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

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Abstract: Six months after the explosion of TEPCO's Fukushima Dai-ichi nuclear power plant, radioactive silver ($^{110m}$Ag), was detected in concentrations of 3754 Bq/kg in Nephila clavata (the orb-web spider; Joro-gumo in Japanese) collected at Nimaibashi, Iitate village in Fukushima Prefecture, whereas $^{110m}$Ag in the soil was 43.1 Bq/kg. A survey of 35 faunal species in the terrestrial environment during the 3.5 years after the accident showed that most of Anthropoda had two orders higher $^{110m}$Ag in their tissues than soils, although silver is not an essential element for their life. However, tracing of the activity of $^{110m}$Ag detected in spider Atypus karschi collected regularly at a fixed location showed that it declined much faster than the physical half-life. These results suggest that $^{110m}$Ag was at once biologically concentrated by faunal species, especially Arthropoda, through food chain. The factors affecting the subsequent rapid decline of $^{110m}$Ag concentration in faunal species are discussed.

Keywords: Atypus karschi, fauna, Fukushima Dai-ichi accident, radioactive silver, radioactive cesium, Nephila clavata

Introduction

After the Great East Japan Earthquake and resulting Tsunami on March 11, 2011, a sequential meltdown of the nuclear fuels occurred at the Fukushima Dai-ichi nuclear power plant (NPP) of Tokyo Electric Power Company (TEPCO), following the discharge of enormous amounts of radioactive materials into the environment. The main components were $^{131}$I, $^{132}$I, $^{134}$Cs and $^{137}$Cs, whose total amounts were estimated to be $1.5 \times 10^{17}$ Bq for $^{131}$I, $1.2 \times 10^{16}$ Bq for $^{137}$Cs, and also $6.3 \times 10^{17}$ Bq in iodine equivalent.¹ Roughly half of the discharged activity had precipitated in soils and entered inland waters, where it was incorporated into flora and fauna in the terrestrial environment.² Among a number of attempts to reveal the scale and distribution of the fallout, our group also started to survey local and biological contamination with the radioactivity in the vicinity of the TEPCO's NPP. On Sept. 20, 2011, we visited one of the heavily contaminated area, Iitate village in Fukushima Prefecture, where had already been declared to be the "planned evacuation zone". It was a stormy day with heavy rain of typhoon and we had to abandon our original plan to collect vegetation samples, instead we collected the orb-web spider Nephila clavata (Joro-gumo in Japanese), which nested in abundance in a bamboo forest. Our initial conjecture was that the spiders might be contaminated with radioactive cesium reflecting contamination of the food chain. Unexpectedly, however, germanium-semiconductor analysis in the laboratory of the N. clavata specimens showed a sharp peak (657.8 keV) of $^{110m}$Ag distinct from the $^{137}$Cs peak (661.7 keV). This was the beginning to recognize marked biological concentration of $^{110m}$Ag in the field fauna during this accident, which was publicized preliminary.³

In fact, radioactive silver, $^{110m}$Ag (half-life: 249.95 days), first attracted attention as the atomic
weapon detritus, whose distinct biological accumulation was detected in the digestive tissues of a marine crustacean and mussels, barnacles, and fishes from the Pacific Ocean due to the repeated test explosions at Entwetok Atoll, Bikini Atoll etc., from 1946 to 1963.\(^4,\)\(^5\) \(^{110m}\text{Ag} \) had also been detected as a minor radioactive nuclide contained in wastewater from NPPs, however, the Chernobyl accident on April 26, 1986 caused its discharge in a large amount into the environment. For example, in Cumbria, northwestern England located \(\sim 2000\) km from Chernobyl, all vegetation in 18 varietal soils sampled in May 1986 was contaminated with \(^{110m}\text{Ag} \) with concentrations ranging from 190 to 480 Bq/kg of dryweight (DW).\(^6\) Many studies were conducted on the metabolism of \(^{110m}\text{Ag} \) in the fallout of the accident by aquatic biota especially in oceans.\(^7\)–\(^9\)

Studies on cattle such as oxen, lambs and sheep revealed that distinct accumulation of \(^{110m}\text{Ag} \) was rather restricted to liver with the biological half-life of 35 days in the case of sheep.\(^6\),\(^10\)–\(^12\) In contrast to these studies, contamination of terrestrial wild fauna involving small insects and animals remained very scarce.

Even in the accident at TEPCO’s NPP, studies on the contamination of fauna were mostly restricted to cattle. After the accident, \(^{110m}\text{Ag} \) was detected in the liver of cattle at a concentration of 177 \pm 176 Bq/kg,\(^13\) and a 3-month supply of clean hay as fodder to the contaminated cattle failed to decrease its contents in liver significantly.\(^14\) In contrast, no published studies exist on the dynamics of \(^{110m}\text{Ag} \) in small faunal species comprising the food chain in terrestrial environments. Therefore, we conducted successive surveillance of \(^{110m}\text{Ag} \) in spiders as well as several small animals and insects during our visits to Fukushima once a month until November 2014. The obtained results confirmed the reproducible accumulation of \(^{110m}\text{Ag} \) in small faunal species. Here, we describe the contamination of 35 of these species by \(^{110m}\text{Ag} \) and \(^{137}\text{Cs} \) (or \(^{134}\text{Cs} \) ) fallout and the fates of those radionuclides in the bodies of the orb-web spider \(N. \text{clavata} \) and the ground spider \(A. \text{karschi} \) in the 3.5 years after the NPP accident.

Materials and methods

**Sampling of fauna species and soils.** From Sept. 20, 2011 until Nov. 3, 2014, we collected 35 faunal species from Benten-yama and Hanami-yama in Fukushima city, Oguni, Miyuki-yama and Ryozen in Date city, Tsushima and Akungi in Namie town, Yamakiya in Kawamata town and Nimaibashi, Ittoi, Hiso, Maeda, Sasu, Usushi and Nagadoro in Iitate village in Fukushima Prefecture (Fig. 1). The sampling sites were located more than 30 km from Fukushima Dai-ichi NPP. Most of the collected species were separately stored in plastic 100-mL cups to avoid their preying on each other. Names of all collected species are listed in Table 3. The collected fauna were starved at room temperature for \(\sim 3\) weeks in the U8-cups, which were stored in desiccators. During the starvation process, some of the fauna excreted feces and urine before their death. Starvation was necessary to allow measurement of the true radioactivity in the body tissues of the collected fauna, after they had excreted residual, non-metabolized contamination in their feces and urine. For some of the fauna, the radioactivity in the desiccated feces was also measured.

Our visit to the Hiso on Oct. 13, 2013 coincided with the pregnancy phase of \(N. \text{clavata} \) females. We collected 85 of the pregnant spiders, separately storing them in U8-cups. These spiders were starved under dark conditions until some of them had spun webs and shed their egg sacs into the cups. Spider bodies, excrement (feces + urine), web thread, and egg sacs were separately collected and their radioactivity was measured. We also collected soil from the sites where \(N. \text{clavata} \) and \(A. \text{karschi} \) had been sampled. Surface soils were collected from an area of \(10 \times 10 \times 2\) cm. A vertical depth of 2 cm was chosen because \(>95\% \) of nuclear fallout accumulates within a vertical depth of 2 cm from the soil surface.\(^15\),\(^16\)

**Measurement of radioactivity.** The detection of the discharged \(^{110m}\text{Ag} \) in the atmospheric dust from the TEPCO’s NPP was first recognized by several institutions from March to May in 2011,\(^17\)–\(^19\) and its total amount was estimated to be \(4.04–6.41 \times 10^{15}\) Bq in the cores of Units 1–3 of the NPP.\(^20\) The Ministry of Education, Culture, Sports, Science and Technology, Japan, published a rough distribution map of \(^{110m}\text{Ag} \) in soils on Oct. 31, 2011 (Fig. 1),\(^21\) and its distribution in surface soils in a wide area around the power plant\(^22\)–\(^24\) as well as the vertical distribution profile in Fukushima soils\(^16\) were described a year later the accident.

The aerial dose (\(\mu\text{Sv/h} \) ) was measured in the contaminated fields at a height of 1 m from the sampling sites using an Aloka \(\gamma\)-survey meter (TCS-172; Hitachi-Aloka, Tokyo, Japan). In the laboratory, all materials were analyzed on germanium-semiconductor detectors (GEM type and GMX type, Seiko EG&G, Tokyo, Japan). The counting time was set at 220,000 s. \(^{110m}\text{Ag} \), \(^{137}\text{Cs} \), and \(^{134}\text{Cs} \) activities were
determined using the γ-ray detector, detecting peaks at 884.7, 661.7, and 604.7 keV, respectively. Multiple number of animals in each sample (Table 3) was put together in a cup and measured. Therefore no statistical errors were expressed in Tables. The concentration of the measured radioactivity was expressed as Bq/kgDW. Radioactivity in snakes, slugs, and tadpoles, all of which had high water content, and melted during starvation process was measured without desiccation of the specimens and expressed as Bq/kgFW, where FW is the fresh weight.

**Imaging analysis.** *N. clavata* specimens were gently flattened between paper towels, allowed to air-dry completely, and then exposed to BAS imaging plates (IP-plate; Fujifilm, Tokyo, Japan). An image of the vertical distribution of the radioactivity in the soil profile was obtained by exposing the vertical soil layer collected in a lunchbox (13 × 7.5 × 6 cm) to an IP-plate. In both cases, polyethylene film was placed between the sample and the IP-plate to avoid chemical- or water-related disturbances of the sensitivity of the IP-plate to the samples. After ~1
month of exposure, the IP-plates were scanned using an Image-analyzer (FLA-5000; FujiFilm). The feces of lizards and other fauna were observed by digital microscopy (KEYENCE VH-6300).

Results

Detection of $^{110m}$Ag in $N$. clavata. Figure 2 shows the $\gamma$-radiation from $N$. clavata (sample no. 3 in Table 3) detected by Ge-semiconductor analysis. Energy peaks corresponding to $^{137}$Cs (661.7 keV), $^{110m}$Ag (657.8 keV), and three other peaks characteristic of $^{110m}$Ag (884.7, 937.5, and 1384 keV) were obtained from the same sample. The radioactivity ratio of $^{110m}$Ag/$^{134}$Cs/$^{137}$Cs was 1/1.12/1.50.

Concentration of radioactivity in $N$. clavata and $A$. karschi. We collected $N$. clavata and surface soils from three different locations in Fukushima Prefecture and calculated the radioactivity concentration ratios, defined as ($^{110m}$Ag Bq/kg $N$. clavata)/($^{110m}$Ag Bq/kg soil). A similar ratio was calculated for $^{137}$Cs. As shown in Table 1, the radioactivity concentration ratios were 27.8, 74.5, and 87.3 for $^{110m}$Ag and 0.036, 0.078, and 0.071 for $^{137}$Cs at Oguni, Benten-yama, and Nimaibashi, respectively. These data were then used to calculate the radioactivity concentration ratios of $^{110m}$Ag to $^{137}$Cs, defined as ($^{110m}$Ag Bq/kg spider/$^{110m}$Ag Bq/kg soil)/($^{137}$Cs Bq/kg spider/$^{137}$Cs Bq/kg soil). The values were 774, 960, and 1234, at Ogumi, Benten-yama, and Nimaibashi, respectively.

We also measured the radioactivity concentration ratio in $A$. karschi living only on the soil surface as a ground spider, which would have received $^{110m}$Ag directly from the soil-based food chain, while $N$. clavata resides in aerial webs mainly depending on flying preys. As shown in Table 1, the concentration ratios of $^{110m}$Ag and $^{137}$Cs in $A$. karschi in samples collected at Benten-yama and Iitoi were 448 and 166, and 1.30 and 0.45, respectively, corresponding to $^{110m}$Ag to $^{137}$Cs radioactivity concentration ratios of 344 and 369 (Table 1). Thus, in both spider species, the concentration ratio of radioactive $^{110m}$Ag was two–three orders higher than that of $^{137}$Cs.

Distribution of radionuclides in the body, thread, excrement, and egg sac of $N$. clavata. Figure 3 shows the lifestyle of the female $N$. clavata spiders in the U8-cups. All $N$. clavata collected from Hiso on Oct. 13, 2013 were pregnant. During starvation in the U8-cups incubated at room temperature, one $N$. clavata delivered one egg sac containing...
many larval spiders (Fig. 3a). The egg sac was covered with a silky thread and hung on the web strung across the cup interior (Fig. 3d). Table 2 shows that the spider’s body and excrement contained equivalent concentrations of $^{110m}$Ag, whereas the concentration in the egg sacs was more than twofold higher. Web silk did not contain $^{110m}$Ag. However, the concentration of $^{137}$Cs (and $^{134}$Cs) in the spiders’ body and thread was nearly the same. Cesium might be incorporated as a contaminant into thread, replacing potassium. Spider excrement contained 4.5 times more $^{137}$Cs (and $^{134}$Cs) than measured in the spider body. Concentrated radioactivity was observed possibly at the midgut gland of

**Table 1.** Radioactivity concentration ratio between soil and *Nephila clavata* or *Atypus karschi*. To calculate the concentration ratio, the spider’s radioactivity, as shown in Table 3b, was divided by the corresponding soil radioactivity data shown in Table 6. The results obtained using the $^{134}$Cs and $^{137}$Cs data were almost the same (data not shown).

| Species            | Sampling place          | Ratio of the radioactivity concentrations: Spider (Bq/kg)/Soil (Bq/kg) | $^{110m}$Ag : $^{137}$Cs ratio |
|--------------------|-------------------------|---------------------------------------------------------------|-------------------------------|
|                    |                         | $^{110m}$Ag (a) | $^{137}$Cs (b) | (a)/(b) |
| *Nephila clavata*  | Oguni (2)               | 27.8          | 0.036        | 774 |
|                    | Benten-yama (1)         | 74.5          | 0.078        | 960 |
|                    | Nimaibashi (3)          | 87.3          | 0.071        | 1234 |
| *Atypus karschi*   | Benten-yama (16)        | 448           | 1.30         | 344 |
|                    | Iitoi (14)              | 166           | 0.45         | 369 |

Fig. 3. *N. clavata*. a: egg sac; b: web threads collected from the spiders; c: feces and urine collected from the spiders; d: dead body of *N. clavata* after release of the egg sac. An egg sac covered with a silky thread is shown. These threads usually had adhered dried residual materials. e: *N. clavata* during pregnancy.
N. clavata, while with lower concentrations of radioactivity in other body parts (Fig. 4).

Radionuclide contents in N. clavata during the 3.5 years after the NPP accident. Figure 5 shows the trend in the radionuclide contents of N. clavata over the sampling period. Although the dates and sites of N. clavata collection differed, the concentration of $^{110m}$Ag in the spiders clearly decreased rapidly within 3.5 years after the NPP accident, except in sample no. 11, which still had a detectable amount of $^{110m}$Ag and a very high amount of $^{137}$Cs (and $^{134}$Cs). This sample was collected at Tsushima, where aerial doses were higher (8.2 µSv/h) than those (1.3–4.6 µSv/h) at other sampling sites (Table 3a).

Amounts of $^{110m}$Ag, $^{134}$Cs, and $^{137}$Cs in various faunal species. Table 3a shows the original $^{110m}$Ag, $^{134}$Cs, and $^{137}$Cs data obtained at the faunal sampling times between Sept. 20, 2011 and Nov. 3, 2014, and Table 3b presents the decay-corrected data of Table 3a with reference to March 15, 2011, when the NPP discharged gaseous materials and the plume spread to the northwest. The $\gamma$-ray peak patterns of most of the faunal samples collected in 2011–2012 were similar to that of N. clavata (Fig. 2, Table 3a). As shown in the column n.d.$^{110m}$Ag Bq/kg" of Table 3b, silk spiders (sample no. 11), ground spiders (14, 16), pill bugs (24, 25), snails (30), dragonflies (54), crabs (60) and centipede (65) contained more than 5000 Bq/kg, whereas silk spiders (1, 2, 3, 4), ground spiders (15, 17, 19), other spiders (21, 22), the beetle

Table 2. Distribution of radionuclides derived from Nephila clavata into four parts. It was sampled at Hiso on Oct. 13, 2013 (sample no. 9 in Table 3). See the culture conditions of N. clavata in the U8-cup in the Materials and methods. Figures of each part are shown in Fig. 3.

| N. clavata    | Radioactivity (Bq/kg) |
|---------------|----------------------|
|               | $^{110m}$Ag | $^{134}$Cs | $^{137}$Cs |
| Whole body    | 78.5       | 546        | 1339       |
| Excrement     | 70.1       | 2480       | 6040       |
| Thread        | n.d.       | 756        | 1330       |
| Egg sacs      | 150        | 155        | 395        |

n.d.: not detected.
| Sample number | Faunal species | Number of measured samples | Radioactivity (Bq/kgDW) | Date of sampling | Sampling place | Aerial dose ($\mu$Sv/h) |
|---------------|---------------|----------------------------|------------------------|-----------------|---------------|----------------------|
| 1             | *Nephila clavata* (silk spider) | 5                          | 731                   | 989             | 1052          | 3.5                  |
| 2             | *Nephila clavata* (silk spider) | 6                          | 707                   | 949             | 1180          | 2.1                  |
| 3             | *Nephila clavata* (silk spider) | 6                          | 1398                  | 1559            | 2066          | 3.2                  |
| 4             | *Nephila clavata* (silk spider) | 10                         | 400                   | 980             | 1340          | 4.3                  |
| 5             | *Nephila clavata* (silk spider) | 5                          | n.d.                  | 772             | 1653          | 4.3                  |
| 6             | *Nephila clavata* (silk spider) | 4                          | n.d.                  | 512             | 1093          | 4.6                  |
| 7             | *Nephila clavata* (silk spider) | 6                          | n.d.                  | 538             | 1077          | 4.4                  |
| 8             | *Nephila clavata* (silk spider) | 3                          | 226                   | 993             | 2163          | 1.3                  |
| 9             | *Nephila clavata* (silk spider) | 35                         | 78.5                  | 546             | 1339          | 4.5                  |
| 10            | *Nephila clavata* (silk spider) | 14                         | 48.4                  | 107             | 385           | 3.5                  |
| 11            | *Nephila clavata* (silk spider) | 4                          | 139                   | 2369            | 8965          | 8.2                  |
| 12            | *Nephila clavata* (silk spider) | 8                          | 18.2                  | 378             | 1395          | 3.1                  |
| 13            | *Araneus ventricosus* (*Onigumo*) | 2                          | 162                   | 424             | 508           | 2.3                  |
| 14            | *Atypus karschi* (ground spider) | 8                          | 3211                  | 6346            | 8016          | 5.4                  |
| 15            | *Atypus karschi* (ground spider) | 12                         | 618                   | 1912            | 3169          | 4.5                  |
| 16            | *Atypus karschi* (ground spider) | 10                         | 2866                  | 11464           | 17445         | 4.5                  |
| 17            | *Atypus karschi* (ground spider) | 13                         | 232                   | 1383            | 2932          | 3.7                  |
| 18            | *Atypus karschi* (ground spider) | 4                          | n.d.                  | 300             | 1206          | 2.7                  |
| 19            | *Atypus karschi* (ground spider) | 4                          | 86.8                  | 513             | 1800          | 3.1                  |
| 20            | *Atypus karschi* (ground spider) | 4                          | n.d.                  | 436             | 686           | 2.9                  |
| 21            | *Argiope anoea* (*Koqanegumo*)  | 10                         | 140                   | 174             | 436           | 4.6                  |
| 22            | *Spider* (Unknown)                | 1                          | 182                   | 1218            | 2506          | 4.2                  |
| 23            | *Doricus herensis* (*Seaka-gominushi*) | 6                          | 491                   | 1006            | 1271          | 2.5                  |
| 24            | *Armadillidium vulgare* (pill bug) | 23                         | 2099                  | 8073            | 10759         | 2.7                  |
| 25            | *Armadillidium vulgare* (pill bug) | 25                         | 3906                  | 26734           | 34482         | 3.0                  |
| 26            | *Aphaenogaster faneleca* (ant)    | 20                         | n.d.                  | 11360           | 14768         | 4.2                  |
| 27            | *Aphaenogaster faneleca* (ant)    | 10                         | n.d.                  | 6233            | 7672          | 2.8                  |
| 28            | *Aphaenogaster faneleca* (ant)    | 32                         | 317                   | 871             | 1271          | 3.1                  |
| 29            | *Oligochaeta* (earthworm)         | 1                          | 20.8                  | 357             | 476           | 3.8                  |
| 30            | *Euhadra* (snail: *Maimai*)       | 43                         | 3118                  | 11466           | 14778         | 5.1                  |
| 31            | *Bufo japonicus formosus* (frog: *Hikigaeru*) | 1                          | 426                   | 5709            | 12130         | 5.5                  |
| 32            | *Rana rugosa* (frog: *Tsuchigaeru*) | 5                          | 206                   | 17785           | 23544         | 4.5                  |
| 33            | *Frog* (unknown)                  | 1                          | 53                    | 6519            | 18544         | 3.5                  |
| 34            | *Frog* (unknown)                  | 1                          | 74.6                  | 2584            | 7671          | 3.7                  |
| 35            | *Lacertidae* (lizard)             | 4                          | n.d.                  | 11500           | 14700         | 5.1                  |
| 36            | *Lacertidae* (lizard)             | 3                          | n.d.                  | 3300            | 4900          | 4.5                  |
| 37            | *Lacertidae* (lizard)             | 1                          | n.d.                  | 431             | 1338          | 2.7                  |
| 38            | *Elaphe climacophora* (snake)†    | 1                          | 18.2                  | 274             | 504           | 2.2                  |
| 39            | *Patanga japonica* (locust)       | 8                          | 25.7                  | 816             | 829           | 3.5                  |
| 40            | *Patanga japonica* (locust)       | 4                          | n.d.                  | 111             | 251           | 2.3                  |
| 41            | *Diestrammena apicalis* (*Kanadouna*) | 4                          | 65                    | 1348            | 2563          | 2.2                  |
| 42            | *Thereuonema tuberculata* (millipede: *Gej*) | 3                          | 578                   | 1724            | 2550          | 2.0                  |
| 43            | *Megkinitmon bilineatum* (slug)†   | 5                          | 165                   | 2833            | 5095          | 3.4                  |
| 44            | *Megkinitmon bilineatum* (slug)†   | 6                          | 233                   | 6856            | 13272         | 3.3                  |
| 45            | *Anonymlina dictatoma* (female)(beetle) | 1                          | n.d.                  | 845             | 1445          | 1.4                  |

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Seaka-gomimushi (23), ants (28), frogs (34), millipedes (42), slugs (44), dragonflies (56), and river shrimp (57) contained 1000–5000 110mAg Bq/kg. In contrast, 110mAg was not detectable in lizards, locusts, beetles, longicorn beetles, honeybees, butterflies, tadpoles, and newts, although 137Cs (or 134Cs) was detected in all of them. Table 3b also shows that radionuclide accumulation in spider bodies in the area of Iitoi, Iitate village, from Oct. 18, 2011 to May 5, 2012, the radioactivity was measured without desiccation and expressed as Bq/kgFW.

### Table 3b. Radionuclide Contents of Faunal Species from the Contaminated Soils Examined

| Sample number | Faunal species | Number of measured samples | Radionuclide (Bq/kgDW) | Date of sampling | Sampling place | Aerial dose (µSv/h) | 110mAg 134Cs 137Cs 110mAg 134Cs 137Cs |
|---------------|---------------|----------------------------|------------------------|------------------|------------------|-------------------|------------------------|
| 46 | Alomyrina dichotoma (male/bee) | 1 | n.d. | 580 1007 | 2012/7/12 | Benten-yama | 1.3 0.58 | n.d. |
| 47 | Cerambicidae (longicorn: Kamikiri) | 1 | n.d. | 70.1 105 | 2012/8/16 | Benten-yama | 1.2 0.67 | n.d. |
| 48 | Gryptopsaltria usurofuscata (Aburazemi) | 10 | n.d. | 699 1196 | 2012/8/17 | Benten-yama | 1.5 0.58 | n.d. |
| 49 | Pentatoma japonica (Kamemushi) | 4 | n.d. | 129 207 | 2012/6/15 | Maeda | 4.0 0.62 | n.d. |
| 50 | Vespa mandarinia (hornet: Suzumebachi) | 4 | 273 | 8206 11320 | 2011/11/9 | Iitoi | 4.5 0.72 | 0.024 |

Notes: 1: measured without desiccation and expressed as Bq/kgFW. n.d.: not detected, not determined.

### Discovery of 110mAg in faunal species after Fukushima NPP accident

No. 4  
Continued.

Seaka-gomimushi (23), ants (28), frogs (34), millipedes (42), slugs (44), dragonflies (56), and river shrimp (57) contained 1000–5000 110mAg Bq/kg. In contrast, 110mAg was not detectable in lizards, locusts, beetles, longicorn beetles, honeybees, butterflies, tadpoles, and newts, although 137Cs (or 134Cs) was detected in all of them. Table 3b also shows that radionuclide accumulation in spider bodies in the area of Iitoi, Iitate village, from Oct. 18, 2011 to May 5, 2012, the radioactivity was measured without desiccation and expressed as Bq/kgFW. n.d.: not detected, not determined.

### Radionuclide contents of A. karschi during the 3.5 years after the NPP accident

A. karschi was collected as the representative spider to monitor radionuclide accumulation in spider bodies in the area of Iitoi, Iitate village, from Oct. 18, 2011 to Oct. 13, 2014. As an inhabitant of the surface soil, A. karschi was assumed to more directly reflect soil radionuclides available through the food chain than N. clavata, which mostly resides in aerial webs. The fixed sampling site, where the aerial rate of radioactivity accumulation was 5.37 µSv/h on the first sampling date, Oct. 18, 2011, but had decreased to 2.75 µSv/h on the fourth sampling date, Oct. 13, 2014. As shown in Table 5, during the 6 months from Oct. 18, 2011 to May 5, 2012, the radioactivity of 110mAg, 134Cs, and 137Cs decayed from 3211 to 8016 Bq/kg, and 8016 to 3169 Bq/kg, respectively. If each radionuclide found
Table 3b. Decay corrected data of Table 3a. All data of Table 3a was corrected following physical half-life of each nuclide to the date Mar. 15, 2011

| Sample number | Faunal species                      | Bq/kgDW | 110mAg | 134Cs | 137Cs | 110mAg/137Cs |
|---------------|------------------------------------|---------|--------|-------|-------|--------------|
| 1             | Nephila clavata (silk spider)      | *1640   | 1293   | 1072  |       | 1.5          |
| 2             | Nephila clavata (silk spider)      | *1559   | 1286   | 1201  |       | 1.3          |
| 3             | Nephila clavata (silk spider)      | *3754   | 2163   | 2144  |       | 1.8          |
| 4             | Nephila clavata (silk spider)      | *1074   | 1360   | 1370  |       | 0.78         |
| 5             | Nephila clavata (silk spider)      | n.d.    | 1524   | 1709  | n.d.  |              |
| 6             | Nephila clavata (silk spider)      | n.d.    | 1011   | 1130  | n.d.  |              |
| 7             | Nephila clavata (silk spider)      | n.d.    | 878    | 1129  | n.d.  |              |
| 8             | Nephila clavata (silk spider)      | 986     | 1618   | 2237  | 0.78  |
| 9             | Nephila clavata (silk spider)      | n.d.    | 878    | 1129  | n.d.  |              |
| 10            | Nephila clavata (silk spider)      | 495     | 259    | 409   |       | 1.2          |
| 11            | Nephila clavata (silk spider)      | **5167  | 7852   | 9734  | 0.53  |
| 12            | Nephila clavata (silk spider)      | 677     | 1253   | 1515  | 0.45  |
| 13            | Araneus ventricosus (Onigumo)      | 344     | 544    | 517   | 0.67  |
| 14            | Atypus karschi (ground spider)     | **6811  | 8142   | 8154  | 0.84  |
| 15            | Atypus karschi (ground spider)     | *1955   | 2800   | 3253  | 0.61  |
| 16            | Atypus karschi (ground spider)     | **9852  | 17261  | #17942| 0.55  |
| 17            | Atypus karschi (ground spider)     | *2089   | 2863   | 3082  | 0.68  |
| 18            | Atypus karschi (ground spider)     | n.d.    | 1193   | 1309  | n.d.  |              |
| 19            | Atypus karschi (ground spider)     | *3882   | 1800   | 1960  | 2.0   |
| 20            | Atypus karschi (ground spider)     | n.d.    | 1630   | 784   | n.d.  |              |
| 21            | Argiope amoena (Kogamegumo)        | *1260   | 360    | 458   | 1.8   |
| 22            | Spider (Unknown)                   | *1639   | 2523   | 2634  | 0.62  |
| 23            | Doricus herensis (Seaka · gonimushi)| *1469  | 1447   | 1303  | 1.1   |
| 24            | Armadillidium vulgare (pill bug)   | **5482  | 11097  | #10997| 0.50  |
| 25            | Armadillidium vulgare (pill bug)   | **13998 | 40812  | #35869| 0.39  |
| 26            | Aphaenogaster famelica (ant)       | n.d.    | 14942  | #15049| n.d.  |
| 27            | Aphaenogaster famelica (ant)       | n.d.    | 9214   | 7831  | n.d.  |              |
| 28            | Aphaenogaster famelica (ant)       | *1234   | 1366   | 1311  | 0.94  |
| 29            | Oligochaeta (earthworm)            | 46      | 464    | 485   | 0.095 |
| 30            | Euhadra (snail: Mainsai)           | *14504  | 19084  | #15304| 0.95  |
| 31            | Bufo japonicus (frog: Hikigaeru)   | *5517   | 13341  | #12858| 0.43  |
| 32            | Rana rugosa (frog: Tsuchigaeru)    | 650     | 26026  | #24957| 0.026 |
| 33            | Frog (unknown)                     | 778     | 15878  | #19713| 0.039 |
| 34            | Frog (unknown)                     | *1095   | 6294   | 8155  | 0.13  |
| 35            | Lacertidae (lizard)                | n.d.    | 13949  | *1426 | n.d.  |
| 36            | Lacertidae (lizard)                | n.d.    | 5507   | 5320  | n.d.  |              |
| 37            | Lacertidae (lizard)                | n.d.    | 1429   | 1452  | n.d.  |              |
| 38            | Elaphe climacophora (snake)        | 85.8    | 458    | 522   | 0.16  |
| 39            | Patanga japonica (locust)          | 57.6    | 1066   | 844   | 0.068 |
| 40            | Patanga japonica (locust)          | n.d.    | 236    | 264   | n.d.  |              |
| 41            | Dietrammena apicalis (Kanadousa)   | 382     | 2424   | 2668  | 0.14  |
| 42            | Theromopara tuberculata (millipede: Geji) | *3329 | 3080 | 2654  | 1.3  |
| 43            | Meghinatium bilineatum (slug)      | *932    | 3529   | 5300  | 0.18  |
| 44            | Meghinatium bilineatum (slug)      | *1130   | 11570  | #13758| 0.082 |
| 45            | Allomyrina dichotoma (female beetle)| n.d.  | 1413   | 1497  | n.d.  |              |
| 46            | Allomyrina dichotoma (male beetle) | n.d.    | 970    | 1043  | n.d.  |              |

Continued on next page.
in A. karschi on Oct. 18, 2011 had decayed following its physical half-life, then the values on May 5, 2012 should have been 1859, 5294, and 7916 Bq/kg, respectively. However, the detected radioactivity was far less. Moreover, on Oct. 13, 2014, $^{110}\text{mAg}$ was no longer detected in A. karschi, although $^{137}\text{Cs}$ and $^{134}\text{Cs}$ were still present in large amounts.

Radioactivity in soils. In contrast, the $\gamma$-ray traces of all surveyed soils showed very low peaks for $^{110}\text{mAg}$. $^{110}\text{mAg}$ was detected in soils at concentrations between 22 and 238 Bq/kg (Table 6). For example, the radioactivity ratio of $^{110}\text{mAg}/^{134}\text{Cs}/^{137}\text{Cs}$ in Nimaibashi soil collected on the same day as N. clavata was 1/675/703, which was very different from that of N. clavata (1/1.12/1.50. See Results). The radioactivity ratio of $^{110}\text{mAg}/^{137}\text{Cs}$ was between 0.0014 and 0.0023. These values were almost the same as those of the riverbed sediments of the four sampling locations of Nitta River (0.0016–0.0036), Takanokura Dam (0.0008) of the Nitta River, and

| Sample number | Faunal species       | $^{110}\text{mAg}$ | $^{134}\text{Cs}$ | $^{137}\text{Cs}$ | $^{110}\text{mAg}/^{137}\text{Cs}$ |
|---------------|---------------------|--------------------|-------------------|-------------------|--------------------------------|
| 47            | Cerambycidae (longicorn) | n.d.              | 117               | 109               | n.d.                          |
| 48            | Graitopsaltria nigrofuscata (Aburazemi) | n.d.              | 1503              | 1239              | n.d.                          |
| 49            | Pentatoma japonica (Kamemushi) | n.d.              | 236               | 216               | n.d.                          |
| 50            | Vespa mandarina (hornet: Suzunebuchi) | 649               | 10933             | $^{a}$11545       | 0.056                         |
| 51            | Apis cerana japonica Rad (honeybee) | n.d.              | 789               | 851               | n.d.                          |
| 52            | Rhopalocera (butterfly: Unknown) | n.d.              | 296               | 264               | n.d.                          |
| 53            | Rhopalocera (butterfly: Monshiroeco) | n.d.              | 4324              | 3434              | n.d.                          |
| 54            | Anotogaster sieboldi (dragonfly) | **8177            | 16979             | $^{a}$19774       | 0.41                          |
| 55            | Sympetrum croceolab (dragonfly) | 849               | 1065              | 1543              | 0.55                          |
| 56            | Sympetrum frequens (dragonfly) | *3465             | 4899              | 3628              | 0.96                          |
| 57            | Palaemon paucidens (river shrimp) | *2688             | 2375              | 2585              | 1.0                           |
| 58            | Geothelphusa dehaani (crab: Sawagani) | 970               | 311               | 240               | 1.0                           |
| 59            | Geothelphusa dehaani (crab: Sawagani) | 4025              | 31384             | 32926             | 0.14                          |
| 60            | Geothelphusa dehaani (crab: Sawagani) | **11039           | 13825             | $^{a}$15084       | 0.73                          |
| 61            | Bellamy quadrata histrica (Tanishi) | 58.8              | 439               | 503               | 0.12                          |
| 62            | Cybester japonicus (Gengoro) | 274               | 255               | 268               | 1.0                           |
| 63            | tadpole | n.d. | 135 | 155 | n.d. |
| 64            | Cynops pyrrhogaster (newt: Akaharaimori) | n.d. | 484 | 1760 | n.d. |
| 65            | Epinomphisa (centipede: Mulade) | **18782           | 3099              | 3595              | 5.2                           |

*: 1000–5000 $^{110}\text{mAg}$ Bq/kg. **: >5000 $^{110}\text{mAg}$ Bq/kg. $^{a}$: >10000 $^{137}\text{Cs}$ Bq/kg. $^{b}$: >1.0 of $^{110}\text{mAg}/^{137}\text{Cs}$. n.d.: not detected, not determined.

Table 4. $^{110}\text{mAg}/^{137}\text{Cs}$ radioactivity ratio in the feces of some of the studied faunal species and the radioactivity concentration ratio for their feces vs. soil. All data are calculated after decay correction to the date Mar. 15, 2011
of fl...top soil, which includes a litter...2 cm of the top soil, which includes a litter

Figure 6 shows the autoradiograph of the forest soil...March 15, 2011, with contamination occurring by precipitation.

During our field research on the contamination of flora in Ibaraki,25) and Fukushima,26) we coinci-
dently detected large amounts of $^{110m\text{Ag}}$ in the spider $N. \text{clavata}$ (Fig. 2). This finding was subsequently confirmed in other faunal species. Among the many species surveyed (Table 3a), those belonging to

Tetsuzen Dam (0.01) of Ota River.16) These sampling places are located in Minamisoma city, Fukushima Prefecture and to the northwest from Fukushima Dai-ichi NPP. The almost constant $^{110m\text{Ag}}/^{137\text{Cs}}$ ratios in those soils suggested that our research area (100 km × 60 km) was contaminated with $^{110m\text{Ag}}$ and $^{137\text{Cs}}$ (and $^{134\text{Cs}}$) all at once shortly after the release of huge amount of radioactivity on March 15, 2011 (Table 3b). In contrast, $^{110m\text{Ag}}$ concentrations in soils were very low, along with the $^{110m\text{Ag}}/^{137\text{Cs}}$ ratio in soils (0.0014–0.0023) (Table 6) compared to the concentrations of these two radionuclides in all faunal species in which $^{110m\text{Ag}}$ were detected (Table 3b).

In both spider species ($N. \text{clavata}$ and $A. \text{karschi}$), the radioactivity concentration ratio of $^{110m\text{Ag}}$ was three orders of magnitude higher than that of $^{137\text{Cs}}$ (Table 1). The most plausible explanation for this unexpected result is that $^{137\text{Cs}}$ was more rapidly bound to soil clay minerals, especially mica15) or illite,27) than $^{110m\text{Ag}}$, resulting in less $^{137\text{Cs}}$ available to flora and/or fauna in the soil. Indeed, the $^{137\text{Cs}}$ content of most of the vegetables and rice in Fukushima Prefecture has decreased very quickly in the 3 years since the NPP accident.28) Alternatively, the Ag-transporter activity of both spiders may be much higher than their Cs-transporter activity, resulting in the more efficient uptake of $^{110m\text{Ag}}$. Otherwise, as mentioned later the binding activity of $^{110m\text{Ag}}$ to hemocyanin in lymph fluid in both spiders may be very strong, thus $^{110m\text{Ag}}$ may be more slowly metabolized than radioactive Cs to be excreted. Another explanation is that $^{110m\text{Ag}}$ is more quickly turned over through the food chain to spiders in terrestrial environments than $^{137\text{Cs}}$. To confirm the latter three possibilities will require advanced biochemical and molecular biological works on mineral (Ag and/or Cs) transporters in flora and fauna.

Although we could not segregate the radioactivity sources in the labeling image of $N. \text{clavata}$ and thus unable to localize $^{110m\text{Ag}}$ and $^{137\text{Cs}}$ (or $^{134\text{Cs}}$) anatomically, the image shown in Fig. 4 suggests that $^{110m\text{Ag}}$ is circulated in the lymph fluid, thus binding to hemocyanin, the O₂ carrier protein that also plays an important role in metal transport and distribution in decapod crustaceans.29) The trunk, head, and legs of $N. \text{clavata}$, as depicted in Fig. 4B, were probably labeled mainly with radioactive Cs. Autoradiographs (BAS-images) of many small animals collected from the contaminated field were taken by one of the authors (S. Mori) and published online.30),31) The muscles of frog and snake are very strongly labeled with radioactive Cs. These observations are consistent with the very low $^{110m\text{Ag}}/^{137\text{Cs}}$ ratio of those fauna (see the $^{110m\text{Ag}}/^{137\text{Cs}}$ ratio of snake no. 38 and frog nos. 32–34 in Table 3a).

Discussion

During our field research on the contamination of flora in Ibaraki,25) and Fukushima,26) we coinci-dently detected large amounts of $^{110m\text{Ag}}$ in the spider $N. \text{clavata}$ (Fig. 2). This finding was subsequently confirmed in other faunal species. Among the many species surveyed (Table 3a), those belonging to

$^a$ Radioactivity in the samples collected on the first sampling date (Oct. 18, 2011) decayed following the physical half-life of each radionuclide.

$^b$ Radioactivity in the sample nos. 14, 15, 17 and 18 in Table 3a, respectively, at the fixed observation site in Iitoi. Parentheses indicate the calculated data based on the assumption that the radioactivity in the samples collected on the first sampling date (Oct. 18, 2011) decayed following the physical half-life of each radionuclide.

Table 6. Concentration of soil radioactivity and the $^{110m\text{Ag}}/^{137\text{Cs}}$ ratio. All data was corrected following physical half-life of each nuclide to the date Mar. 15, 2011

| Date of sampling | $^{110m\text{Ag}}$ (Bq/kg) | $^{134\text{Cs}}$ (Bq/kg) | $^{137\text{Cs}}$ (Bq/kg) | $^{110m\text{Ag}}/^{137\text{Cs}}$ |
|------------------|--------------------------|--------------------------|--------------------------|--------------------------|
| 2011/10/18       | 3211                     | 6346                     | 8016                     |                          |
| 2012/5/5         | 618 (1859)               | 1912 (5294)              | 3169 (7916)              |                          |
| 2013/6/5         | 232 (742)                | 1383 (3909)              | 2932 (7754)              |                          |
| 2014/10/13       | 0 (144)                  | 360 (2184)               | 1206 (7469)              |                          |

Table 6. Concentration of soil radioactivity and the $^{110m\text{Ag}}/^{137\text{Cs}}$ ratio. All data was corrected following physical half-life of each nuclide to the date Mar. 15, 2011

| Soils            | Radioactivity (Bq/kg) in soils | $^{110m\text{Ag}}$ (Bq/kg) | $^{134\text{Cs}}$ (Bq/kg) | $^{137\text{Cs}}$ (Bq/kg) | $^{110m\text{Ag}}/^{137\text{Cs}}$ |
|------------------|-------------------------------|---------------------------|---------------------------|---------------------------|---------------------------|
| 1 Oguni          | 56                            | 32000                     | 33400                     | 0.0017                    |
| 2 Hanami-yama    | 42                            | 25600                     | 26500                     | 0.0016                    |
| 3 Benten-yama    | 22                            | 12900                     | 13800                     | 0.0016                    |
| 4 Tsushima       | 238                           | 105900                    | 108000                    | 0.0022                    |
| 5 Iitoi          | 41                            | 17200                     | 18100                     | 0.0023                    |
| 6 Nimaibashi     | 43                            | 29100                     | 30300                     | 0.0014                    |
| 7 Hiso           | 37                            | 18200                     | 21300                     | 0.0017                    |
Fig. 6. Radioactivity image of the forest soil profile of Tsushima. See the data for soil no. 4 in Table 6.

Fig. 7. Body fragments of *Atypus karschi* were the major components of the feces of the lizards collected at the observation site in Iitoi. The digital microscopy photo was taken on Jan. 30, 2012.
The fact that the concentration of $^{110m}$Ag in the egg sac of *N. clavata* (Fig. 3a) was more than twice that of the spider's body (Table 2) suggests that $^{110m}$Ag was transferred from the female spider to the egg sac. Because one egg sac incubates hundreds of larval spiders, separating them from the fragile sac after it was crushed was difficult (Fig. 3a). Thus, the portion of $^{110m}$Ag transferred from female spiders to its larvae could not be determined.

In Arthropoda and Mollusca that inhabit the sea, such as crab, octopus, and squid, etc., $^{110m}$Ag was suggested to replace copper in the active center of hemocyanin. It is reported that the regeneration of holohemocyanin by the combination of apoprotein with added cuprous chloride was inhibited by Ag$^+$. Although this has yet to be proven more directly using $^{110m}$Ag. The same may be true in the case of land fauna species living in freshwater, such as river shrimp, crab, and diving beetle *Gengoro*, whose $^{110m}$Ag contents and $^{110m}$Ag/$^{137}$Cs ratios were relatively high (Table 3). The amount of $^{110m}$Ag concentrated by phytoplankton or zooplankton may be a determining factor, as both would serve as the primary source of $^{110m}$Ag in the food chain of freshwater ecosystems. It was interesting that snail (sample no. 30 in Table 3) belonging to Mollusca had also relatively high $^{110m}$Ag content and $^{110m}$Ag/$^{137}$Cs ratio.

$^{110m}$Ag was not detectable in lizards, locusts, beetles, longicorn beetles, honeybees, butterflies, tadpoles, and newts, which may have been due to very low Ag-transporter activity in the digestive organs of these species. The lizards were collected at the same fixed point where we also collected the ground spider *A. karschi* annually. As shown in Fig. 7, a major component of lizard feces comprised fragmented *A. karschi*. This spider highly concentrated $^{110m}$Ag in its body (sample nos. 14–20 in Table 3a), whereas the lizard did not (sample nos. 35–37 in Table 3a). This high amount of $^{110m}$Ag in the feces of lizards suggests that the lizards excreted highly concentrated $^{110m}$Ag without absorption of the radionuclide from the digested *A. karschi*. Thus, lizards may have very low intestinal Ag-transporter activity. *A. karschi*, however, almost completely absorbed $^{110m}$Ag into its body from unknown foods because no detectable $^{110m}$Ag was found in the feces of this spider (Table 4).

The radioactivity image of the soil profile (Fig. 6) agrees with the results of Lepage *et al.* (see Figs. 5, 7 and 8 in their recent report). Although we could not differentiate $^{110m}$Ag from $^{137}$Cs and $^{134}$Cs in the image, the three radionuclides seemed to have co-localized in the topsoil within a depth of 2 cm. Silver tends to concentrate in the surface litter plus humus horizon, suggesting that it mixes with organic matter. Increased acidity facilitates the removal of Ag from this horizon, whereas at a pH above 4, Ag is relatively immobile. If we define the residence half time for $^{110m}$Ag in soil as the time required for one-half of an acute $^{110m}$Ag deposit to pass through the soil layer of interest, the residence half time resulting from the migration velocity of Ag within the 5-cm soil top layer would be $33 \pm 3$ years. Therefore, the vertical migration of $^{110m}$Ag may be highly dependent on the soil type.

This study shows that faunal concentrations of $^{110m}$Ag released during the Fukushima Dai-ichi NPP accident are decreasing more rapidly than the physical half-life of the radionuclide in the contaminated terrestrial environment of Fukushima (Fig. 5, Table 5). Due to the Chernobyl and Fukushima disasters, the precise physicochemical dynamics of radioactive Cs with soil minerals have received considerable attention from soil scientists around the world. However, studies on the biological dynamics of radioactive Cs in the “soil sphere” are lacking. Clarification of not only the physicochemical dynamics of soil minerals, but also the biological dynamics of $^{110m}$Ag in the “soil sphere”, will likely provide the information needed to explain why faunal concentrations of $^{110m}$Ag released by the Fukushima Dai-ichi NPP meltdown are decreasing faster through the food chain than predicted by their physical half-life.

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