. Osaki and T. Shinano. 2015. Interactions between Cs, Sr, and other nutrients and trace element accumulation in *Amaranthus* shoot in response to variety effect. *J. Agric. Food Chem.* 63: 2355–2363.
- Committee of Standard Methods for Soil Analysis (Japanese Society of Soil Science and Plant Nutrition). 1986. Standard methods for soil analysis (In Japanese). Hakuyusha Co., Ltd., Tokyo.
- Cooper, J. A. 1970. Factors determining the ultimate detection sensitivity of Ge (Li) gamma-ray spectrometers. *Nucl. Instrum. Meth.* 82: 273–277.
- Djedidi, S., K. Kojima, N. Ohkama-Ohtsu, S. D. Bellingrath-Kimura and T. Yokoyama. 2016. Growth and ^{137}Cs uptake and accumulation among 56 Japanese cultivars of *Brassica rapa*, *Brassica juncea* and *Brassica napus* grown in a contaminated field in Fukushima: Effect of inoculation with a *Bacillus pumilus* strain. *J. Environ. Radioact.* 157: 27–37.
- Endo, S., T. Kajimoto and K. Shizuma. 2013. Paddy-field contamination with ^{134}Cs and ^{137}Cs due to Fukushima Dai-ichi Nuclear Power Plant accident and soil-to-rice transfer coefficients. *J. Environ. Radioact.* 116: 59–64.
- Fesenko, S. V., R. M. Alexakhin, M. I. Balonov, I. M. Bogdevitch, B. J. Howard, V. A. Kashparov, N. I. Sanzharova, A. V. Panov, G. Voigt and Y. M. Zhuchenka. 2007. An extended critical review of twenty years of countermeasures used in agriculture after the Chernobyl accident. *Sci. Total Environ.* 383: 1–24.
- Fuhrmann, M., M. M. Lasat, S. D. Ebbs, L. V. Kochian and J. Cornish. 2002. Uptake of cesium-137 and strontium-90 from contaminated soil by three plant species; application to phytoremediation. *J. Environ. Qual.* 31: 904–909.
- Fujimura, S., K. Yoshioka, T. Saito, M. Sato, M. Sato, Y. Sakuma and Y. Muramatsu. 2013. Effects of applying potassium, zeolite and vermiculite on the radiocesium uptake by rice plants grown in paddy field soils collected from Fukushima prefecture. *Plant Prod. Sci.* 16: 166–170.
- Higaki, T., S. Higaki, M. Hirota and S. Hasezawa. 2014. Radiocesium distribution in bamboo shoots after the Fukushima nuclear accident. *PLoS ONE* 9: e97659.
- Hirayama, T., H. Sekizawa and M. Sato. 2011. Effect of radiocesium on *Brassica napus* and its transfer to oil (In Japanese) <<http://www.naro.affrc.go.jp/org/tarc/seika/jyouhou/H23/hatasaku/H23hatasaku006.html>>.
- Hirose, K. 2016. Fukushima Daiichi Nuclear Plant accident: Atmospheric and oceanic impacts over the five years. *J. Environ. Radioact.* 157: 113–130.
- Kaunisto, S., L. Aro and A. Rantavaara. 2002. Effect of fertilisation on the potassium and radiocaesium distribution in tree stands (*Pinus sylvestris* L.) and peat on a pine mire. *Environ. Pollut.* 117: 111–119.
- Kondo, M., T. Makino, T. Eguchi, A. Goto, H. Nakano, T. Takai, Y. Arai-Sanoh and T. Kimura. 2015. Comparative analysis of the relationship between Cs and K in soil and plant parts toward control of Cs accumulation in rice. *Soil Sci. Plant Nutr.* 61: 144–151.
- Kozhakhnov, T. E., S. N. Lukashenko and N. V. Larionova. 2014. Accumulation of artificial radionuclides in agricultural plants in the area used for surface nuclear tests. *J. Environ. Radioact.* 137: 217–226.
- Kusaba, S., K. Matsuoka, K. Abe, H. Ajito, M. Abe, N. Sakuma, Y. Saito, H. Shimura, N. Kihou and K. Hiraoka. 2016. Effect of soil surface management on radiocesium concentrations in apple orchard and fruit. *Hort. J.* 85: 30–36.
- Lasat, M. M., W. A. Norvell and L. V. Kochian. 1997. Potential for phytoextraction of Cs-137 from a contaminated soil. *Plant Soil* 195: 99–106.
- MAFF (Ministry of Agriculture, Forestry and Fisheries) and NARO (The National Agriculture and Food Research Organization). 2015. Factors causing high radiocesium concentration in soybean and its control (In Japanese) <http://www.maff.go.jp/j/kanbo/joho/saigai/pdf/youin_daizu_3.pdf>.
- Mori, S., A. Hirato, K. Tanoi, K. Takeda, T. Yamakawa and H. Nakanishi. 2012. Radioactive cesium flow in *Rhus vernicifera*. *Soil Sci. Plant Nutr.* 58: 611–617.
- Nisbet, A. F. and S. Shaw. 1994. Summary of a five-year lysimeter study on the time dependent transfer of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$ and ^{241}Am to crops from three contrasting soil types. 2 Distribution between different plant parts. *J. Environ. Radioact.* 23: 171–187.
- Nobori, T., N. I. Kobayashi, K. Tanoi and T. M. Nakanishi. 2014. Effects of potassium in reducing the radiocesium translocation to grain in rice. *Soil Sci. Plant Nutr.* 60: 772–781.
- Ohse, K., N. Kihou, K. Kurishima, T. Inoue and I. Taniyama. 2015. Changes in concentrations of I-131, Cs-134 and Cs-137 in leafy vegetables, soil and precipitation in Tsukuba city, Ibaraki, Japan, in the first 4 months after the Fukushima Daiichi nuclear power plant accident. *Soil Sci. Plant Nutr.* 61: 225–229.
- Paasikallio, A., A. Rantavaara and J. Sippola. 1994. The transfer of cesium-137 and strontium-90 from soil to food crops after the Chernobyl accident. *Sci. Total Environ.* 155: 109–124.
- Penrose, B., K. A. Johnson nee Payne, A. Arkhipov, A. Maksimenko, S. Gaschak, M. C. Meacham, N. J. M. Crout, P. J. White, N. A. Beresford and M. R. Broadley. 2016. Inter-cultivar variation in soil-to-plant transfer of radiocaesium and radiostrontium in *Brassica oleracea*. *J. Environ. Radioact.* 155–156: 112–121.
- Ramzaev, V., A. Barkovsky, Y. Goncharova, A. Gromov, M. Kaduka and I. Romanovich. 2013. Radiocesium fallout in the grasslands on Sakhalin, Kunashir and Shikotan Islands due to Fukushima accident: the radioactive contamination of soil and plants in 2011. *J. Environ. Radioact.* 118: 128–142.
- R Core Team. 2014. R: A language and environment for statistical computing. R Foundation for Statistical Computing, Vienna, Austria. <<http://www.R-project.org/>>.
- Sato, M., K. Abe, H. Kikunaga, D. Takata, K. Tanoi, T. Ohtsuki

- and Y. Muramatsu. 2015. Decontamination effects of bark washing with a high-pressure washer on peach [*Prunus persica* (L.) Batsch] and Japanese persimmon (*Diospyros kaki* Thunb.) contaminated with radiocaesium during dormancy. Hort. J. 84: 295–304.
- Shibuya, T., T. Watanabe, H. Ikeda and Y. Kanayama. 2015. Ionomeric analysis of horticultural plants reveals tissue-specific element accumulation. Hort. J. 84: 305–313.
- Terashima, I., M. Shiyomi and H. Fukuda. 2014. ^{134}Cs and ^{137}Cs levels in a grassland, 32 km northwest of the Fukushima 1 Nuclear Power Plant, measured for two seasons after the fallout. J. Plant Res. 127: 43–50.
- Yasutaka, T., H. Miyoshi and K. Ito. 2014. Transfer of radiocaesium from hydroponic medium to potherb mustard and tomato plants. Soil Sci. Plant Nutr. 60: 818–823.
- Yong, H.-Y., C. Wang, I. Bancroft, F. Li, X. Wu, H. Kitashiba and T. Nishio. 2015. Identification of a gene controlling variation in the salt tolerance of rapeseed (*Brassica napus* L.). Planta 242: 313–326.