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Radioactive contamination of arthropods from different trophic levels in hilly and mountainous areas after the Fukushima Daiichi nuclear power plant accident

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A B S T R A C T

In order to understand the influence of the Fukushima Daiichi nuclear power plant accident on the ecosystem in hilly and mountainous areas of Fukushima Prefecture, chronological changes in the levels of radiocesium in arthropod species were investigated. From 2012 to 2014, arthropods from different trophic levels were sampled and the air radiation dose rates at the sampling sites were analyzed. The air radiation dose rates showed a significant and constant reduction over the 2 years at the sampling sites in Fukushima. The median radiocesium concentration (^{134}Cs + ^{137}Cs) detected in the rice grasshopper, *Oxya yezoensis*, and the Emma field cricket, *Teleogryllus emma*, dropped continuously to 0.080 and 0.078 Bq/g fresh weight, respectively, in 2014. In contrast, no significant reduction in radioactive contamination was observed in the Jorô spider, *Nephila clavata*, in which the level remained at 0.204 Bq/g in 2014. A significant positive correlation between radiocesium concentration and the air radiation dose rate was observed in the rice grasshopper, the Emma field cricket and the Jorô spider. The highest correlation coefficient ($\rho = 0.946$) was measured in the grasshopper.

1. Introduction

Five years after the accident at the Fukushima Daiichi nuclear power plant (hereafter referred to as FDNPP), approximately one hundred thousand refugees remain unable to return home (Fukushima Prefectural Government, 2016), and many difficulties still exist for agricultural recovery in the disaster-affected areas. A major factor behind this uninhabitability is the environmental contamination by radionuclides, especially radioactive cesium as reported in several studies (e.g. Endo et al., 2013; Kamada et al., 2012; Tsuiki and Maeda, 2012; Yoshihara et al., 2013).

Although decontamination operations have continuously been underway (Ministry of the Environment, 2016), their effects are unstable. After decontamination, air radiation dose measurements are unstable even though surveys are conducted within a limited area and/or time span. This is likely due to Fukushima's ecological characteristics which include a large proportion of hilly and mountainous regions, estimated at 82% of the prefectural area (Fukushima Prefectural Government, 2010). In fields backed by wooded hills or in small valleys, runoff from uphill can recontaminate the lowlands; however, there are still merits to conducting measurements of the air radiation dose rate. These include the immediacy and convenience of estimating radioactive contamination levels in the agroecosystem compared to measurements of soil radionuclide deposition using a germanium semiconductor detector.

Hilly and mountainous areas, as well as other farmlands in Japan, are inhabited by a diverse range of organisms including aquatic and terrestrial animals, birds and plants, all of which are linked through their activities to the outside ecosystems. Arthropods in particular maintain relationships between multiple ecological guilds, such as terrestrial herbivores, omnivores, and carnivores, through the food chain. In such an ecosystem, radioactive contamination may differently influence the arthropod species depending on their trophic level. By periodically

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monitoring the amount of radionuclides in these arthropods, the overall dynamics of radionuclides within an ecosystem can be elucidated. If these data correlate with representative air radiation doses, such as the mean and median values within that area, arthropod species could be utilized as a novel environmental indicator of radioactive contamination.

A high level of concern also exists pertaining to environmental protection and the effects of radiation on wildlife. With regard to the influence of radioactive contamination in arthropods after the FDNPP accident, Møller et al. (2013) conducted field censuses of birds and arthropods and reported a significant negative correlation between animal population density and the air radiation dose rate. Hiyama et al. (2012) reported that the lycaenid butterfly, *Zizeeria maha*, that emerged in contaminated areas in 2011 exhibited morphological abnormalities, and that those abnormalities were inherited by the next generation, which exhibited a higher proportion of abnormalities than the first generation. Akimoto (2014) compared the morphology and viability of first instar nymphs in populations of gall-forming aphids from Fukushima and uncontaminated areas and found that the proportion of morphological abnormalities and mortality was significantly higher in Fukushima populations than in those from control areas. Assuming these abnormalities are due to the FDNPP accident, the exposure dose rate should be estimated to clarify the effects of radiation on these organisms. In order to do so, the concentration of whole-organism radioactivity is the most important factor to quantify, as this parameter estimates the internal exposure of an individual organism (ICRP, 2009).

Furthermore, radioactive contamination should be investigated in multiple arthropod species belonging to different trophic levels in the food web to determine the influence of the accident on the regional ecosystem. From the 1970's to the early 1980's, investigations into watershed contamination by effluents from a former reactor in the United States demonstrated a significant correlation between radiocesium concentrations in plants and arthropods, suggesting producer–herbivore–predator associations in terrestrial and aquatic food webs (Anderson et al., 1973; Brisbin et al., 1989). After the Chernobyl reactor accident in 1986, radiocesium was detected in earthworms (142 Bq/kg fresh weight) and in the Eurasian woodcock (730 Bq/kg fresh weight) that fed on earthworms in Norway (Kålås et al., 1994). However, few field studies have been conducted which analyze radioactive contamination in sympatric arthropod species with different food habits, excluding a few reports conducted after the FDNPP accident (e.g. Nakanishi et al., 2015).

The main objectives of this study are: (1) to understand how geographic and temporal factors influence the accumulation of radioactive nuclides in terrestrial arthropod species; (2) to clarify the correlation between the radionuclide concentration in arthropods with the median air radiation dose rate within the sampling area; and (3) to confirm the difference in radionuclide accumulation between arthropod species belonging to different trophic levels. In the present study, we investigated the chronological changes in radiocesium concentration in grasshoppers, field crickets and web-making spiders, which belong to different trophic levels, collected from hilly and mountainous areas of Fukushima from 2012 to 2014.

2. Materials and methods

2.1. Sampling sites

The locations of the sampling sites are illustrated in Fig. 1. Profiles of these sites are summarized in Table 1. Three sites, labeled A, B and C, are located in hilly and mountainous areas of Fukushima Prefecture, at an altitude of 450–490 m and 40–46 km northwest or north-northwest of the FDNPP. Site D, located in Kanagawa Prefecture about 270 km south-west of the FDNPP, was selected as the control site. In the administrative division which includes sites A and B, decontamination activities under the direct control of the Ministry of Environment began in September 2012, and by the end of September 2014 included 14.4% of the target farmland and forest areas (Ministry of the Environment, 2014a, 2014b). In the subdivision including site C, decontamination activities organized by the municipal office were carried out in paddy fields around the site beginning in November 2012 (Monma et al., 2015). Surveys were carried out from September to October in 2012–2014. In 2012 and 2013, surveys were conducted at all four sites, while the survey was conducted only at site A in 2014.

2.2. Air radiation dose rate

The air radiation dose rate at each site was measured with a NaI (Tl) scintillation survey meter (Hitachi-Aloka TCS-172B) at a height of 1 m above the ground surface. For each site, dose rates were measured at five points of approximately 20 m intervals in distance.

2.3. Arthropod samples

At each sampling site, arthropods were collected by sweep-net sampling and hand collection. Samples were preserved in 70% ethanol in the field and refrigerated until sorted. Each sample was identified and sorted by species. Prior to radioactive measurement, samples were weighed after air-drying for 10 min.

2.4. Radioactive measurements

The radioactivity in arthropod samples was measured by gamma spectrometry with a high-purity germanium semiconductor detector (GC-2020, Canberra Industries) and a multichannel analyzer (DSA1000, Canberra Industries) for 3600 to 10,800 s. The counting efficiency of the detector was determined by measuring a certified mixed radioactive standard gamma volume source set (MX-033-U8PP, Japan Radioisotope Association). For each sample, 10 to 50 individual arthropods were placed into plastic containers (U8 container: 47 mm in diameter, 60 mm in height). The radioactivity of each sample was obtained as Bq/g fresh weight and was corrected for radioactive decay to the sampling date of the arthropods. When the radioactive measurement was below the detection limit, the amount of radionuclides was regarded as equivalent to the detection limit.

2.5. Statistical analyses

For both the air radiation dose rate and the amount of radionuclides in the arthropod samples, the lower (Q_1) and higher (Q_3) quartiles and the interquartile range ($\mathrm{IQR} = Q_3 - Q_1$) were calculated. Data points lying outside the range from ($Q_1 - 1.5 \times \mathrm{IQR}$) to ($Q_3 + 1.5 \times \mathrm{IQR}$) were identified as outliers and excluded (Tukey, 1977). Differences in the values between sampling sites and years were analyzed by a Kruskal-Wallis test. Spearman's rank correlation was used to test the association between the air radiation dose rate and the amount of radioactive nuclides in the arthropod samples. Statistical analyses were performed using R version 2.15.3 (R Core Team, 2013).

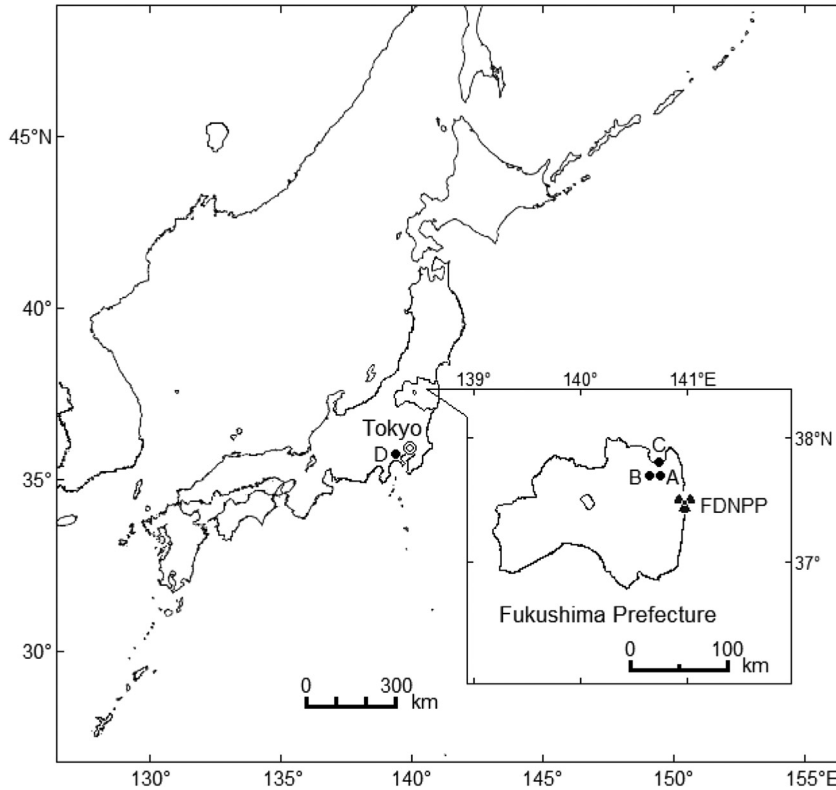

Fig. 1. The map shows the location of the sampling sites and the FDNPP. Sites A, B, and C are located in Fukushima Prefecture within 50 km northwest or north-northwest of the FDNPP. In contrast, site D is more than 250 km southwest of the FDNPP, located in the Tokyo metropolitan region (see Table 1).

Table 1
Distance from the FDNPP to the sampling sites and their geographic and evacuation status.

Site	Latitude, longitude and altitude	Distance and direction from FDNPP	Description[a]
A	37°41′38″ N 140°44′10″ E 446 m	40.1 km NW	A high school campus, currently closed, equipped with sports fields, shrub gardens and experimental farms. Located in the designated area (Area 2) in which residents are not permitted to live.
B	37°41′40 N 140°40′31 E 485 m	43.8 km NW	Fallow paddy fields between a stream and a mixed forest. Located in the designated area (Area 1) in which evacuation orders are ready to be lifted.
C	37°45′52 N 140°44′28 E 490 m	46.0 km NNW	Abandoned and fallow paddy fields near a canal and a broad-leaved forest.
D	35°26′18″ N 139°21′01″ E 26 m	267.2 km SW	Cultivated rice paddy fields just after harvesting, located in the suburbs of the Tokyo metropolitan region.

[a] Definitions of evacuation areas are from the Ministry of Economy, Trade and Industry (2012).

3. Results

3.1. Air radiation dose rate

Air radiation dose rates measured at a height of 1 m above the ground surface at the survey sites from 2012 to 2014 are summarized in Tables 2–4, respectively. In 2012, the highest air radiation dose rate was measured at site A (4.06 μSv/h). Measurements at site A, with a median of 3.74 μSv/h, were significantly higher than those at site D. In 2013, the highest dose rate was again measured at site A (2.83 μSv/h). Measurements at site A, with a median of 2.64 μSv/h, were significantly higher than those at site D. In 2014, the air radiation dose at site A ranged from 1.84 to 2.06 μSv/h with a median of 1.96 μSv/h. At site A, the median air radiation dose rate showed significant and consistent reductions from 3.74 to 1.96 μSv/h between 18 and 42 months (2012–2014) after the FDNPP accident ($H = 11.571$, $df = 2$, $P = 0.003$; Fig. 2).

3.2. Geographic variation in radioactive nuclide contamination in arthropods

The two arthropod species most commonly collected from the four sites were the rice grasshopper, *Oxya yezoensis* Shiraki (Orthoptera: Catantopidae) and the Emma field cricket, *Teleogryllus emma* (Ohmachi et Matsuura) (Orthoptera: Gryllidae). The Jorô spider, *Nephila clavata* L. Koch (Araneae: Nephilidae) was collected from sites A, B and D, but not collected from site C. The wasp spider, *Argiope bruennichi* (Scopoli) (Araneae: Araneidae) was collected from sites A and C.

The radioactive nuclides detected in the arthropod samples

Table 2
Air radiation dose rate and amount of radionuclides in arthropods at the sampling sites in 2012.

Site	Air radiation dose rate (μSv/h)		Amount of radionuclides (10^{-2} Bq/g fresh weight)															
			Rice grasshopper				Emma field cricket				Jorô spider				Wasp spider			
	n	Median (range)	No. of individuals/sample	Radio-nuclide	*n*	Median (range)	No. of individuals/sample	Radio-nuclide	*n*	Median (range)	No. of individuals/sample	Radio-nuclide	*n*	Median (range)	No. of individuals/sample	Radio-nuclide	*n*	Median (range)
A	5	3.74 (3.55–4.06) a	50	^{134}Cs	4	17.2 (14.8–19.9)	10	^{134}Cs	4	5.8 (5.0–6.4)	20	^{134}Cs	4	12.4 (10.2–14.0)	20	^{134}Cs	4	5.4 (4.8–6.4)[a]
				^{137}Cs	4	29.5 (23.6–31.7)		^{137}Cs	4 (1)	9.7 (9.2–16.2)		^{137}Cs	4 (1)	18.1 (16.9–19.4)		^{137}Cs	4	6.3 (5.3–6.6)
				Total	**4**	**46.9 (38.4–51.3) a**		**Total**	**4**	**15.6 (14.1–22.6)**		**Total**	**4 (1)**	**31.0 (28.3–31.1) a**		**Total**	**4 (1)**	**11.4 (10.9–11.5)[a] a**
B	5 (1)	1.95 (1.77–2.02) ab	50	^{134}Cs	4 (1)	7.6 (6.6–7.9)	10	^{134}Cs	2	7.5 (3.8–11.1)[a]	20	^{134}Cs	4 (1)	6.6 (6.4–7.2)	–	–	–	–
				^{137}Cs	4 (1)	10.8 (10.0–11.4)		^{137}Cs	2	12.7 (5.2–20.2)		^{137}Cs	4	14.6 (11.7–19.2)		–	–	–
				Total	**4 (1)**	**18.4 (16.6–19.3) ab**		**Total**	**2**	**20.2 (9.0–31.4)[a]**		**Total**	**4**	**21.4 (18.3–29.3) ab**		–	–	–
C	5 (1)	1.16 (1.14–1.20) ab	50	^{134}Cs	4	1.6 (1.5–1.6)[a]	10	^{134}Cs	4 (1)	3.8 (3.3–3.8)[a]	–	–	–	–	20	^{134}Cs	4	2.9 (2.8–3.0)[a]
				^{137}Cs	4	1.4 (1.2–1.5)[a]		^{137}Cs	4 (1)	4.0 (2.6–4.0)[a]		–	–	–		^{137}Cs	4	2.6 (2.1–3.3)[a]
				Total	**4**	**3.0 (2.8–3.2)[a] ab**		**Total**	**4 (1)**	**7.3 (6.3–7.8)[a]**		–	–	–		**Total**	**4**	**5.6 (4.9–6.1)[a] b**
D	5 (2)	0.04 (0.04–0.04) b	50	^{134}Cs	4	1.2 (0.9–1.3)[a]	10	^{134}Cs	4	3.5 (3.1–5.2)[a]	20	^{134}Cs	4	2.3 (2.1–2.9)[a]	–	–	–	–
				^{137}Cs	4	1.2 (1.0–1.3)[a]		^{137}Cs	4	4.5 (4.1–5.2)[a]		^{137}Cs	4 (1)	2.1 (1.9–2.1)[a]		–	–	–
				Total	**4 (1)**	**2.4 (2.4–2.6)[a] b**		**Total**	**4 (1)**	**7.8 (7.5–7.9)[a]**		**Total**	**4 (1)**	**4.3 (4.2–4.4)[a] b**		–	–	–
Kruskal–Wallis test	$H = 14.133$ $df = 3$ $P = 0.003$		$H = 12.200$ $df = 3$ $P = 0.007$				$H = 8.628$ $df = 3$ $P = 0.035$				$H = 7.318$ $df = 2$ $P = 0.026$				$H = 4.500$ $df = 1$ $P = 0.034$			

Numbers in parentheses after the number of analyzed samples (*n*) represent the number of outliers among the samples (see text). Values of air radiation dose rate and total radionuclide amounts followed by the same letter within the column were not significantly different ($p > 0.05$) in multiple comparisons by Scheffe's method following a Kruskal–Wallis test.
–: No arthropod sample was available at this site.
[a] Data include the detection limits since the measured amounts were below the limits (see text).

Table 3
Air radiation dose rate and amount of radionuclides in arthropods at the sampling sites in 2013.

Site	Air radiation dose rate (μSv/h)		Amount of radionuclides (10^{-2} Bq/g fresh weight)											
			Rice grasshopper				Emma field cricket				Jorô spider			
	n	Median (range)	No. of individuals/ sample	Radio-nuclide	*n*	Median (range)	No. of individuals/ sample	Radio-nuclide	*n*	Median (range)	No. of individuals/ sample	Radio-nuclide	*n*	Median (range)
A	5	2.64 (2.47–2.83) a	40	^{134}Cs	5	4.4 (3.9–4.6)	20	^{134}Cs	5	3.5 (3.0–4.8)	20	^{134}Cs	5 (1)	9.2 (4.6–10.4)[a]
				^{137}Cs	5	10.0 (8.3–11.2)		^{137}Cs	5	7.8 (6.1–10.1)		^{137}Cs	5	22.9 (7.4–35.1)
				Total	**5**	**13.9 (12.8–15.8) a**		**Total**	**5**	**11.3 (9.0–14.9) a**		**Total**	**5**	**33.3 (12.0–51.2)[a] a**
B	5 (1)	1.25 (1.24–1.36) ab	40	^{134}Cs	5	1.3 (0.9–1.6)	20	^{134}Cs	3	1.9 (1.3–2.5)	20	^{134}Cs	5	9.1 (6.0–11.9)
				^{137}Cs	**5**	2.8 (2.6–3.3)		^{137}Cs		3.7 (2.8–4.5)		^{137}Cs	5	21.4 (13.4–23.9)
				Total	**5**	**4.1 (3.5–4.9) ab**		**Total**	**3**	**6.1 (4.1–6.5) ab**		**Total**		**30.4 (19.4–35.1) ab**
C	5	0.81 (0.73–0.86) ab	40	^{134}Cs	**4**	1.0 (0.9–1.3)[a]	20	^{134}Cs	**5**	1.1 (1.0–3.2)	–	–	–	–
				^{137}Cs	4	1.6 (0.9–3.0)[a]		^{137}Cs	5	2.0 (1.3–6.3)		–	–	–
				Total	**4**	**2.5 (1.8–4.3)[a] ab**		**Total**	**5**	**3.1 (2.3–9.5)**		–	–	–
D	5	0.04 (0.03–0.05) b	40	^{134}Cs	5 (1)	0.8 (0.7–0.9)[a]	20	^{134}Cs	4	0.8 (0.8–0.9)[a]	20	^{134}Cs	4 (1)	1.3 (1.3–1.5)
				^{137}Cs	5	0.9 (0.7–1.0)[a]		^{137}Cs	4	0.8 (0.8–0.8)[a]		^{137}Cs	4	1.2 (0.8–1.4)[a]
				Total	**5**	**1.6 (1.4–1.8)[a] b**		**Total**	**4**	**1.6 (1.6–1.7)[a] b**		**Total**	**4**	**2.5 (1.7–2.9)[a] b**
Kruskal–Wallis test	$H = 16.939$ $df = 3$ $P < 0.001$		$H = 15.853$ $df = 3$ $P = 0.001$				$H = 13.294$ $df = 3$ $P = 0.004$				$H = 8.051$ $df = 2$ $P = 0.018$			

Numbers in parentheses after the number of analyzed samples (*n*) represent the number of outliers among the samples (see text). Values of air radiation dose rate and total radionuclide amounts followed by the same letter within the column are not significantly different ($p > 0.05$) in multiple comparisons by Scheffe's method following a Kruskal–Wallis test.
–: No arthropod sample was available at this site.
[a] Data include detection limits since the measured amounts were below the limits (see text).

Table 4
Air radiation dose rate and amount of radionuclides in arthropods at sampling site A in 2014.

Site	Air radiation dose rate (μSv/h)		Amount of radionuclides (10^{-2} Bq/g fresh weight)											
			Rice grasshopper				Emma field cricket				Jorô spider			
	n	Median (range)	No. of individuals/ sample	Radio-nuclide	*n*	Median (range)	No. of individuals/ sample	Radio-nuclide	*n*	Median (range)	No. of individuals/ sample	Radio-nuclide	*n*	Median (range)
A	5 (1)	1.96 (1.84–2.06)	20	^{134}Cs	4	2.0 (1.2–2.8)[a]	20	^{134}Cs	3	2.0 (1.6–4.8)	20	^{134}Cs	3	4.9 (3.4–5.6)[a]
				^{137}Cs	4	5.7 (4.7–8.1)		^{137}Cs	3	5.8 (3.9–6.3)		^{137}Cs	3	15.5 (10.3–16.7)
				Total	**4 (1)**	**8.0 (5.9–8.0)[a]**		**Total**	**3**	**7.8 (5.5–11.1)**		**Total**	**3**	**20.4 (13.7–22.2)[a]**

Numbers in parentheses after the number of analyzed samples (*n*) represent the number of outliers among the samples (see text).
[a] Data include detection limits since the measured amounts were below the limits (see text).

were ^{134}Cs and ^{137}Cs. Radioactive nuclides ^{40}K and ^{131}I were not detected in the samples throughout the investigation. The median concentration and range of ^{134}Cs and ^{137}Cs in the arthropod samples collected from 2012 to 2014 are summarized in Tables 2–4, respectively. In 2012, the total radiocesium (^{134}Cs + ^{137}Cs) contamination in the grasshopper, field cricket, and Jorô and wasp spiders was significantly different between the sampling sites (Kruskal–Wallis test, $P < 0.05$; Table 2). Among the arthropod species collected, the highest median concentration of radiocesium was detected in the grasshoppers (0.469 Bq/g, collected from site A), followed by Jorô spiders (0.310 Bq/g, from site A), field crickets (0.202 Bq/g, from site B), and wasp spiders (0.114 Bq/g, from site A). In 2013, the total levels of radiocesium contamination in the grasshopper, field cricket and Jorô spider were significantly different between sampling sites (Table 3). Among the arthropod species collected, the highest median concentration of radiocesium was detected in Jorô spiders (0.333 Bq/g, from site A), followed by grasshoppers (0.139 Bq/g, from site A) and field crickets (0.113 Bq/g, from site A). In 2014, of the arthropod species collected from site A, the highest median concentration of radiocesium was detected in Jorô spiders (0.204 Bq/g), followed by grasshoppers (0.080 Bq/g) and field crickets (0.078 Bq/g; Table 4).

3.3. Chronological changes in radiocesium contamination in arthropods

The change in radiocesium contamination over a three-year period was investigated in samples of three arthropod species

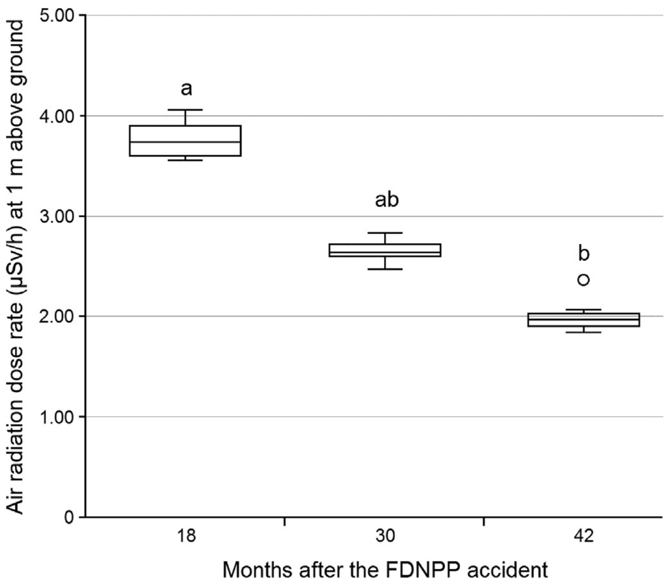

Fig. 2. Air radiation dose rate (microsieverts per hour at 1 m above ground) at site A, significantly differed with time after the FDNPP accident (Kruskal–Wallis test, $H = 11.571$, $df = 2$, $P = 0.003$). Minimum and maximum dose rates are depicted by whiskers. The box signifies the upper and lower quartiles, and the median is represented by a horizontal line within the box for each year. The outlier, which is a value more than 1.5 times the interquartile range away from the top or bottom of the box, is represented by a circle. Different letters above the box plots denote significant differences in multiple comparisons by Scheffe's method ($P < 0.05$).

collected from site A. In grasshoppers, the median concentration of ^{134}Cs and ^{137}Cs was significantly reduced from 0.469 to 0.080 Bq/g between 18 and 42 months after the FDNPP accident ($H = 9.692$, $df = 2$, $P = 0.008$; Fig. 3A). Field crickets also showed a significant reduction in radiocesium contamination, from 0.156 to 0.078 Bq/g ($H = 7.256$, $df = 2$, $P = 0.027$; Fig. 3B). In contrast, the median amount of radiocesium in Jorô spiders was estimated to be 0.310, 0.333 and 0.204 Bq/kg at 18, 30 and 42 months after the accident; these amounts were not significantly different between survey times ($H = 3.382$, $df = 2$, $P = 0.184$; Fig. 3C). The decreasing rate of median radiocesium concentration from 18 to 42 months after the accident was calculated to be 82.9%, 50.0% and 34.1% for grasshoppers, crickets and Jorô spiders, respectively.

3.4. Correlation between the air radiation dose and radiocesium contamination in arthropods

The strength of the association was tested between the concentration of radiocesium in samples of three arthropod species and the air radiation dose rate 1 m above the ground at the sample collection sites from 2012 to 2014 (Fig. 4). The wasp spider was excluded from the test since it was collected only once from each of two sites in a year. A significant positive correlation ($P < 0.05$) in the radiocesium concentration to the air radiation dose rate was observed in the rice grasshopper, the Emma field cricket and the Jorô spider. The highest correlation coefficient was measured in the grasshopper ($\rho = 0.946$).

4. Discussion

Results show that the air radiation doses significantly differed between the four sampling sites located in Fukushima and Kanagawa Prefectures in 2012 and 2013. Within Fukushima Prefecture, the order of the air radiation dose rates at the different sites reflects their distance from the FDNPP. This is consistent with the distribution of the air radiation dose rates reported after the accident by the Ministry of Education, Culture, Sports, Science and Technology (2012, 2013), which showed a northwesterly expansion from the FDNPP. At site A, where the survey was conducted continuously, the total radiocesium concentration in the grasshopper and field cricket decreased significantly over the two years from 2012 to 2014 along with the air radiation dose.

The contribution of ^{134}Cs to the total radiation is relatively high (Eckerman and Ryman, 1993), because the physical half-life of ^{134}Cs (2.06 y) is much shorter than that of ^{137}Cs (30.17y). In the present study, the decreasing rate of ^{134}Cs between 2012 and 2014 was higher than that of ^{137}Cs in the arthropod species ($^{134}Cs/^{137}Cs = 88.4\%/80.7\%$ in the grasshopper; 65.5%/40.2% in the field cricket; and 60.5%/14.4% in the Jorô spider). This could explain the constant reduction in air radiation dose rates and the total radiocesium concentration in arthropods that were observed in this survey. This suggests that the reduction in air radiation dose rates is slowing, as the contribution of ^{134}Cs is close to a negligible level.

The effects of weather exposure on the annual decrease in environmental air radiation dose are also important. Several studies have demonstrated that rainfall and typhoons promoted the dispersal of radiocesium into rivers (Matsunaga et al., 2013; Nagao et al., 2013). The additional influence of decontamination activities should also be considered.

The multi-year investigation of arthropods from different trophic levels demonstrated significant differences in the level of radioactive contamination. In contrast to the grasshopper and field cricket, in which the median amount of radiocesium showed a constant reduction from 2012 to 2014, no significant reduction in radioactive contamination was observed in the Jorô spider. This is likely due to the different food resource pathways at the respective trophic levels, such as grazing and detrital food chains (Polis and Strong, 1996). Murakami et al. (2014) reported high ^{137}Cs concentrations in detritivore groups and suggested that the ^{137}Cs transferred between organisms flows up to higher trophic levels through the detritus food chain. Grasshoppers rely on the grazing food chain, feeding exclusively on green plants, while field crickets and spiders make use of the detritus food chains (ElEla et al., 2010; Shimazaki and Miyashita, 2005). Different patterns of chronological change in radiocesium concentrations might be attributable to differences in the feeding habits of the arthropods.

Ayabe et al. (2015a) reported that the mean level of radiocesium contamination in the Jorô spider decreased from 2012 to 2013, but highly contaminated spiders (>9.0 Bq/g dry weight) were still collected in 2013. Web-making spiders such as the Jorô spider, rely on prey such as saprophagous flies for their food more than herbivores, increasing the likelihood of transferring radionuclides through the detritus food chain (Rudge et al., 1993; Wood et al., 2009). This could result in a wide distribution of contamination levels between samples. Recently, Ayabe et al. (2015b) demonstrated that the accumulation of alkali metals in the Jorô spider is dependent on the type of prey it consumes, and this predation is also a factor in determining the level of ^{137}Cs contamination. The authors suggest that variation in ^{137}Cs contamination among herbivores caused by their diverse feeding habits would be passed on through the food chain to spiders at higher trophic levels.

Although no morphological abnormalities were observed in the individual arthropod samples collected in the present study, data from this kind of long-term investigation will be necessary for estimating the extent to which these organisms are influenced by internal and external exposure to the fallout from the FDNPP accident. This radioactive fallout is a potential contributing factor to morphological abnormalities in wildlife. The biological half-life of cesium in an arthropod body has been reported to be relatively short: 4–5 days for ^{137}Cs in the eastern lubber grasshopper,

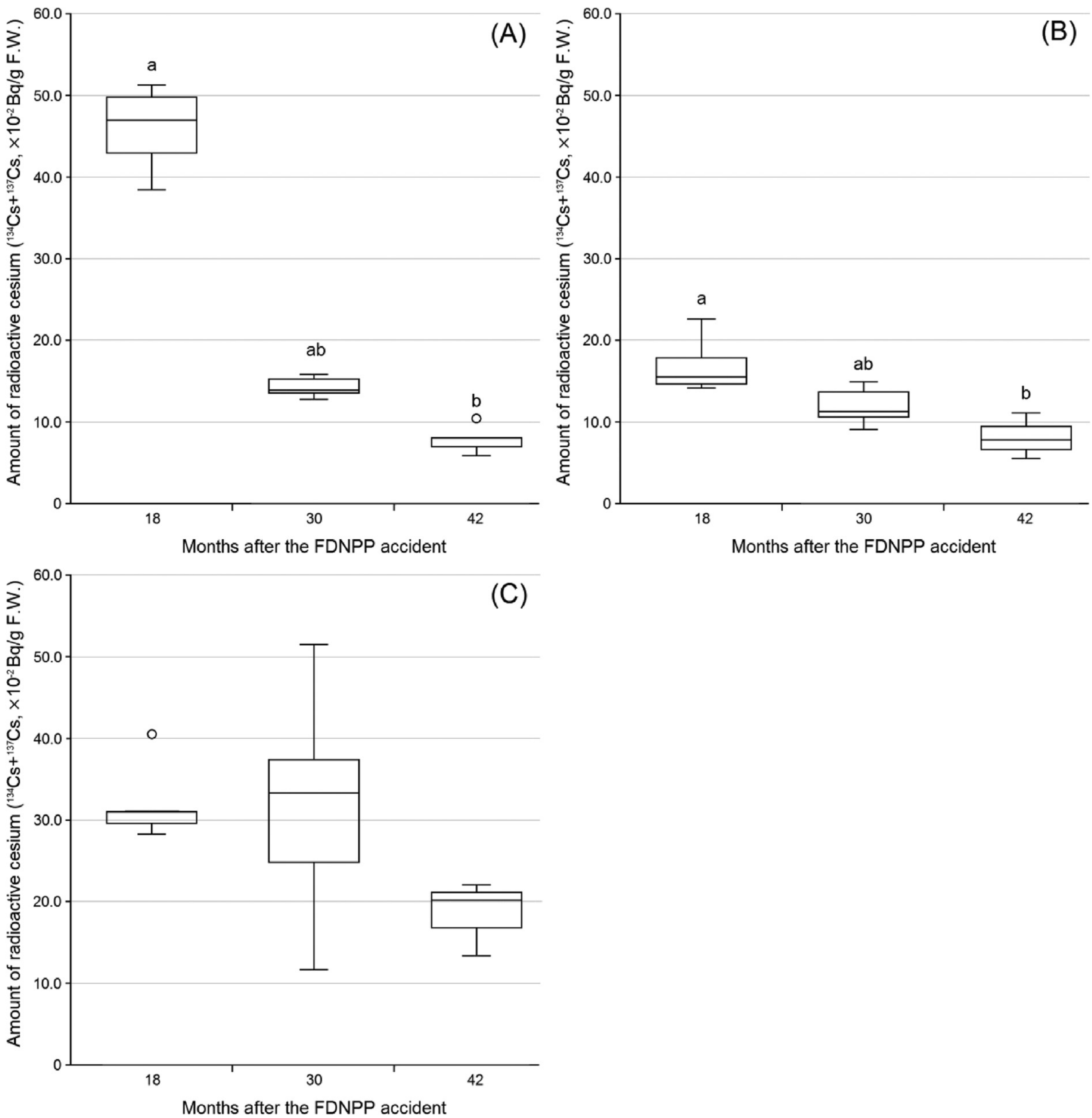

Fig. 3. Radiocesium contamination (becquerels per gram of fresh weight) in the rice grasshopper, *Oxya yezoensis* (A) and Emma field cricket, *Teleogryllus emma* (B) at site A significantly changed over time after the FDNPP accident (A: $H = 9.692$, $df = 2$, $P = 0.008$; B: $H = 7.256$, $df = 2$, $P = 0.027$ using the Kruskal–Wallis test). However, no significant change was observed in the Jorô spider, *Nephila clavata* (C; $H = 3.382$, $df = 2$, $P = 0.184$). Minimum and maximum concentrations are depicted by whiskers. The box signifies the upper and lower quartiles, and the median is represented by a horizontal line within the box for each year. The outlier, which is a value more than 1.5 times the interquartile range away from the top or bottom of the box, is represented by a circle. Different letters above the box plots denote significant differences in multiple comparisons by Scheffe's method ($P < 0.05$).

Romalea microptera (Crossley and Pryor, 1960); 4.9 h–3.9 days for ^{134}Cs in the brown cricket, *Acheta domesticus* (DiGregorio et al., 1978; Van Hook and Crossley, 1969); and 11.5–51.0 days for ^{134}Cs in various species of spiders (Lycosidae, Gnaphosidae and Thomisidae) (Moulder and Dodson, 1967). In the present study, all of the sampled arthropod species are univoltine, having one generation per year (Chikuni, 2008; Murai and Ito, 2011), which indicates that arthropods in the 4th generation after the accident are continuing to ingest radiocesium through the food chain in the local ecosystem in spite of the decontamination activities.

The present study was the first to conduct a multi-year survey of radioactive contamination levels in arthropod species from different trophic levels after the FDNPP accident. Results identified a trend in which radioactive contamination in the Jorô spider was higher than that in the herbivorous grasshopper or omnivorous field cricket, except for 2012 when the contamination level in the grasshopper was higher than that in the field cricket or the two species of spiders.

No radionuclides other than ^{134}Cs and ^{137}Cs were detected in the arthropod samples throughout the present study. It is possible that the detection time adopted in this study (3600–10,800 s) could be too short for detection of radionuclides such as ^{110m}Ag. Nakanishi et al. (2015) reported that ^{110m}Ag was detected in Jorô spiders collected from Fukushima Prefecture six months after the FDNPP accident.

Significant positive correlations were found between the

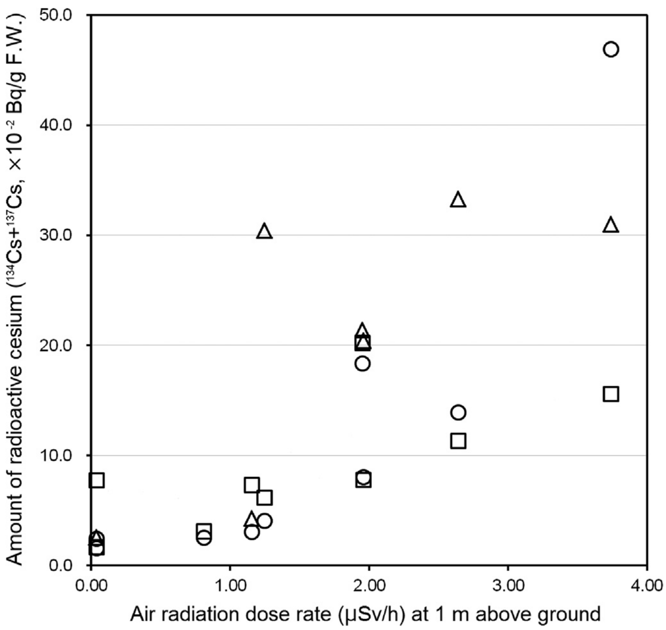

Fig. 4. The amount of radiocesium detected in arthropod samples is significantly correlated with the air radiation dose rate at the sites where the samples were collected from 2012 to 2014. Circles, squares and triangles represent the rice grasshopper, the Emma field cricket and the Jorô spider, respectively. Spearman's rank correlation coefficient (ρ) and P values for the arthropod species are as follows: rice grasshopper: $\rho = 0.946$, $P < 0.001$; Emma field cricket: $\rho = 0.753$, $P = 0.019$; Jorô spider: $\rho = 0.821$, $P = 0.034$.

radiocesium concentrations in the arthropod species and the air radiation dose rates at the sites where the samples were collected from 2012 to 2014. Ayabe et al. (2014) demonstrated a correlation between the radiocesium contamination in Jorô spiders and air radiation dose rates in Fukushima Prefecture during a single year (2012). Cumulative data from these types of investigations over time will be useful for predicting the reduction in radioactive contamination within the affected area.

5. Conclusion

The present study suggests that arthropods are an important factor in the circulation of radiocesium in the ecosystem from a long-term perspective. We demonstrated significant differences in chronological change in radiocesium concentrations in arthropod populations from different trophic levels. These differences are likely caused by different food chain pathways, suggesting behavioral correlates to radiocesium levels in the ecosystem. To understand the contamination dynamics of radiocesium in hilly and mountainous areas in Fukushima where restoration is delayed, it is essential to conduct long-term monitoring surveys of arthropods commonly distributed in both the farmlands and forested areas, and which belong to different trophic levels in the food web. These investigations will also lead to an accurate assessment of the impact of radiation on non-human biota for the purposes of environmental radiation protection. The present study provides basic information on radioactive contamination and trophic level flow in arthropods after the FDNPP accident. The level of radiation exposure in wildlife compared to the radioactive contamination caused by the nuclear accident, regardless of the air radiation dose, greatly varies depending on habitat and feeding habits of the individual species. Therefore, further investigation is needed to clarify the impact of low dose radiation on non-human biota as well as the basis for the different patterns of chronological change in radiocesium contamination within trophic levels.

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References

Akimoto, S., 2014. Morphological abnormalities in gall-forming aphids in a radiation-contaminated area near Fukushima Daiichi: selective impact of fallout? Ecol. Evol. 4, 355–369.

Anderson, G.E., Gentry, J.B., Smith, M.H., 1973. Relationships between levels of radiocesium in dominant plants and arthropods in a contaminated streambed community. Oikos 24, 165–170.

Ayabe, Y., Kanasashi, T., Hijii, N., Takenaka, C., 2014. Radiocesium contamination of the web spider *Nephila clavata* (Nephilidae: arachnida) 1.5 years after the Fukushima Dai-ichi nuclear power plant accident. J. Environ. Radioact. 127, 105–110.

Ayabe, Y., Kanasashi, T., Hijii, N., Takenaka, C., 2015a. Monitoring radiocesium contamination of the web spider *Nephila clavata* (Nephilidae: arachnida) in Fukushima forests. J. Jpn. For. Soc. 97, 70–74.

Ayabe, Y., Kanasashi, T., Hijii, N., Takenaka, C., 2015b. Relationship between radiocesium contamination and the contents of various elements in the web spider *Nephila clavata* (Nephilidae: arachnida). J. Environ. Radioact. 150, 228–235.

Brisbin Jr., I.L., Breshears, D.D., Brown, K.L., Ladd, M., Smith, M.H., Smith, M.W., Towns, A.L., 1989. Relationships between levels of radiocaesium in components of terrestrial and aquatic food webs of a contaminated streambed and floodplain community. J. Appl. Ecol. 26, 173–182.

Chikuni, Y., 2008. Pictorial Encyclopedia of Spiders in Japan, Revised Ver. Kaisei-sha, Tokyo.

Crossley Jr., D.A., Pryor, M.E., 1960. The uptake and elimination of cesium-137 by a grasshopper—*Romalea microptera*. Health Phys. 4, 16–20.

DiGregorio, D., Kitchings, T., Van Voris, P., 1978. Radionuclide transfer in terrestrial animals. Health Phys. 34, 3–31.

Eckerman, K.F., Ryman, J.C., 1993. External Exposure to Radionuclides in Air, Water, and Soil, Federal Guidance Report No. 12, EPA-402-R-93–081. Office of Radiation and Indoor Air, U.S. Environmental Protection Agency, Washington, DC.

ElEla, S.A., ElSayed, W., Nakamura, K., 2010. Mandibular structure, gut contents analysis and feeding group of orthopteran species collected from different habitats of Satoyama area within Kanazawa City. Jpn. J. Threat. Taxa 2, 849–857.

Endo, S., Kajimoto, T., Shizuma, K., 2013. Paddy-field contamination with ^{134}Cs and ^{137}Cs due to Fukushima Dai-ichi Nuclear Power Plant accident and soil-to-rice transfer coefficient. J. Environ. Radioact. 116, 59–64.

Fukushima Prefectural Government, 2010. The Strategy for the Promotion of Depopulated Hilly and Mountainous Area in Fukushima Prefecture. Fukushima Prefectural Government, Fukushima.

Fukushima Prefectural Government, 2016. Transition of Evacuation Instruction Zones: Evacuation Zone Map Update (accessed 16.06.27). http://www.pref.fukushima.lg.jp/site/portal-english/en03-08.html.

Hiyama, A., Nohara, C., Kinjo, S., Taira, W., Gima, S., Tanahara, A., Otaki, J.M., 2012. The biological impacts of the Fukushima nuclear accident on the pale grass blue butterfly. Sci. Rep. 2, 570.

ICRP, 2009. Environmental protection: transfer parameters for reference animals and plants (ICRP Publication 114). Ann. ICRP 39, 1–111.

Kålås, J.A., Bretten, S., Byrkjedal, I., Njåstad, O., 1994. Radiocesium (^{137}Cs) from the Chernobyl reactor in Eurasian woodcock and earthworms in Norway. J. Wildl. Manage 58, 141–147.

Kamada, N., Saito, O., Endo, S., Kimura, A., Shizuma, K., 2012. Radiation doses among residents living 37 km northwest of the Fukushima Dai-ichi nuclear power plant. J. Environ. Radioact. 110, 84–89.

Matsunaga, T., Koarashi, J., Atarashi-Andoh, M., Nagao, S., Sato, T., Nagai, H., 2013. Comparison of the vertical distributions of Fukushima nuclear accident radiocesium in soil before and after the first rainy season, with physicochemical and mineralogical interpretations. Sci. Tot. Environ. 447, 301–314.

Ministry of Economy, Trade and Industry, 2012. Restricted Areas and Areas to which Evacuation Orders have been issued (accessed 16.04.09). http://www.meti.go.jp/english/earthquake/nuclear/roadmap/pdf/20121130_01.pdf.

Ministry of Education, Culture, Sports, Science and Technology, 2012. Distribution

Map of Radiation Dose: Airborne Monitoring (accessed 16.04.09). http://ramap.jmc.or.jp/map/engpdf/pdf/air/20120628/dr/5640-C.pdf.

Ministry of Education, Culture, Sports, Science and Technology, 2013. Distribution Map of Radiation Dose: Airborne Monitoring (accessed 16.04.09). http://ramap.jmc.or.jp/map/engpdf/pdf/air/20130928/dr/5640-C.pdf.

Ministry of the Environment, 2014a. Progress of Decontamination in the Special Target Areas, Oct. 17, 2014a (accessed 16.06.23). http://josen-plaza.env.go.jp/info/weekly/pdf/weekly_141017a.pdf.

Ministry of the Environment, 2014b. Progress of Decontamination Based on the Special Target Area Project, Oct. 17, 2014b (accessed 16.06.23). http://josen-plaza.env.go.jp/info/weekly/pdf/weekly_141017c.pdf.

Ministry of the Environment, 2016. Off-site Decontamination Measures (accessed 16.05.30). http://josen.env.go.jp/en/.

Møller, A.P., Nishiumi, I., Suzuki, H., Ueda, K., Mousseau, T.A., 2013. Differences in effects of radiation on abundance of animals in Fukushima and Chernobyl. Ecol. Indic. 24, 75–81.

Monma, T., Lurhathaiopath, P., Kawano, Y., Byambasuren, D., Ono, Y., Evine, Q., 2015. Developing and trialing a system to monitor radionuclides in individual plots of farmland to help reconstruction farming in contaminated areas. In: Monma, T., Goto, I., Hayashi, T., Tachiya, H., Ohsawa, K. (Eds.), Agricultural and Forestry Reconstruction after the Great East Japan Earthquake: Tsunami, Radioactive, and Reputational Damages. Springer, Japan, Tokyo, pp. 149–167.

Moulder, B.C., Dodson, G.J., 1967. Elimination rates of radiocesium by spiders as a function of temperature and body size. In: Morgan, K.Z., Snyder, W.S., Struxness, E.G. (Eds.), Health Physics Division Annual Progress Report: for Period Ending July 31, 1967. Oak Ridge National Laboratory, Oak Ridge, pp. 86–88.

Murakami, M., Ohte, N., Suzuki, T., Ishii, N., Igarashi, Y., Tanoi, K., 2014. Biological proliferation of cesium-137 through the detrital food chain in a forest ecosystem in Japan. Sci. Rep. 4, 3599.

Murai, T., Ito, F., 2011. A Field Guide to the Orthoptera of Japan. Hokkaido University Press, Sapporo.

Nagao, S., Kanamori, M., Ochiai, S., Tomihara, S., Fukushi, K., Yamamoto, M., 2013. Export of ^{134}Cs and ^{137}Cs in the Fukushima river systems at heavy rains by Typhoon Roke in September 2011. Biogeosciences 10, 6215–6223.

Nakanishi, H., Mori, A., Takeda, K., Tanaka, H., Kobayashi, N., Tanoi, K., Yamakawa, T., Mori, S., 2015. Discovery of radioactive silver (^{110m}Ag) in spiders and other fauna in the terrestrial environment after the meltdown of Fukushima Dai-ichi nuclear power plant. Proc. Jpn. Acad. Ser. B 91, 160–174.

Polis, G.A., Strong, D.R., 1996. Food web complexity and community dynamics. Am. Nat. 147, 813–846.

R Core Team, 2013. R: a Language and Environment for Statistical Computing. ISBN 3-900051-07-0. R Foundation for Statistical Computing, Vienna (accessed 16.04.10). http://www.R-project.org.

Rudge, S.A., Johnson, M.S., Leah, R.T., Jones, S.R., 1993. Biological transport of radiocaesium in a semi-natural grassland ecosystem. 1. Soils, vegetation and invertebrates. J. Environ. Radioact. 19, 173–198.

Shimazaki, A., Miyashita, T., 2005. Variable dependence on detrital and grazing food webs by generalist predators: aerial insects and web spiders. Ecography 28, 485–494.

Tsuiki, M., Maeda, T., 2012. Spatial distribution of radioactive cesium fallout on grasslands from the Fukushima Daiichi nuclear power plant in 2011. Grassl. Sci. 58, 153–160.

Tukey, J.W., 1977. Exploratory Data Analysis, first ed. Addison-Wesley, Reading.

Van Hook Jr., R.I., Crossley Jr., D.A., 1969. Assimilation and biological turnover of cesium-134, iodine-131, and chromium-51 in brown crickets, *Acheta domesticus* (L.). Health Phys. 16, 463–467.

Wood, M.D., Leah, R.T., Jones, S.R., Copplestone, D., 2009. Radionuclide transfer to invertebrates and small mammals in a coastal sand dune ecosystem. Sci. Total Environ. 407, 4062–4074.

Yoshihara, T., Matsumura, H., Hashida, S., Nagaoka, T., 2013. Radiocesium contaminations of 20 wood species and the corresponding gamma-ray dose rates around the canopies at 5 months after the Fukushima nuclear power plant accident. J. Environ. Radioact 115, 60–68.