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**Environmental mobility of  $^{110m}\text{Ag}$ : lessons learnt from Fukushima accident (Japan) and potential use for tracking the dispersion of contamination within coastal catchments**

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1 **Environmental mobility of  $^{110m}\text{Ag}$ : lessons learnt from Fukushima accident**  
2 **(Japan) and potential use for tracking the dispersion of contamination within**  
3 **coastal catchments**

4

5 **Abstract**

6 Silver-110 metastable ( $^{110m}\text{Ag}$ ) has been far less investigated than other anthropogenic  
7 radionuclides, although it has the potential to accumulate in plants and animal tissues. It is  
8 continuously produced by nuclear power plants in normal conditions, but emitted in much  
9 larger quantities in accidental conditions facilitating its detection, which allows the  
10 investigation of its behaviour in the environment. We analysed  $^{110m}\text{Ag}$  in soil and river drape  
11 sediment (i.e., mud drapes deposited on channel-bed sand) collected within coastal  
12 catchments contaminated in Fukushima Prefecture (Japan) after the Fukushima Dai-ichi  
13 Nuclear Power Plant accident that occurred on 11 March 2011. Several field experiments  
14 were conducted to document radiosilver behaviour in the terrestrial environment, with a  
15 systematic comparison to the more documented radiocesium behaviour. Results show a  
16 similar and low mobility for both elements in soils and a strong affinity with the clay fraction.  
17 Measurements conducted on sediment sequences accumulated in reservoirs tend to confirm  
18 a comparable migration and deposition of those radionuclides even after their redistribution  
19 due to erosion and deposition processes. Therefore, as the  $^{110m}\text{Ag}$ : $^{137}\text{Cs}$  initial activity ratio  
20 varied in soils across the area, we justified the relevance of using this tool to track the  
21 dispersion of contaminated sediment from the main inland radioactive pollution plume  
22 generated by FDNPP accident.

## 23 1. Introduction

24 Silver-110 metastable ( $^{110m}\text{Ag}$ ) is a radioisotope with a rather short half-life of 249 days  
25 produced in nuclear power plants (NPP) as an activation product of  $^{109}\text{Ag}$ . In NPP, natural  
26 silver (which contains 48% of  $^{109}\text{Ag}$ ) is found in control rods or in the alloy used to seal the  
27 head of the reactor (Calmon and Garnier-Laplace 2002; Chelet 2006; IAEA 1998). In addition,  
28  $^{109}\text{Ag}$  may also be a fission product from uranium and plutonium (Chelet, 2006).  $^{110m}\text{Ag}$  can  
29 be measured by spectrometry gamma (with the main peak at 658 keV, and secondary peaks  
30 at 885 and 937 keV).

31 Le Petit et al. (2012) described the  $^{110m}\text{Ag}$  as a volatile fuel particle displaying some common  
32 characteristics with semi-volatile substances, due to its well-known strong retention within  
33 the reactor core structures. Even though  $^{110m}\text{Ag}$  is produced by NPP in normal operational  
34 conditions, its release into the environment remains limited (Ciffroy et al., 2005; Eyrolle et  
35 al., 2012) (Table 1), and research on this radionuclide has mainly gained interest after  
36 Chernobyl and Fukushima Dai-ichi accidents when it was emitted in much larger quantities,  
37 which facilitated its measurement. A search of the Web of Knowledge citation index (July  
38 2013) shows that the number of publications about  $^{110m}\text{Ag}$  increased significantly after  
39 Chernobyl accident (Fig. 1). A similar increase in attention has already been observed shortly  
40 after Fukushima accident.

41 Few information are available on the environmental mobility of  $^{110m}\text{Ag}$ , and most of the  
42 studies regarding this radioisotope were conducted in laboratory (Calmon and Garnier-  
43 Laplace 2002) because the conditions necessary to conduct field experiments were rarely  
44 met, as samples should be collected within the months following a release. They should  
45 alternatively be recovered in large quantities and analysed with high sensitivity detectors

46 during relatively long counting times. Still, it remains important to better understand its  
47 behaviour in the environment, as it has been demonstrated that silver is one of the most  
48 toxic metals (Bryan, 1971; Ratte, 1999) and that  $^{110m}\text{Ag}$  may be transferred from the soils to  
49 the plants (Handl et al., 2000; Shang and Leung, 2003), and accumulated in animal tissues  
50 (Adam et al., 2001; Beresford et al., 1998; Bryan and Langston, 1992; Khangarot and Ray,  
51 1987; Martin and Holdich, 1986; Oughton, 1989). It can also contaminate natural silver  
52 (isotopic contamination) used for photographic industry and jewellery manufacturing  
53 (Vuković, 2002).

54 Besides this interest to better understand the radioecology of  $^{110m}\text{Ag}$ , it has been shown that  
55 this radioisotope was emitted by the Fukushima Dai-ichi Nuclear Power Plant (FDNPP)  
56 accident and subsequently detected in the environment (Fukuda et al., 2013; MEXT, 2011;  
57 Tazoe et al., 2012; Watanabe et al., 2012). It is estimated that 20% of the radionuclides  
58 emitted were deposited on the soils of Fukushima Prefecture as a result of wet and dry  
59 atmospheric fallout, creating a contamination plume extending up to 70 km to the  
60 northwest of the FDNPP site (Kinoshita et al., 2011; Yasunari et al., 2011). It was estimated  
61 that between 7 and 36 PBq of  $^{137}\text{Cs}$  ( $T_{1/2} = 30.17\text{y}$ ) were released (Chino et al., 2011; Stohl et  
62 al., 2012; Winiarek et al., 2012), and  $^{110m}\text{Ag}$  release rate represented about 11% of the  $^{137}\text{Cs}$   
63 rate (Petit et al., 2012).

64 Depending on the behaviour of the radionuclides and their affinity with the fine mineral  
65 particles and the associated organic matter fraction, they may subsequently be redistributed  
66 across hillslopes as a consequence of runoff and soil erosion processes and be delivered to  
67 downstream rivers that may finally supply them to the ocean (Tanaka et al., 2012; Tateda et  
68 al., 2013; Ueda et al., 2013). Even though the bulk of initial marine contamination originated

69 from atmospheric deposition, erosion processes may have partly supplied the  $^{110\text{m}}\text{Ag}$  that  
70 was detected in zooplankton in the Pacific Ocean shortly after the accident (Buessler et al.,  
71 2012).

72 In order to document this transfer, Chartin et al. (2013) showed that the spatial patterns of  
73  $^{110\text{m}}\text{Ag}:^{137}\text{Cs}$  activity ratio in soils varied across space within the Fukushima contamination  
74 plume. They subsequently used this ratio to track the dispersion of contaminated sediment  
75 along a coastal river draining a catchment located within the main radioactive plume.

76 However, to date, most studies conducted in Fukushima post-accidental conditions focused  
77 on  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  behaviour (Inoue et al. 2013; Koarashi et al. 2012; Kato et al. 2012a,  
78 2012b; Matsunaga et al. 2013). Investigations reporting the presence of  $^{110\text{m}}\text{Ag}$  in soil did not  
79 detail its distribution with depth nor its behaviour (Fukuda et al., 2013; Tazoe et al., 2012;  
80 Watanabe et al., 2012). In this context, there is a lack of data collected in this specific post-  
81 accidental context to document the relative behaviour of  $^{110\text{m}}\text{Ag}$  and  $^{137}\text{Cs}$  in soils and  
82 sediment in order to check the relevance of using this ratio to track the dispersion of  
83 contaminated material along rivers. Even though both radionuclides are particle-reactive,  
84 the  $^{110\text{m}}\text{Ag}:^{137}\text{Cs}$  activity ratio in mobilized sediment might not simply reflect the catchment  
85 soil average of this ratio.

86 In this context, this paper aims to compile original experimental data acquired in Fukushima  
87 Prefecture during the months that followed the accident to document  $^{110\text{m}}\text{Ag}$  behaviour in  
88 soils and sediment. This paper will particularly address four issues:

- 89 (1) the affinity of  $^{110\text{m}}\text{Ag}$  with different grain size fractions, to check whether there is a  
90 different behaviour of  $^{110\text{m}}\text{Ag}$  and  $^{137}\text{Cs}$  depending on the particle size;
- 91 (2) the migration of  $^{110\text{m}}\text{Ag}$  in the soil, to investigate its potential movement with depth;

92 (3) the evolution of  $^{110m}\text{Ag}$  activities within sediment sequences accumulated in  
93 reservoirs, to document whether characteristics of contaminated sediments  
94 delivered to the reservoir by successive flood events change with time;

95 (4) the evolution of  $^{110m}\text{Ag}$  activities within sediment drape deposits, that consist of mud  
96 drapes deposited on channel-bed sand (Olley et al., 2013), collected at the same  
97 locations along the rivers after a succession of typhoon and snowmelt events to  
98 investigate the impact of those events on  $^{110m}\text{Ag}$  redistribution in catchments.

99 We will systematically compare the radiosilver behaviour to the one of the well documented  
100 radiocaesium, and to the literature data. The implications of those results will finally be  
101 discussed to decide whether the use of the  $^{110m}\text{Ag}:$  $^{137}\text{Cs}$  activity ratio is relevant to track the  
102 dispersion of contaminated sediment from the main radioactive pollution plume generated  
103 by FDNPP accident.

## 104 **2. Materials and methods**

### 105 2.1. Study area

106 The study was conducted in Fukushima Prefecture, located in Northeastern Japan. We  
107 focused on several coastal catchments (i.e. Mano, Nitta, Ota and Odaka catchments) (1000  
108 km<sup>2</sup>) draining the main part of the radioactive plume extending to the northwest of FDNPP  
109 (Fig. 2). Those catchments extend from the coastal mountain range (at approximately 30-km  
110 distance of the coast) to the Pacific Ocean and their elevation ranges from 0 to 900 m.

111 Plateaus on the west consist of Cretaceous granite and granodiorite, evolving into a complex  
112 patchwork of volcanic (both intrusive and extrusive), metamorphic (gneiss and schist) and  
113 plutonic (granite mainly) rocks. In the middle part of the catchments, marine and continental

114 sedimentary rocks of various ages separated by fault systems are found. In contrast, eastern  
115 coastal plains are mainly composed of Tertiary and Quaternary marine and continental  
116 sedimentary rocks.

117 Woodland is the main land use in this area (covering 68–78% of the total surface), followed  
118 by cropland (10–13%). The region is exposed to typhoons and spring snowmelt events  
119 leading to severe soil erosion and subsequent export of sediment in rivers (Chartin et al.,  
120 2013; Evrard et al., 2013). We therefore conducted three sampling campaigns after each of  
121 those major hydro-sedimentary events (i.e., Nov. 2011, April 2012, and Nov. 2012).

## 122 2.2. Sample collection, preparation and description of experiments.

123 After the accident, the Ministry of Education, Culture, Sports, Science and Technology in  
124 Japan (MEXT) supervised the sampling of soils at 2200 sites located within a radius of  
125 approximately 100 km around FDNPP in June and July 2011, and the analysis of their  
126 activities in several radionuclides contained in their 5-cm upper layer ( $^{110m}\text{Ag}$  activities were  
127 only provided for a selection of 345 sites; MEXT 2011).

128 Based on this dataset, a map of  $^{110m}\text{Ag}$ : $^{137}\text{Cs}$  activity ratio values in soils was drawn (Chartin  
129 et al., 2013). Unlike  $^{134}\text{Cs}$ : $^{137}\text{Cs}$  activity ratio, it revealed the presence of different  
130 contamination patterns across the area (Fig. 3), with lower values across mountains and  
131 higher values in the coastal lowlands.

132 The relatively low abundance of  $^{110m}\text{Ag}$  in soils of the study area in Fukushima (activities  
133 ranging between 2 – 2400 Bq kg<sup>-1</sup>) compared to those in  $^{134}\text{Cs}$  and  $^{137}\text{Cs}$  (500 – 1,245,000 Bq  
134 kg<sup>-1</sup>) required relatively long counting times and the use of low-background high-resolution  
135 detectors to allow its detection. This probably explains why this radioisotope has not been

136 measured more widely during the months that followed the accident. We therefore decided  
137 to restrict this study to the main contamination plume (Table 2) in order to minimize  
138 uncertainties associated with  $^{110m}\text{Ag}$  measurements.

### 139 **A. Behaviour of $^{110m}\text{Ag}$ in soils**

#### 140 *Experiment 1 – Affinity with different grain size fractions*

141 Four soil samples (~300g/sample – Soil 1 to Soil 4 - Fig. 3) were collected using non-metallic  
142 trowels in different parts of the contamination plume where  $^{110m}\text{Ag}$ : $^{137}\text{Cs}$  activity ratio varies  
143 from 0.002 to 0.008. After drying at 40°C, samples were dry-sieved to 1mm, 500µm, 250µm  
144 and 63µm using an automatic device, in order to measure the respective activities present in  
145 the different grain size fractions.

#### 146 *Experiment 2 – Migration in soil*

147 To investigate the potential migration of radionuclides with depth in soil, gamma  
148 measurements were conducted on each 2-mm increment of the uppermost 2-cm of a soil  
149 profile sampled near Kawamata city (Profile - Fig. 3). Soil layers were collected using a  
150 scraper plate composed by a metal frame and a metal plate (Loughran et al., 2002; Kato et  
151 al., 2012b). To avoid contamination by top layers, a spray glue was used to fix the sample.

152  $^{137}\text{Cs}$  concentration in undisturbed Japanese soils is expected to show an exponential decline  
153 with depth (Kato et al., 2012b; Koarashi et al., 2012; Matsunaga et al., 2013). Cumulative  
154 inventory can be expressed as described in Eq. (1):

$$155 \quad I_{(x)} = I_t (1 - \exp^{-x/h_0}) \quad (1)$$

156 Where  $I(x)$  is the radiocesium inventory ( $\text{Bq}\cdot\text{m}^{-2}$ ) at the  $x$  ( $\text{kg}\cdot\text{m}^{-2}$ ) depth,  $I_t$  is the total  
157 radiocesium inventory and  $h_0$  is the relaxation mass depth ( $\text{kg}\cdot\text{m}^2$ ), an index characterising  
158 the radiocesium penetration in the soil. In order to use this formula, we calculated the  $^{137}\text{Cs}$   
159 inventory using  $^{137}\text{Cs}$  activities ( $\text{Bq}\cdot\text{kg}^{-1}$ ) (Lee et al., 2013) at the successive ( $x$ ) depths (Eq. 2):

$$160 \quad I(x) = \rho(x) * h(x) * A(x) \quad (2)$$

161 Where  $\rho$  is the dry bulk density ( $\text{g}\cdot\text{m}^{-3}$ ),  $h$  is the thickness (cm) of the layer and  $A$  the activity  
162 ( $\text{Bq}\cdot\text{kg}^{-1}$ ) of this layer.

### 163 **B. Behaviour of $^{110\text{m}}\text{Ag}$ in rivers**

#### 164 *Experiment 3 – Activities in sediment sequences accumulated in reservoirs*

165 During the November 2012 fieldwork campaign, we had the opportunity to collect samples  
166 of the different layers representative of the deep sediment sequence that accumulated  
167 behind Tetsuzen dam on Ota River (1.6 m) and behind Takanokura dam on Mizunashi River  
168 (Nitta catchment) (0.5 m) in order to investigate  $^{110\text{m}}\text{Ag}$  behaviour in respectively 9 and 7  
169 different layers (Dam Sed 1 and Dam Sed 2 - Fig. 3). We removed the exposed sediment  
170 lateral surface to avoid artificial sample contamination during fieldwork.

171 Particle size of the samples was measured using a SALD-3100 Laser (Shimadzu Co., Ltd.,  
172 Kyoto, Japan) following standard procedures. The particles were classified into 42 size ranges  
173 between 0.05 and 450 $\mu\text{m}$ . The surface specific area was estimated using the density of  
174 quartz ( $2.65\text{g cm}^{-3}$ ) and assuming that particles were spherical (Santamarina and Klein,  
175 2002). Grain size distribution was corrected using sieving data for classes  $>450\mu\text{m}$ , and final  
176 classes were reclassified according to the grain-size ranges proposed by AFNOR X 316107  
177 (i.e., clay  $<2\mu\text{m}$ ,  $2\mu\text{m} < \text{silt} < 50\mu\text{m}$ ,  $50\mu\text{m} < \text{sand} < 2\text{mm}$ ).

178 *Experiment 4 – Impact of typhoon and snowmelt on activities in river drape sediment*

179 A last experiment was conducted in upper parts of the Nitta River catchment (with  
180 elevation > 400m), where the  $^{110m}\text{Ag}:$  $^{137}\text{Cs}$  activity ratio in soils is low but overall radioactive  
181 contamination is high. This area was impacted by summer typhoons and spring snowmelt  
182 events that generated soil erosion on hillslopes and led to an increase in river discharge  
183 (Fukuyama et al., 2005; Iida et al., 2012; Ueda et al., 2013). Sediment drape deposits (SDD)  
184 were selected as an alternative to suspended sediment in order to increase the spatial  
185 coverage of the survey within the catchments, and to avoid the logistical problems  
186 associated with the collection of suspended sediment in rivers. Olley et al. (2013)  
187 demonstrated that sediment source apportionment conducted on SDD and suspended  
188 matters showed similar results. To prepare a representative sample, five to ten subsamples  
189 that are likely to have deposited after the last major flood were collected at several locations  
190 selected randomly down to the underlying coarser cobble or gravel layer across a 10m<sup>2</sup>  
191 surface. Sediment drape deposits are likely to integrate deposits of the last hydro-  
192 sedimentary events of low to intermediate magnitude (Evrard et al., 2011) even though the  
193 occurrence of large floods probably leads to a complete flush of the system justifying the  
194 relevance of conducting field surveys to collect sediment drape deposits as frequently as  
195 possible.

196 Activities were measured in 4 representative SDD samples collected at the same location  
197 during the four successive fieldwork campaigns (SDD 1 to SDD 4 - Fig. 3). Grain size  
198 measurements were conducted by following the same protocol as described for Experiment  
199 3.

200 2.3 Gamma spectrometry measurements

201 Before measurement, samples were dried at 40°C (105°C for soil), ground to a fine powder  
202 and then packed into 15mL polyethylene specimen cups. Radionuclide activities ( $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ,  
203  $^{110\text{m}}\text{Ag}$ ) in all samples were determined by gamma spectrometry using very low-background  
204 coaxial N- and P-types HPGe detectors (Canberra / Ortec). Counting times of soil and  
205 sediment samples varied between  $8\times 10^4$  and  $200\times 10^4$  s to allow the detection of  $^{110\text{m}}\text{Ag}$ ,  
206 which was present in much lower activities in the samples (2–2400 Bq.kg<sup>-1</sup>) than  $^{134}\text{Cs}$  and  
207  $^{137}\text{Cs}$  (500–1,245,000 Bq.kg<sup>-1</sup>).

208 The  $^{137}\text{Cs}$  activities were measured at the 661 keV emission peak. The  $^{134}\text{Cs}$  ( $T_{1/2} = 2\text{y}$ )  
209 activities were calculated as the mean of activities derived from measurements conducted at  
210 604 keV and 795 keV ( $^{228}\text{Ac}$  activities being negligible compared to  $^{134}\text{Cs}$  activities) as both  
211 peaks are associated with the largest gamma emission intensities of this radionuclide.

212 The presence of  $^{110\text{m}}\text{Ag}$  was confirmed by the detection of an emission peak at 885 keV (as  
213 the peak associated with the largest emission intensity at 658 keV was masked by the 661-  
214 keV peak of  $^{137}\text{Cs}$ ). Minimum detectable activities in  $^{110\text{m}}\text{Ag}$  for 24h counting times reached 2  
215 Bq.kg<sup>-1</sup>. Detection of  $^{110\text{m}}\text{Ag}$  was considered relevant when the 885 keV peak was present.

216 Because of their low level, activities in  $^{110\text{m}}\text{Ag}$  were associated with larger uncertainties than  
217 activities in  $^{137}\text{Cs}$ . Counting efficiencies and quality assurance were conducted using internal  
218 and certified International Atomic Energy Agency (IAEA) reference materials prepared in the  
219 same specimen cups as the samples. Uncertainties on results were estimated by combining  
220 counting statistics and calibration uncertainties. Summing and self-absorption effects were  
221 taken into account by analysing standards with similar densities and characteristics as the  
222 collected samples. All radionuclide activities were decay corrected to the date of 14 March  
223 2011 corresponding to the date of the first radionuclide deposits on soils. Most of them

224 were estimated to have deposited on 15 March in Fukushima Prefecture (Kinoshita et al.,  
225 2011; Shozugawa et al., 2012).

### 226 **3. Results and discussion**

#### 227 **A. Behaviour of $^{110m}\text{Ag}$ in soils**

##### 228 **Experiment 1 – Affinity with different grain size fractions (Soil 1 to Soil 4)**

229 During this experiment,  $^{110m}\text{Ag}$  activities could only be detected in all fractions for 2 of the 4  
230 analysed samples (Soil 1 and Soil 2 – Fig. 3) as a result of low  $T_{1/2}$  and low  $^{110m}\text{Ag}$  initial  
231 deposition level. Samples were indeed collected in the vicinity of the main contamination  
232 plume as access to the most affected area was restricted by Japanese authorities. It also has  
233 been detected in the finest fraction of the Soil 4 sample. Fig. 4 describes the distribution of  
234  $^{110m}\text{Ag}$  and  $^{137}\text{Cs}$  radionuclides in the different grain size fractions of Soil 1 and Soil 2 samples.  
235 Activity distribution is similar for both radionuclides and the major part (from 33% to 65%) of  
236 their activity was measured in the finest fraction (<63 $\mu\text{m}$ ; Table 3). We could therefore not  
237 detect any difference in the particle-size effects of adsorption between both radionuclides.  
238 This finding is crucial in order to use the  $^{110m}\text{Ag}:^{137}\text{Cs}$  activity ratio to track the dispersion of  
239 contaminated particles in catchments, as the finest particles are the most susceptible to be  
240 eroded and redistributed (Motha et al., 2002).

##### 241 **Experiment 2 – Migration in soil (Profile)**

242 Activities measured in the uppermost part of the soil profile (Fig. 3) confirmed the low  
243 mobility of  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$  and  $^{110m}\text{Ag}$  (Table 4). The bulk of their inventories ( $\approx 80\%$ ) were  
244 contained in the first layers (0 – 8 mm) and decreased rapidly with depth (Fig. 5).

245 The  $^{137}\text{Cs}$  total inventory reached about  $417 \text{ kBq}\cdot\text{m}^{-2}$  and the relaxation mass depth  $h_0$  was  
246  $7.1 \text{ kg}\cdot\text{m}^{-2}$  for  $^{137}\text{Cs}$  (Fig. 6). This value is close to the one found by Kato et al. (2012b) in a  
247 cultivated soil ( $9.1 \text{ kg}\cdot\text{m}^{-2}$ ) in the vicinity of our sampling site. This value also remained in the  
248 same order of magnitude as in several cultivated soils investigated by Ivanov (Ivanov et al.,  
249 1997) near the Chernobyl Power Plant (ranging from  $5.6$  to  $9.1 \text{ kg}\cdot\text{m}^{-2}$ ).

250 In order to compare the behaviour of both radionuclides, we calculated in the same way the  
251 relaxation mass depth for the  $^{110\text{m}}\text{Ag}$  and found  $7.6 \text{ kg}\cdot\text{m}^{-2}$  with a total  $^{110\text{m}}\text{Ag}$  inventory value  
252 of  $1 \text{ kBq}\cdot\text{m}^{-2}$ , showing that penetration of both radionuclides in the soil was similar.

253 Our results are consistent with the ones provided by other studies that investigated the  
254 behaviour of the same radionuclides in soils. Alloway (1995) showed that  $^{110\text{m}}\text{Ag}$  is not  
255 mobile as it is the case for  $^{137}\text{Cs}$  (Sawi-iney, 1972; Spezzano, 2005) and they both remain in  
256 the uppermost 5cm of the soil (Handl et al., 2000; Kato et al., 2012b; Shang and Leung, 2003)

257 We investigated the migration of radionuclides in a profile of bare soil, but the transposition  
258 of those results to soils covered with vegetation is not straightforward, as the radionuclide  
259 interception and migration processes were reported to play a major role in forested  
260 environments (Kato et al., 2012a). Furthermore, Martin et al. (1989) reported that during the  
261 first months that followed Chernobyl fallout,  $^{110\text{m}}\text{Ag}$  required more time to migrate from  
262 grass to soil (50% reduction) than  $^{137}\text{Cs}$  (90% reduction) due to foliar leaching. However, this  
263 effect should have remained limited in Fukushima, as most cultivated soils (i.e., paddy fields)  
264 were found to be bare when atmospheric fallout occurred in March 2011.

265 These results confirm the low mobility of  $^{110m}\text{Ag}$  in cultivated soils. As it remained  
266 concentrated in the first layers, it could be rapidly redistributed across hillslopes as a  
267 consequence of runoff and soil erosion.

### 268 C. Behaviour of $^{110m}\text{Ag}$ in rivers

#### 269 Experiment 3 – Activities in sediment sequences accumulated in reservoirs (Dam Sed 1 & 2)

270 The pH measured in the four investigated rivers varied between 5.0 and 6.0. It was  
271 demonstrated in the literature that pH plays a major role on  $^{110m}\text{Ag}$  behaviour in both soil  
272 and water. Its mobility in soils (Khan et al., 1982; Shang and Leung, 2003) increases when pH  
273 raises and its affinity for suspended particulate matter (SPM) rises when pH exceeds 7, to  
274 reach maximum values at a pH of 9 (Fukai and Murray, 1974; Murray and Murray, 1972). In  
275 contrast,  $^{137}\text{Cs}$  was shown not to be affected by a pH variation (Adam et al., 2001). Because  
276 the observed pH values remained significantly lower than 7,  $^{110m}\text{Ag}$  behaviour and  
277  $^{110m}\text{Ag}:$  $^{137}\text{Cs}$  activity ratio were not expected to be affected by pH variations in Fukushima  
278 rivers.

279 Analysis of the two sediment profiles (Dam Sed 1 and 2 – Fig. 3) tends to confirm the similar  
280 post-depositional behaviour of the radionuclides (Figs. 7 and 8). Their activities were  
281 concentrated in the top of the profile (Tables 5 and 6), and most likely resulted from  
282 deposition of sediment eroded after the initial radionuclide fallout in March 2011. Before  
283 the FDNPP accident,  $^{137}\text{Cs}$  activities in sediment were generally lower than 100Bq/kg in  
284 Japanese soils (Fukuyama et al., 2005), which corresponds to activities found in deeper  
285 layers of sediment accumulated behind Tetsuzen dam (Dam Sed 1). Higher contamination in  
286 lower layers in the second profile (> 500Bq/kg) most likely resulted from contamination

287 originating from upper levels during sampling (scraping). However, this will not impact the  
288 conclusions drawn from this experiment as they strictly focus on the observations made on  
289 the top layers.

290 In Tetsuzen dam sediment (Dam Sed 1), a lower activity was measured in the surface layer  
291 than in the underlying layer suggesting that a minimum of two significant erosion events had  
292 supplied contaminated sediment to the reservoir by November 2012 (Fig. 7; Table 5). Also,  
293  $^{110m}\text{Ag}/^{137}\text{Cs}$  activity ratio remained constant for these layers. Furthermore, a significant  
294 difference in particle size between the first two layers and the third layer confirms the  
295 occurrence of an intense erosion event, probably during the dam release that was carried  
296 out during the period of heavy typhoons that took place during summer in 2011. Based on  
297 this information, sedimentation rates were estimated to  $20\text{cm yr}^{-1}$  in Tetsuzen dam, and to  
298  $5\text{cm yr}^{-1}$  in Takanokura dam.

299

300 The similar percentage of radionuclide inventories found in the successive layers and the  
301 calculation of constant  $^{110m}\text{Ag}:^{137}\text{Cs}$  activity ratios confirmed that  $^{110m}\text{Ag}$  and  $^{137}\text{Cs}$  have a  
302 similar behaviour with regard to their adsorption onto soil particles and sediment. This result  
303 was expected owing to the high distribution coefficients reported in the literature for both  
304 Cs and Ag (Ciffroy et al., 2001; Fournier-Bidoz and Garnier-Laplace, 1994).  $K_d$  values  
305 remaining in the same order of magnitude were estimated for both radionuclides in winter  
306 and summer (i.e.  $K_{d\text{Cs-}^{137}} : 0.1-100$ ;  $K_{d\text{Ag-}^{110m}} : 1-100$ ). Ciffroy et al. (2001) also quantified the  
307 kinetics of the adsorption and desorption of these radionuclides in freshwater systems, and  
308 concluded to their similar behaviour. Fournier-Bidoz and Garnier-Laplace (1994) and Garnier  
309 et al. (2006) showed that  $^{110m}\text{Ag}$  has a strong affinity for river SPM and especially for their

310 clay fraction. A large number of studies demonstrated that Cs has the same behaviour  
311 (Ojima et al. , 1965 in Ancelin et al., 1979; Garnier-Laplace et al., 1994). He and Walling  
312 (1996) confirmed that  $^{137}\text{Cs}$  affinity increases with specific surface area in the case of a  
313 uniform contamination of the soils. Guéguéniat et al. (1976) reported similar results for  
314  $^{110\text{m}}\text{Ag}$ .

#### 315 **Experiment 4 – Impact of typhoon and snowmelt**

316 During the three successive sampling campaigns that followed the succession of snowmelt  
317 and typhoon events, the inventories of  $^{110\text{m}}\text{Ag}$  and  $^{137}\text{Cs}$  measured in sediment drape  
318 deposits collected systematically at four sampling points in the upper part of the Nitta  
319 catchment (Fig. 3) showed similar patterns (Fig. 9). As expected, samples collected close to  
320 the catchment headwaters (SDD 1 and SDD 4) showed a decrease in contamination  
321 throughout time, probably due to the flush of the most contaminated material during high  
322 water flows (Table 7). In contrast, the samples collected at the 2 downstream locations  
323 (especially SDD 3) were associated with a slight temporal increase in contamination of  $^{137}\text{Cs}$   
324 that may be explained by a supply of upstream – more contaminated – material. This  
325 explanation is consistent with the fact that  $^{110\text{m}}\text{Ag}:$  $^{137}\text{Cs}$  activity ratio measured in sediment  
326 remained in the same order of magnitude as the ratio measured in soils of this upper part of  
327 the Nitta catchment (Table 8).

328 An alternative explanation to the activity increase observed throughout time is that the grain  
329 size of particles could have changed from one campaign to the next, explaining the observed  
330 differences in contamination. Calculation of surface specific area (SSA) shown in Fig. 9 shows  
331 that this might be the case for sample SDD 3 (with an enrichment in fine particles  
332 throughout time), but not for sample SDD 2 (SSA remaining constant in time).

333 Because of different experimental conditions (non-homogeneous fallout deposition and high  
334 radionuclide activities), our results plotted on Fig. 10 do not show a power function as found  
335 by He and Walling (1996) but they display instead a linear function for both  $^{137}\text{Cs}$  ( $r^2 = 0.89$ )  
336 and  $^{110\text{m}}\text{Ag}$  ( $r^2 = 0.91$ ) that is strongly correlated with the SSA.

337 This tends to confirm the similar behaviour for both radionuclides, although particle-size  
338 effects and fractionation could have occurred, given that erosion and transport in rivers are  
339 obviously selective to finer particles. Consequently, these results confirmed the relevance of  
340 using of  $^{110\text{m}}\text{Ag}:^{137}\text{Cs}$  activity ratio to investigate the dispersion and redistribution of  
341 contamination along rivers.

## 342 **Conclusions**

343 Silver-110m ( $^{110\text{m}}\text{Ag}$ ) is an anthropogenic radionuclide that is produced continuously by  
344 nuclear power plants (NPP) in normal conditions, but investigations regarding its behaviour  
345 in the environment are rare despite the potentially important radioecological implications of  
346 its accidental release into the environment. In the Fukushima NPP post-accidental context,  
347 the interest to conduct field studies on the behaviour of  $^{110\text{m}}\text{Ag}$  was reinforced by the fact  
348 that initial deposits displayed  $^{110\text{m}}\text{Ag}:^{137}\text{Cs}$  activity ratios that varied across space within the  
349 main radioactive contamination plume. This study showed that  $^{137}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  were  
350 strongly sorbed by the finest particle fractions (i.e., clay or silt), and that the bulk of their  
351 inventory was stored in the uppermost part of soil profiles (i.e., <2 cm), confirming that the  
352 in-depth mobility was not significant during the two years following the accident. In addition  
353 to this fine particle reactivity, we showed that soil erosion and sediment transport in  
354 catchments that mobilize preferentially the finest particle fractions did not induce different  
355 fractionation effects for both radionuclides. This study therefore confirmed the relevance of

356 using this ratio to track the dispersion of contaminated material along coastal rivers draining  
357 this area. In future, due to the rapid decay of radiosilver and its initially relatively low fallout  
358 level, extraction methods could be used in order to measure precisely radiosilver and  
359 continue to investigate its behaviour and the dispersion of contaminated sediment along  
360 rivers.

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367

368

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527

528

529 **Figure captions**

530 Figure 1. Number of publications per year between 1937 and 2013 based on a search in the  
531 Web of Knowledge citation index, using “radiosilver” (or “Ag-110m” » or “<sup>110m</sup>Ag”) and  
532 “environment” as topic keywords (22 July 2013).

533 Figure 2. Location of the main radiocaesium (<sup>134</sup>Cs + <sup>137</sup>Cs) contamination plume in  
534 Fukushima Prefecture, northeastern Japan (derived from MEXT data decay-corrected to June  
535 2011).

536 Figure 3. Location of the samples collected across the main contamination plume of  
537 Fukushima Prefecture. Background map corresponds to <sup>110m</sup>Ag:<sup>137</sup>Cs activity ratio based on  
538 data measured in Fukushima Prefecture by MEXT in June 2011 (activities decay-corrected to  
539 14 June 2011). SDD: sediment drape deposits, Dam Sed 1: sediment collected in Tetsuzen  
540 dam, Dam Sed 2: sediment collected in Takanokura dam; FDNPP: Fukushima Dai-ichi Nuclear  
541 Power Plant.

542 Figure 4. Percentage of total activity in <sup>110m</sup>Ag and <sup>137</sup>Cs in four particle size classes (d =  
543 diameter) for two soil samples (Soil 1 and Soil 2 - see the location of sampling sites on Figure  
544 3).

545 Figure 5. Radionuclide inventory (%) in successive 2-mm increment layers along a bare soil  
546 profile (Profile - see the location of sampling sites on Figure 3).

547 Figure 6. Distribution of <sup>137</sup>Cs inventory (%) in the bare soil profile as measured by gamma  
548 spectrometry and fitted using Eq. (1), as a function of mass depth ( $\text{kg}\cdot\text{m}^{-2}$ ) (Profile - see the  
549 location of sampling sites on Figure 3).

550 Figure 7. Sediment profile accumulated in Yokokawa reservoir on the Ota River (Dam Sed 1  
551 – see the location of sampling sites on Figure 3) – a) Picture of the profile: the limits of the  
552 numbered sampled layers are represented with horizontal lines - b) Anthropogenic  
553 radionuclide inventories (%) in the successive layers of the profile (mm) – c) Grain size  
554 composition of the different layers.

555 Figure 8. Sediment profile accumulated behind Takanokura dam on the Mizunashi River  
556 (Nitta catchment – Dam Sed 2 – see the location of sampling sites on Figure 3) – a) Picture of  
557 the profile: the limits of the numbered sampled layers are represented with horizontal lines -  
558 b) Activity in anthropogenic radionuclides in the successive layers of the profile – c) Grain  
559 size composition of the different layers.

560 Figure 9. Evolution of the  $^{137}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  contamination inventory (%) and specific surface  
561 area (SSA) ( $\text{m}^2.\text{g}^{-1}$ ) of sediment drape deposits collected in the upper part of the Nitta River  
562 catchment during the campaigns of Nov 2011, April 2012 and Nov 2012 (SDD 1 to SDD 4 –  
563 see the location of sampling sites on Figure 3).

564 Figure 10. Relationships between  $^{137}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  activities ( $\text{Bq}.\text{kg}^{-1}$ ) and specific surface  
565 area ( $\text{m}^2.\text{g}^{-1}$ ) in sediment drape deposits collected in the upper part of the Nitta River  
566 catchment during the campaigns of Nov 2011, April 2012 and Nov 2012 (SDD 1 to SDD 4 –  
567 see the location of sampling sites on Figure 3).

Figure 1.

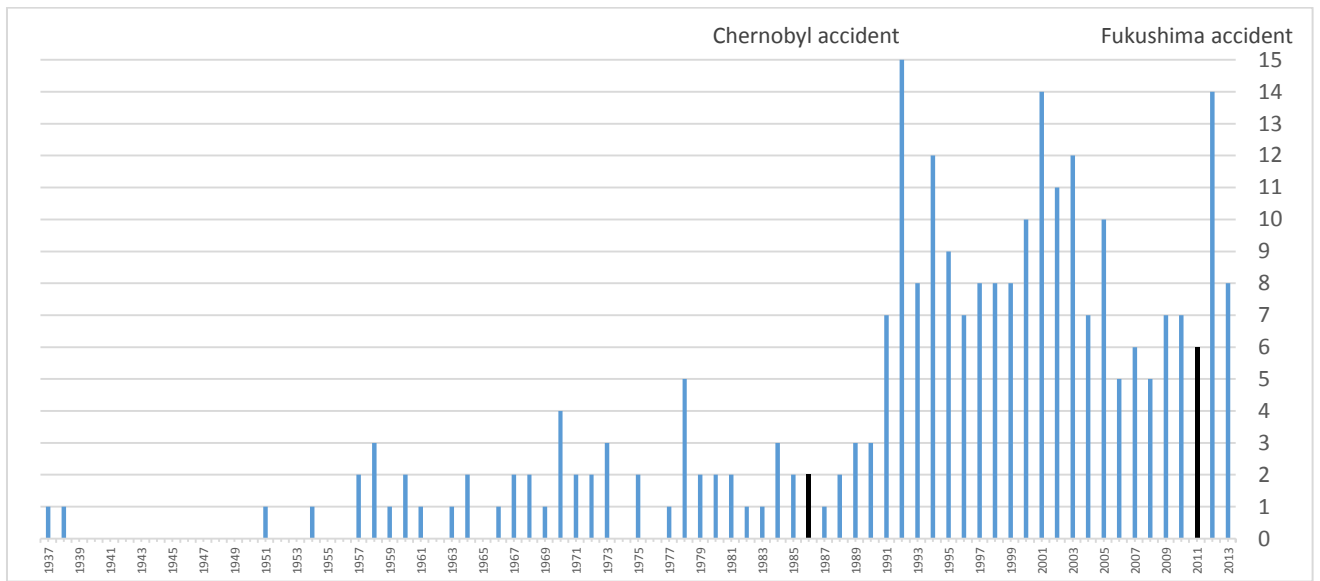


Figure 2.

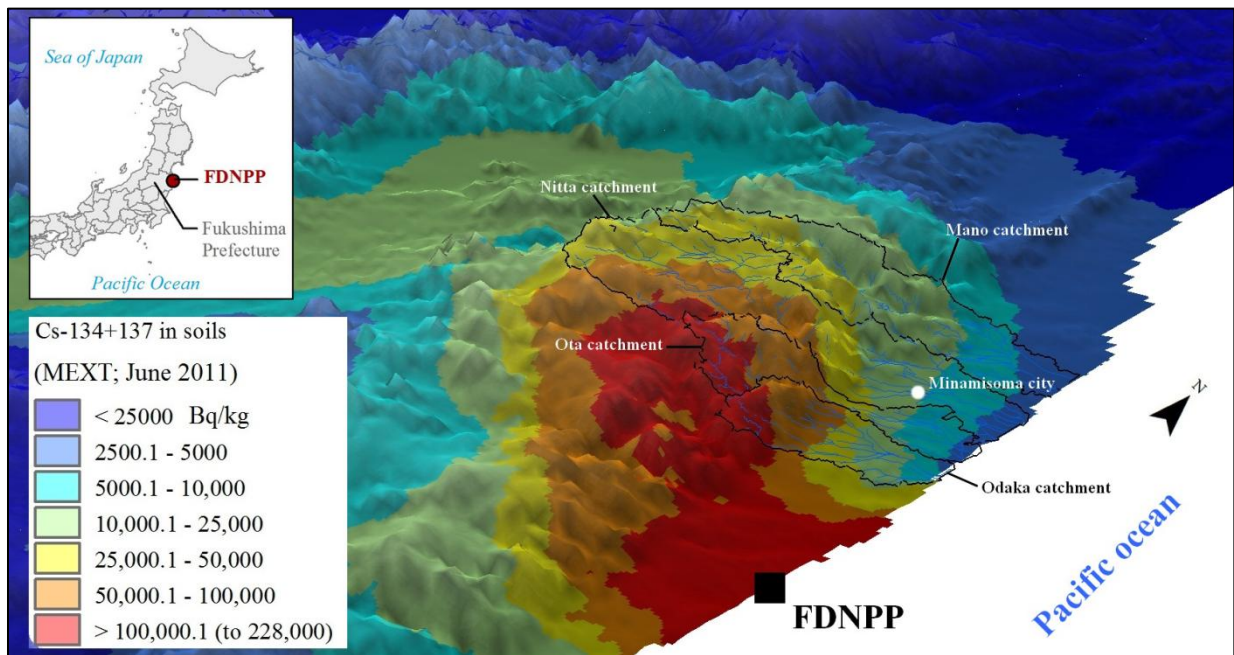


Figure 3.

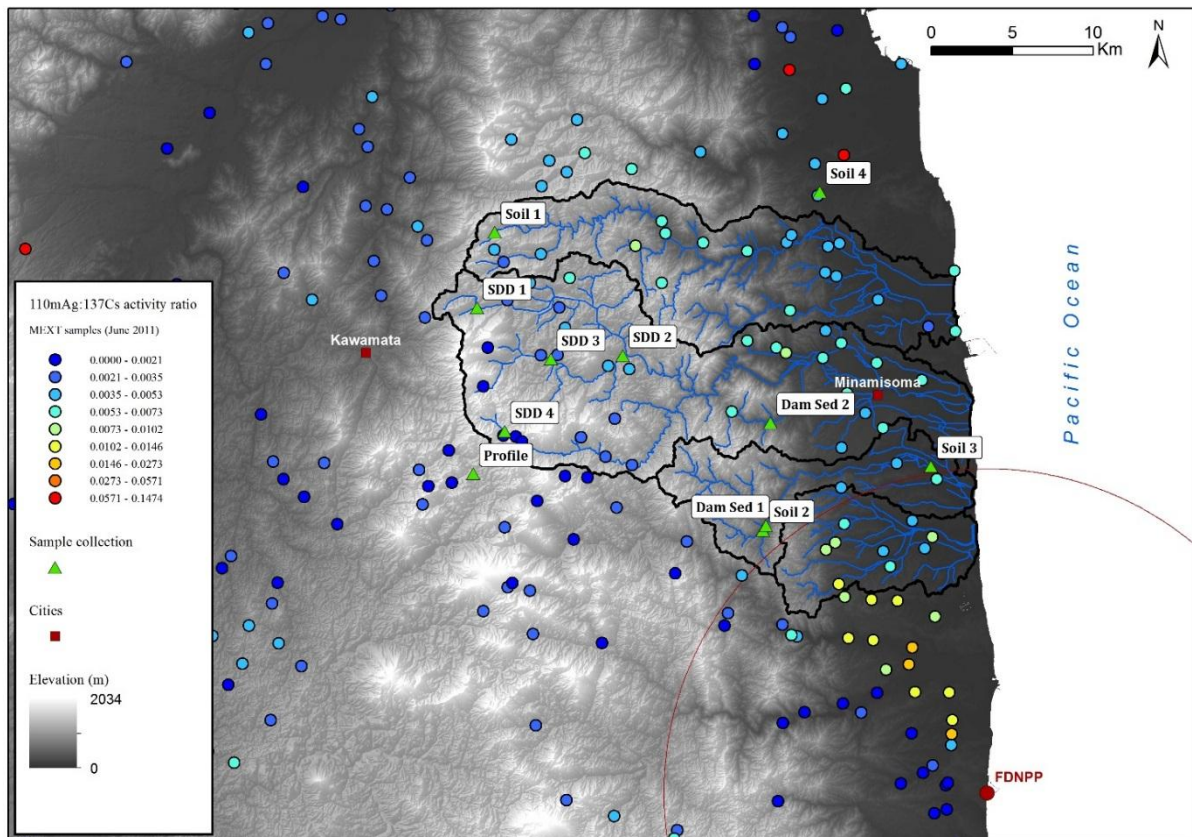


Figure 4.

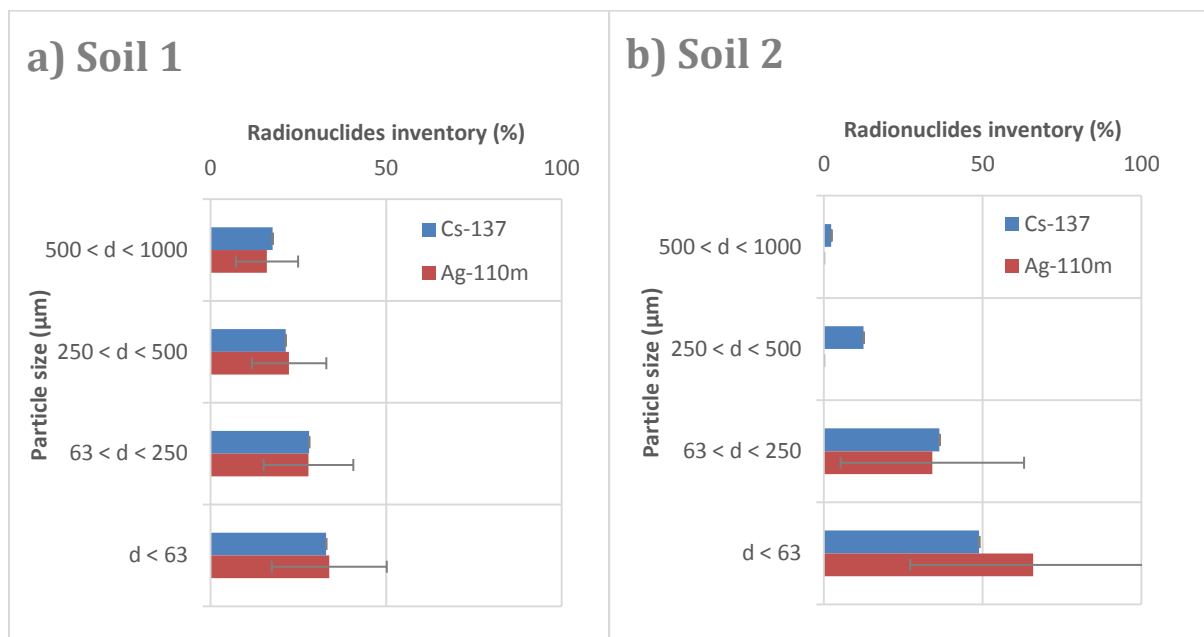


Figure 5.

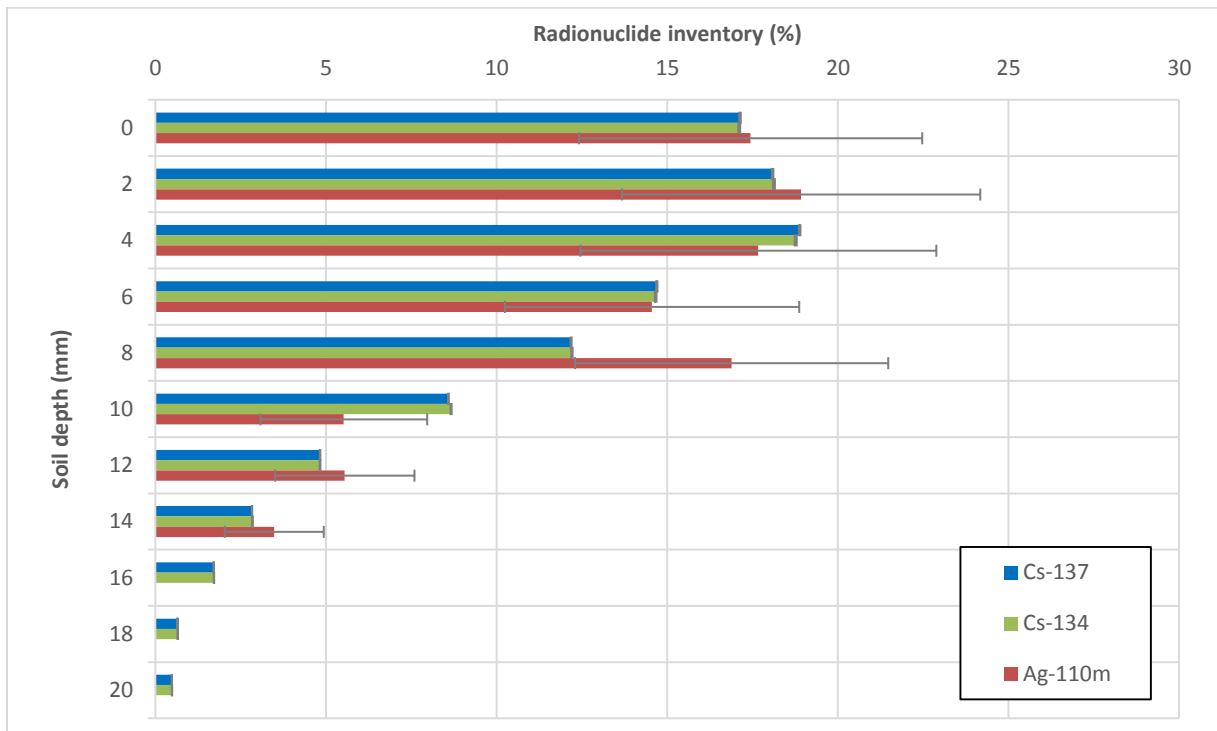


Figure 6.

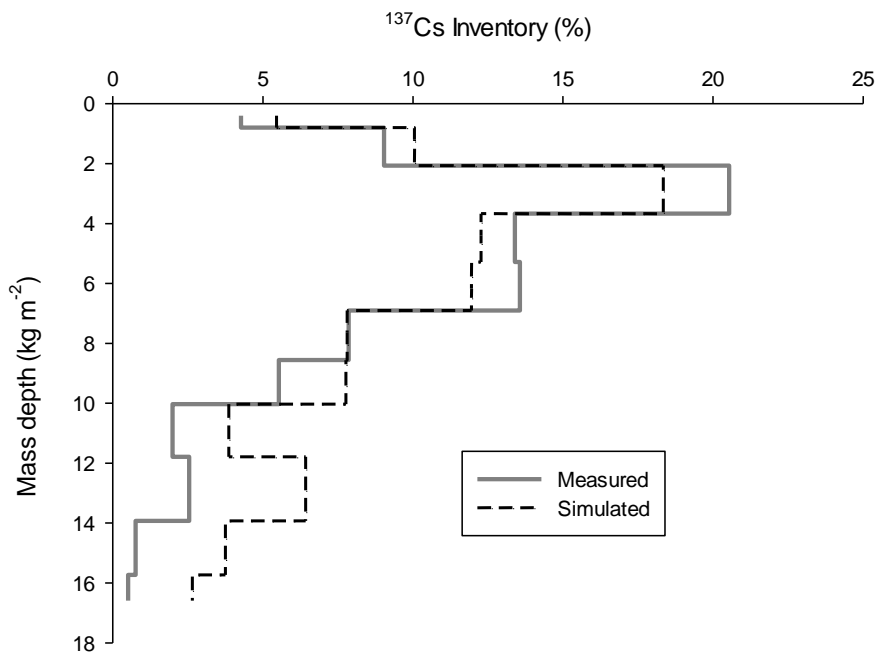


Figure 7.

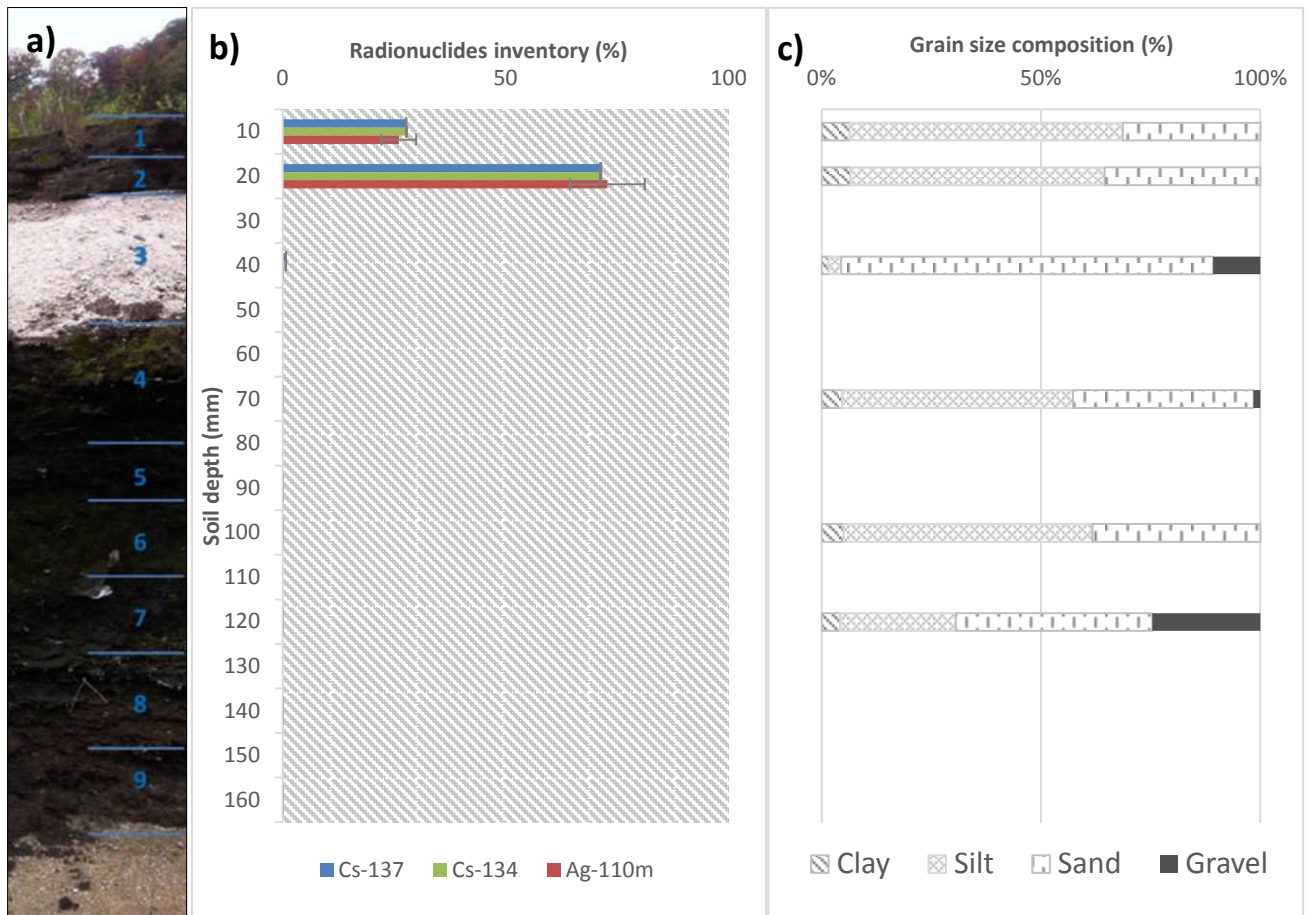


Figure 8.

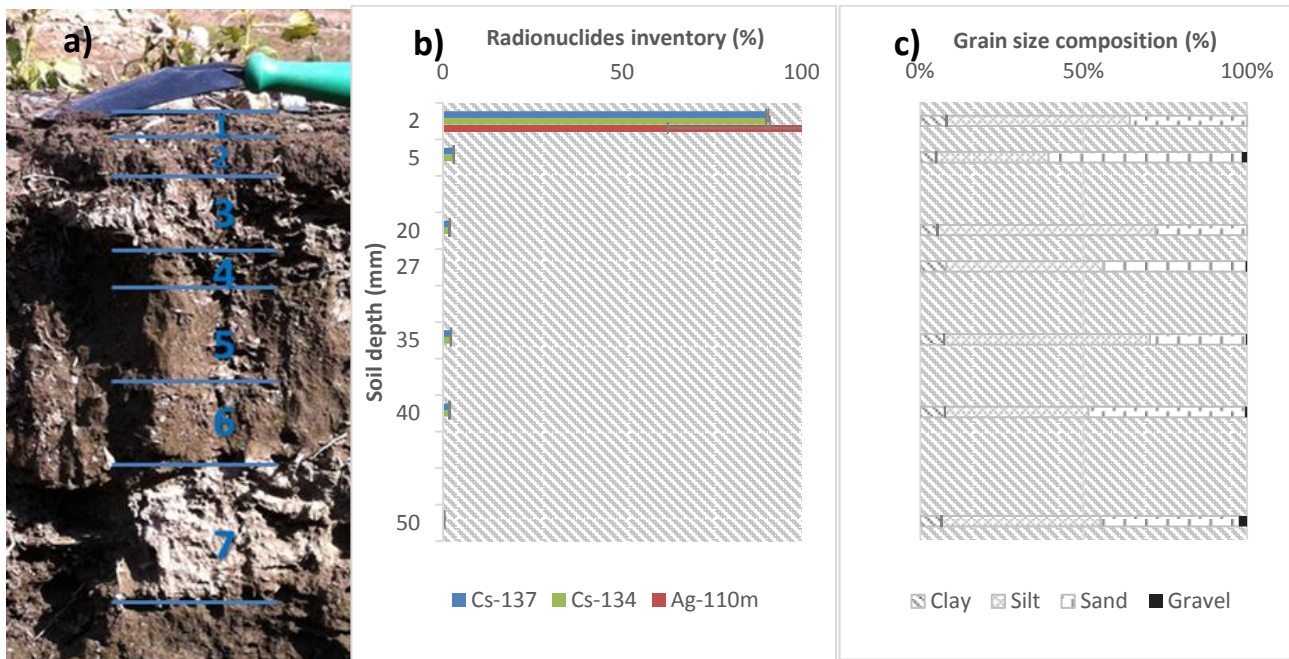


Figure 9.

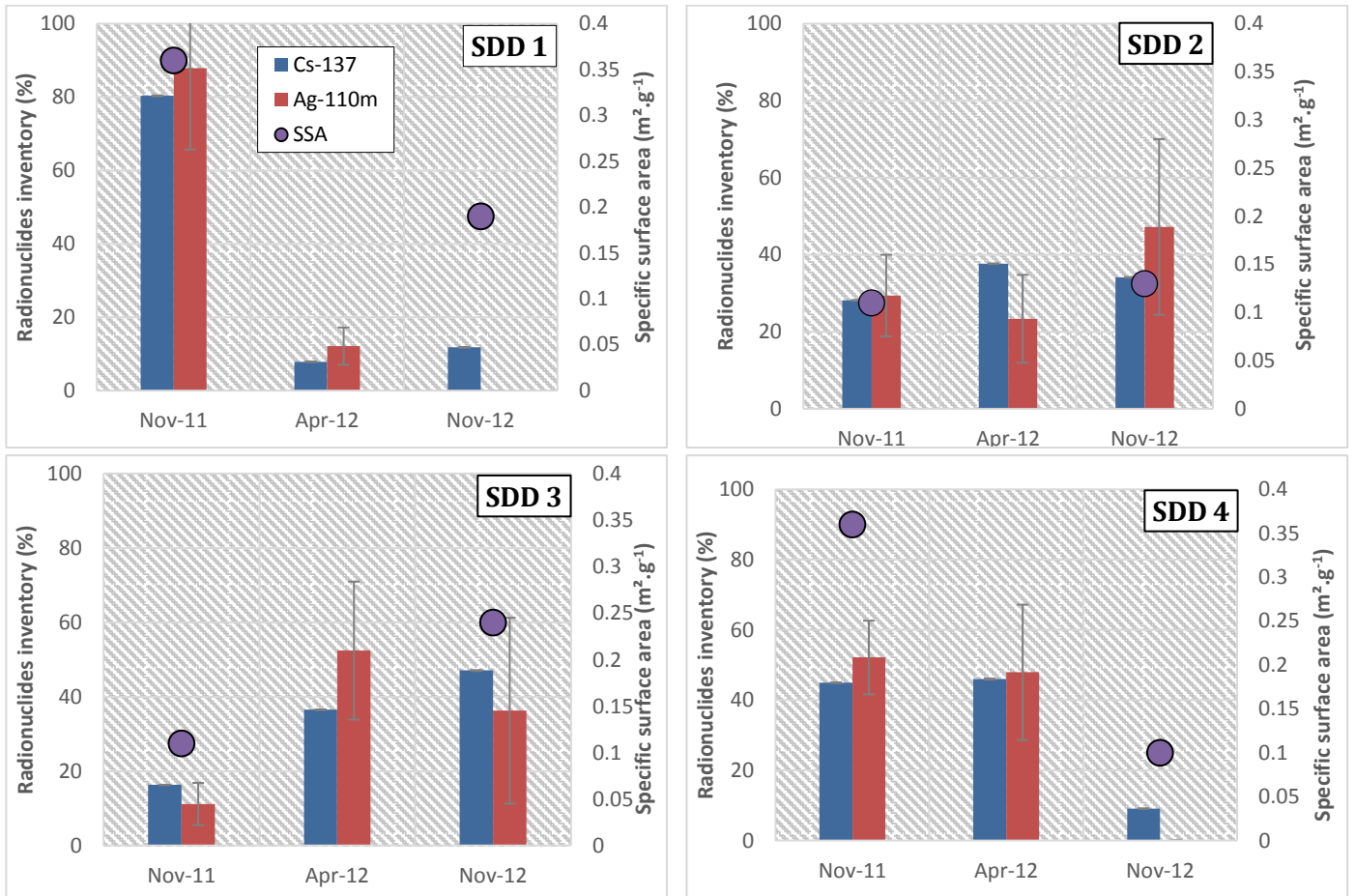
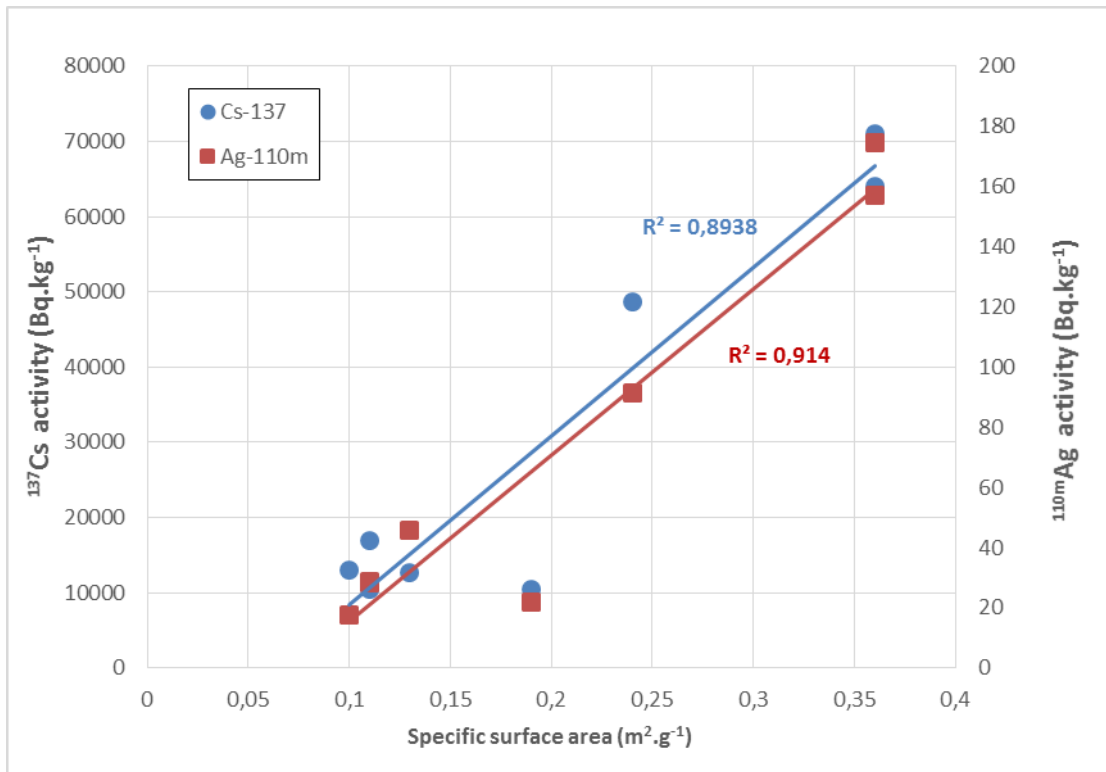


Figure 10



**Table 1**

Literature compilation of radiosilver activities measured in samples collected in the environment in normal and post-accidental conditions.

	Normal conditions				Post-accidental conditions			
	Sample description	Mean (number of samples)	Max	Source	Samples description	Mean (number of samples)	Max	Source
SPM (Bq.kg <sub>dry</sub> <sup>-1</sup> )	Routine monitoring in France (from 2009 to 2012)	<2 (893)	28	(T.Boissieux, IRSN/LS3E, personal communication)	Not investigated to our knowledge			
	Rhone River, France (from 2005 to 2008)	1 (≈10)	<10	Eyrolle et al. (2012)				
Sediment (Bq.kg <sub>dry</sub> <sup>-1</sup> )	Routine monitoring in France (from 2009 to 2012) (Bottom sediment)	<1 (279)	<2.75	(T.Boissieux, IRSN/LS3E, personal communication)	In Fukushima Prefecture (14 March 2011) (Sediment drape deposits)	2 – 2390 (162)	2390	Chartin et al. (2013)
Soil (Bq.kg <sub>dry</sub> <sup>-1</sup> )	Not reported in the literature				In Fukushima Prefecture (11 March 2011)	280 (7)	520	Tazoe et al. (2012)
					In Miyagi Prefecture (26 April 2011)	12 (15)	49	Wanatabe et al. (2012)

SPM : Suspended Particulate Matters

LS3E: Laboratoire de Surveillance de l'Environnement et d'Expertise par Echantillonnage

FDNPP : Fukushima Dai-ichi Nuclear Power Plant

**Table 2.**

Date of collection and location of the samples collected in the framework of this study.

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Sample	Date of collection	(WGS 1984):	Latitude	Longitude	Experiment
Soil 1	09/11/2012		37.732447	140.688330	1
Soil 2	10/11/2012		37.567039	140.877110	1
Soil 3	10/11/2012		37.602735	140.994663	1
Soil 4	10/11/2012		37.755403	140.916649	1
Profile	04/2012		37.598136	140.673917	2
Dam Sed 1 (Tetsuzen)	10/11/2012		37.569687	140.879102	3
Dam Sed 2 (Takanokura)	08/11/2012		37.626889	140.882307	3
SDD 1	See Table 7		37.690509	140.676251	4
SDD 2	See Table 7		37.664095	140.778413	4
SDD 3	See Table 7		37.662099	140.728151	4
SDD 4	See Table 7		37.621797	140.695852	4

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SDD: Sediment Drape Deposit

**Table 3.**

Distribution of radionuclide activities ( $\text{Bq.kg}^{-1}$ ) and weight in four grain size fractions of five soils collected across Fukushima radioactive pollution plume. Total activity ( $\text{Bq.kg}^{-1}$ ) is also given for the bulk sample.

Sample	500 $\mu\text{m}$ < d < 1000 $\mu\text{m}$			250 $\mu\text{m}$ < d < 500 $\mu\text{m}$			63 $\mu\text{m}$ < d < 250 $\mu\text{m}$			d < 63 $\mu\text{m}$		Bulk sample activity		
	Weight (%)	$^{137}\text{Cs}$	$^{110\text{m}}\text{Ag}$	Weight (%)	$^{137}\text{Cs}$	$^{110\text{m}}\text{Ag}$	Weight (%)	$^{137}\text{Cs}$	$^{110\text{m}}\text{Ag}$	Weight (%)	$^{137}\text{Cs}$	$^{110\text{m}}\text{Ag}$	$^{137}\text{Cs}$	$^{110\text{m}}\text{Ag}$
Soil 1	8*	21 166 $\pm$ 50**	83 $\pm$ 36	16	25 636 $\pm$ 56	116 $\pm$ 40	42	33 682 $\pm$ 64	145 $\pm$ 44	32	39 490 $\pm$ 90	175 $\pm$ 62	n/a	n/a
Soil 2	11	929 $\pm$ 14	< 14	14	5009 $\pm$ 32	< 26	27	14 600 $\pm$ 80	93 $\pm$ 64	29	19 608 $\pm$ 66	179 $\pm$ 54	9912 $\pm$ 64	98 $\pm$ 52
Soil 3	8	298 $\pm$ 8	< 10	14	425 $\pm$ 14	< 10	35	446 $\pm$ 10	< 12	39	580 $\pm$ 16	< 8	506 $\pm$ 12	< 12
Soil 4	17	576 $\pm$ 12	< 14	12	787 $\pm$ 14	< 16	34	665 $\pm$ 12	< 14	37	1484 $\pm$ 26	27 $\pm$ 25	634 $\pm$ 26	< 22

d: diameter

n/a: not available

\* The remainder of the sample mass correspond to the gravel fraction (>1000  $\mu\text{m}$ )

\*\* Uncertainties on results were estimated by combining counting statistics and calibration uncertainties. Summing and self-absorption effects were taken into account by analysing standards with similar densities and characteristics as the collected samples.

**Table 4.**

Depth distribution of  $^{137}\text{Cs}$  and  $^{110\text{m}}\text{Ag}$  concentrations in the soil profile.

Depth (mm)	$^{137}\text{Cs}$ (Bq.kg <sup>-1</sup> )	$^{110\text{m}}\text{Ag}$ (Bq.kg <sup>-1</sup> )	Dry bulk density (g.cm <sup>-3</sup> )	Mass depth (kg.m <sup>-2</sup> )
0 – 2	44 698 ± 74	100 ± 34	0.19	0.4
2 – 4	47 189 ± 72	109 ± 34	0.40	1.2
4 – 6	49 254 ± 76	101 ± 38	0.87	2.9
6 – 8	38 335 ± 64	84 ± 32	0.73	4.4
8 – 10	31 776 ± 58	97 ± 28	0.89	6.2
10 – 12	22 393 ± 54	32 ± 24	0.73	7.6
12 – 14	12 575 ± 38	32 ± 18	0.92	9.5
14 – 16	7374 ± 28	20 ± 14	0.56	10.6
16 – 18	4439 ± 22	< 12	1.19	13.0
18 – 20	1674 ± 14	< 16	0.94	14.9
20 - 22	1241 ± 22	< 16	0.86	16.6

**Table 5.**

Depth, radionuclide activities and grain size proportions of the sequence of sediment accumulated behind Tetsuzen Dam (Ota River)

Depth (cm)	Radionuclide activities and activity ratio			Grain size proportions			
	$^{137}\text{Cs}$ (Bq.kg <sup>-1</sup> )	$^{110\text{m}}\text{Ag}$ (Bq.kg <sup>-1</sup> )	$^{110\text{m}}\text{Ag}:^{137}\text{Cs}$	Clay (%)	Silt (%)	Sand (%)	Gravel (%)
0 - 10	64 195 ± 210	649 ± 142	0.010 ± 0.002	6	62	31	0
10 - 20	165 217 ± 280	1813 ± 196	0.011 ± 0.001	7	58	35	0
20 - 40	1790 ± 24	< 20	n/a	2	3	85	11
40 - 70	146 ± 8	< 18	n/a	5	53	41	2
70 - 85	68 ± 8	<22	n/a	n/a	n/a	n/a	n/a
85 - 100	33 ± 8	< 40	n/a	5	57	38	0
100 - 110	147 ± 20	< 68	n/a	4	26	45	25
110 – 140	26 ± 4	< 14	n/a	n/a	n/a	n/a	n/a
140 – 162	77 ± 10	< 18	n/a	n/a	n/a	n/a	n/a

n/a: not available

**Table 6.**

Depth, radionuclide activities and grain size proportions of the sequence of sediment accumulated behind Takanokura Dam (Nitta River).

Depth (cm)	Radionuclide activities (Bq.kg <sup>-1</sup> )		Activity ratio	Grain size proportions			
	<sup>137</sup> Cs	<sup>110m</sup> Ag	<sup>110m</sup> Ag: <sup>137</sup> Cs	Clay (%)	Silt (%)	Sand (%)	Gravel (%)
0 - 2	28 744 ± 120	246 ± 88	0.0085 ± 0.0030	8	56	36	0
2 - 5	967 ± 30	< 32	n/a	5	34	59	2
5 - 20	616 ± 18	< 18	n/a	5	67	28	0
20 - 27	28 ± 4	< 12	n/a	8	48	44	0
27 - 35	728 ± 16	< 18	n/a	7	63	29	0
35 - 40	592 ± 10	< 12	n/a	8	44	48	1
40 - 50	124 ± 6	< 16	n/a	7	49	42	2

n/a: not available

**Table 7.**

Characteristics of the riverbed sediment from the upper part of the Nitta River catchment collected at the same locations during the three fieldwork campaigns.

Sample	Sampling date	<sup>137</sup> Cs activity (Bq.kg <sup>-1</sup> )	<sup>110m</sup> Ag activity (Bq.kg <sup>-1</sup> )	<sup>110m</sup> Ag: <sup>137</sup> Cs	SSA (m <sup>2</sup> .g <sup>-1</sup> )	pH
SDD 1	Nov-2011	71 014 ± 108	157 ± 30	0.0022 ± 0.0004	0.36	n/a
	Apr-2012	6 939 ± 34	22 ± 16	0.0031 ± 0.0023	n/a	n/a
	Nov-2012	10 394 ± 62	< 38	n/a	0.19	6 ± 0.5
SDD 2	Nov-2011	10 385 ± 32	29 ± 10	0.0028 ± 0.0010	0.11	n/a
	Apr-2012	13 885 ± 34	23 ± 18	0.0016 ± 0.0013	n/a	n/a
	Nov-2012	12 583 ± 48	46 ± 34	0.0036 ± 0.0027	0.13	6 ± 0.5
SDD 3	Nov-2011	16 898 ± 66	29 ± 22	0.0017 ± 0.0013	0.11	n/a
	Apr-2012	337 786 ± 72	132 ± 34	0.0035 ± 0.0009	n/a	n/a
	Nov-2012	48 680 ± 178	92 ± 55	0.0019 ± 0.0011	0.24	5.5 ± 0.5
SDD 4	Nov-2011	64 093 ± 100	175 ± 16	0.0027 ± 0.0002	0.36	n/a
	Apr-2012	65 599 ± 106	161 ± 24	0.0024 ± 0.0004	n/a	n/a
	Nov-2012	12 949 ± 74	< 44	n/a	0.1	6

n/a: not available

**Table 8**

Comparison of  $^{110m}\text{Ag}$ : $^{137}\text{Cs}$  activity ratio between soil and sediment drupe deposits collected during the three campaigns in the upper part of the Nitta River catchment. All values were decay-corrected to 14 march 2011.

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$^{110m}\text{Ag}$ : $^{137}\text{Cs}$ activity ratio	Soil (based on MEXT data)	SDD
Nov-11	$0.0023 \pm 0.008$	$0.0023 \pm 0.0005$
Apr-12		$0.0027 \pm 0.0008$
Nov-12		$0.0022 \pm 0.0010$

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SDD: Sediment Drupe Deposits