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Evidence of the radioactive fallout in France due to the Fukushima nuclear accident

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Keywords: Fukushima accident; radioactive fallout; radioiodine; radiocesium; France.

Highlights

- Artificial radionuclides emitted by Fukushima power plant accident were detected in environmental samples collected in France
- Radioactivity measurements on air filters obtained at the highest altitude in France
- ¹³¹I detected in grass, soil and sediment collected between 30 March – 12 April
- ¹³⁴Cs detected in grass collected between 31 March – 10 April
- Total ¹³⁴Cs input to grass of 2 Bq m⁻² (~400 times lower than Chernobyl input)

25 **Abstract**

26 Radioactive fallout due to the Fukushima reactor explosion in Japan was detected in
27 environmental samples collected in France. The presence of ^{131}I in aerosols ($200\pm 6 \mu\text{Bq m}^{-3}$)
28 collected at the *Pic du Midi* observatory, located at 2877 m altitude in the French Pyrénées,
29 indicated that the Japanese radioactive cloud reached France between 22 and 29 March, i.e.
30 less than two weeks after the initial emissions, as suggested by a $^{137}\text{Cs}/^{134}\text{Cs}$ ratio of 1.4.
31 Cesium radioisotopes (^{134}Cs and ^{137}Cs) were not detected in this sample but they were present
32 in the aerosol sample collected the next week, i.e. between 29 March and 05 April (about 10
33 $\mu\text{Bq m}^{-3}$). We also report ^{131}I activities measured in grass ($1.1\text{--}11 \text{ Bq kg}^{-1}$; fresh weight) and
34 soil samples (0.4 Bq kg^{-1}) collected in the Seine River basin between 30 March and 10 April.
35 The ^{134}Cs from the damaged Fukushima power plant was also detected in grass collected in
36 the Seine River basin between 31 March and 10 April ($0.2\text{--}1.6 \text{ Bq kg}^{-1}$ fresh weight, with a
37 $^{137}\text{Cs}/^{134}\text{Cs}$ ratio close to 1, which is consistent with Fukushima radioactive release). Despite
38 the installation of a network of nested stations to collect suspended matter in the upstream part
39 of the Seine River basin, ^{131}I was only detected in suspended matter ($4.5\text{--}60 \text{ Bq kg}^{-1}$)
40 collected at the most upstream stations between 30 March and 12 April. Neither ^{131}I nor ^{134}Cs
41 have been detected in environmental samples since the end of April 2011, because of the
42 rapid decay of ^{131}I and the very low activities of ^{134}Cs (about 400 times lower than after
43 Chernobyl accident).

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49 1. Introduction

50 Tohoku earthquake that occurred on 11 March 2011 was exceptionally powerful
51 (moment magnitude M_w of 9.0; Simons et al., 2011). It triggered a violent tsunami with an
52 exceptional wave, which height reached up to 20 m or even 40 m locally. The coincidence
53 between this earthquake and the tsunami led to the destruction of more than 600 km of
54 Japanese coastline, to numerous deaths and injuries (about 20,000 people were killed or are
55 still missing), and to the immersion of the land up to 10 km inland (Imamura, 2011). Reactors
56 of the affected nuclear power plants were automatically stopped, but the loss of cooling
57 systems was observed at three reactors of Fukushima-Daiichi power plant. People were
58 evacuated within a 20-km radius around the power plant because of the possible occurrence of
59 an explosion and radioactive fallout (Irikura, 2011). Several explosions occurred during the
60 next few days (i.e., 12-15 March). The Nuclear and Industrial Safety Agency of Japan (NISA)
61 attributed a 7-level to this accident according to INES – International Nuclear and
62 Radiological Event Scale (IAEA, 2011). A similar 7-level was also attributed to the
63 Chernobyl accident that occurred on 26 April 1986 in Ukraine. A cloud loaded with
64 radioactive elements was emitted from the damaged reactors, blown out eastward of Japan to
65 the Pacific Ocean and then detected in parts of the Northern hemisphere.

66 The U.S. Environmental Protection Agency reported the presence of 18 fission
67 product radionuclides (e.g., ^{137}Cs , ^{131}I) in air filter samples collected on 22 March 2011 (EPA,
68 2011). The radioactive cloud then moved over the USA, the Atlantic Ocean and then diffused
69 across Europe. Airborne radionuclides are continuously monitored by a European network of
70 measurement stations (150 locations). Based on this monitoring, Masson et al. (2011)
71 provided maps describing the transport and dispersion in the air of Fukushima radionuclides
72 over Europe.

73 Additional radionuclide fallout was reported from melted snow and rainwater samples
74 collected across the European continent (e.g., from Greece, Manolopoulou et al., 2011; from
75 Central Russia, Bolsunovsky and Dementyev, 2011). In contrast and to our knowledge, there
76 are only few studies reporting fallout radionuclide activity measurements in soil, river
77 sediment and grass samples collected in Europe after the Fukushima accident. Pittauerova et
78 al. (2011) reported ^{131}I , ^{134}Cs and ^{137}Cs activities determined in a total of twelve rainwater,

79 river sediment, soil, grass and cow milk samples collected in Northwest Germany. Beresford
80 et al. (in press) provided measurements of ^{131}I originating from Fukushima in grass samples
81 collected across Great Britain. In France, one study investigated the transfer of ^{131}I from
82 Fukushima to vegetation and milk, but it only focused on study sites located in southern
83 France (Parache et al., 2011).

84 This paper compiles and provides additional radionuclide data. The aim of the present
85 study was to determine whether we could find in France any evidence of radioactive fallout
86 associated with the Fukushima accident. Therefore, we analyzed environmental samples
87 collected in France, such as grass, soil and suspended sediment of the Seine River basin to
88 check whether radioisotopes from Fukushima could be detected. To complement this study
89 conducted at low altitude, we also investigated aerosol samples collected at high altitude
90 (2877) in the French Pyrénées. Very few samples were analyzed at such an altitude in Europe.

91 **2. Materials and methods**

92 ***2.1 Collection of aerosols at the Pic du Midi mountain (southern France)***

93 *2.1.1 Sampling*

94 Aerosols were collected using a pumping system linked up to the Whole Air Inlet
95 (WAI) at the *Pic du Midi* observatory located at 2877 m above sea level in the French
96 Pyrénées; Fig. 1a) in the framework of the ORAURE network (*Observations en Réseaux des*
97 *Aérosols à Usage de Recherches Environnementales*). A volume of 400 m³ of air was filtered
98 using Teflon filters (Pall Zefluor, 47 mm diameter, 2.0 μm porosity) during one-week periods.
99 The air flow rate in the WAI system was fixed to capture all aerosols with a diameter lower
100 than 30 μm. We followed strict quality-controlled procedures to avoid any contamination of
101 the samples.

102 *2.1.2 Gamma spectrometry measurements*

103 Radioisotope activities (i.e., ^{131}I , ^{137}Cs and ^{134}Cs as well as ^7Be) on the filters were
104 determined using low-background gamma-ray spectrometry at the LAFARA underground
105 laboratory located in Ferrières, French Pyrénées (van Beek et al., 2010; van Beek et al.,
106 submitted). Two germanium detectors are protected from cosmic radiations under 85 m of
107 rock (215 m water equivalent). A very low background (<5 counts per minute) is thus
108 achieved in this laboratory, allowing the detection of low-gamma activities in environmental

109 samples (van Beek et al., submitted). The filters were analyzed using a high-purity
110 germanium coaxial detector (GEM-FX series) provided by ORTEC/AMETEK (van Beek et
111 al., in press). The germanium crystal has a diameter of 85 mm and a length of 32.2 mm, which
112 is equivalent to a volume of 183 cm³ of germanium. The spectrum of the detector covers a
113 wide range of energies, ranging from 10 to 2700 keV. The detector resolution (FWHM: full
114 width at half maximum) determined at 122 keV and 1.33 MeV are 0.669 keV and 1.65 keV,
115 respectively. The detector is protected by a lead castle: 2 cm of low-activity lead (< 10 Bq kg⁻¹)
116 and additionally 13 cm regular lead (<50 Bq kg⁻¹). Finally, 4 cm of pure copper cover the
117 interior of the lead castle.

118 We used RGU-1 and IAEA Standard #175 powder standards provided by IAEA (International
119 Atomic Energy Agency) that we placed onto a 47 mm filter to determine the detection
120 efficiencies for the radionuclides reported in this study. RGU-1 is a certified reference
121 material prepared using U ore diluted with floated silica powder - with all the daughters of the
122 series being in secular equilibrium. IAEA Standard #175 allows us to determine the detection
123 efficiencies for ¹³⁷Cs and ⁴⁰K. The detection efficiencies for ¹³¹I, ¹³⁴Cs and ⁷Be were then
124 interpolated at the corresponding energies, 365, 604 and 478 keV, respectively. For the short-
125 lived ¹³¹I and ⁷Be, decay correction was applied according to the middle of the sampling
126 period (one week interval).

127

128 ***2.2 Collection of soil, sediment and grass samples in the Seine River basin (northern*** 129 ***France)***

130 ***2.2.1 Sampling***

131 Soil, suspended sediment and grass samples were collected within the upstream part of
132 the Seine River basin (Fig. 1a). The location of sampling points is shown in Fig. 1b. We
133 collected soil samples in cropland fields that are potential sediment source areas. Five
134 subsamples of surface material (top 2 cm) were collected and well-mixed to provide a
135 homogeneous sample. Additionnally, suspended sediment samples were collected in upstream
136 Orgeval and Saulx Rivers (i.e., upstream tributaries of the Seine River) at four different nested

137 stations (i.e., M elarchez, Grange Allard, Ribeaucourt, Vitry-le-Fran ois) between 16 March
138 and 14 June 2011. We used metallic cylindrical traps submerged into the water by using a rope
139 attached to a nearby tree trunk on the river bank. Finally, twelve fresh grass samples were
140 collected from uncut parklands or gardens in both Orgeval and Saulx subcatchments as well
141 as in the vicinity of our laboratory (i.e. LSCE) between 30 March and 14 June 2011.

142 *2.2.2 Gamma spectrometry measurements*

143 Fresh grass samples were weighed and placed into 1-L Marinelli plastic containers.
144 They were analysed as soon as possible after sampling. All results were decay corrected to the
145 day of sampling. Soil and suspended sediment samples were sieved to 2 mm, dried and
146 packed into 15 or 60-ml (depending on the quantity available) pre-tared polyethylene
147 specimen cups and sealed airtight to contain ^{222}Rn and allow ingrowth of its decay products.
148 The ^{131}I (364.5 keV), ^7Be (477.6 keV), ^{137}Cs (661.6 keV), and ^{210}Pb (46.5 keV) activities in
149 this material were determined by gamma spectrometry using the seven very low-background
150 coaxial N- and P-types GeHP detectors (Canberra / Ortec) available at the LSCE. For soil and
151 suspended sediment samples, “excess” ^{210}Pb was calculated by subtracting the supported
152 activity (determined using two ^{238}U daughters, i.e. ^{214}Pb , by taking the average count number
153 at 295.2 and 351.9 keV, and ^{214}Bi at 609.3 keV) from the total activity of ^{210}Pb (measured at
154 46.5 keV; Le Cloarec et al., 2011).

155 When the available material was insufficient (< 5 g), suspended sediment samples
156 were placed in polypropylene tubes, sealed airtight, and counted on a well-type Ge detector.
157 Counting time of soil and suspended sediment samples varied between 8×10^4 and 3×10^5 s.
158 All measured counts were corrected for background levels measured at least every 2 months
159 as well as for detector and geometry efficiencies. Results were systematically expressed in Bq
160 kg^{-1} . Counting efficiencies and quality insurance were conducted using internal and certified

161 IAEA standards (e.g., IAEA-135, IAEA-375, IAEA-CU-2006-03, IAEA-Soil-6, RGU-1,
162 RGTh-1) prepared in the same specimen cups as the samples. Efficiencies were interpolated
163 for ^7Be energy.

164 Furthermore, to provide an indication on the “age” of suspended sediment collected in
165 the river traps, we calculated the $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ ratio, as proposed by Matisoff et al. (2005). A
166 decrease in the $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ ratio can then alternatively be explained by (i) an increase in the
167 sediment residence time (given that ^7Be decay is faster than $^{210}\text{Pb}_{\text{xs}}$ decay) or by (ii) the
168 dilution of sediment enriched in ^7Be by sediment poorer in ^7Be . Consequently, calculation of
169 this ratio could indicate whether aging and/or dilution of sediment occurred in the river
170 (Evrard et al., 2010).

171 **3. Results and discussion**

172 ***3.1 Aerosols collected on top of Pic du Midi (2877 m, southern France)***

173 Radionuclide activities reported in this study are integrated over one-week intervals
174 (Table 1). Data reported for the air filter samples collected at the *Pic du Midi* observatory
175 constitute, to our knowledge, the data obtained at the highest altitude in France. These data
176 provide thus a good complement to the measurements obtained in lowlands and at lower
177 altitudes by the French Institute of Radioprotection and Nuclear Safety (IRSN, 2011). Only
178 two samples (#123 and #124) were analyzed sufficiently quickly after the date of collection to
179 allow detection of ^{131}I (half life of 8.02 days). All other samples were analyzed several weeks
180 after sampling, which prevented the detection of potential ^{131}I activities on the filters. Only
181 ^{137}Cs , ^{134}Cs and ^7Be activities could be determined for these samples. We measured a ^{131}I
182 activity of $200 \pm 6 \mu\text{Bq m}^{-3}$ on the filter that collected aerosols between 22 March and 29
183 March 2011. This activity indicates the presence of the radioactive cloud above France during

184 this period, because ^{131}I activities measured in France are typically $< 0.03 \mu\text{Bq m}^{-3}$ (Loaiza et
185 al., submitted). However, our sample integrates a one-week interval and we cannot determine
186 the exact day during which the radioactive cloud reached France. The ^{131}I activity reported
187 here shows a good agreement with the activity determined on filters collected in the French
188 Alps (Loaiza et al., submitted). These authors collected several filters during the same time
189 interval (25-29 March) and the resulting mean ^{131}I activity varied between $248 \pm 2 \mu\text{Bq m}^{-3}$
190 (25–27 March) and $144 \pm 5 \mu\text{Bq m}^{-3}$ (29 March). Higher ^{131}I activities were reported later,
191 amounting up to $522 \mu\text{Bq m}^{-3}$ (02-03 April; Loaiza et al., submitted). These data can also be
192 compared to another altitude station, Jungfrauoch station (Switzerland, 3450 m) where ^{131}I
193 activity levels were found to reach up to $900 \mu\text{Bq m}^{-3}$ (Masson et al., 2011).

194 ^{134}Cs and ^{137}Cs were not detected in this aerosol sample collected at the end of March
195 (when ^{131}I was detected) but they were present in the aerosol sample collected the following
196 week, i.e. between 29 March and 05 April (sample #125). We report ^{137}Cs and ^{134}Cs activities
197 of 10 ± 2 and $7 \pm 2 \mu\text{Bq m}^{-3}$, respectively. These activities are in the same order of magnitude as
198 ^{137}Cs (mean of $17.5 \mu\text{Bq m}^{-3}$) and ^{134}Cs (mean of $19.5 \mu\text{Bq m}^{-3}$) activities determined on
199 filters collected in the French Alps during the same period (Loaiza et al., submitted). Only one
200 sample (#125) allows us to provide a value of 1.4 for the $^{137}\text{Cs}/^{134}\text{Cs}$ ratio in aerosols. As a
201 comparison, the $^{137}\text{Cs}/^{134}\text{Cs}$ ratio in the Fukushima fallout was found to be close to 1.0,
202 whereas it was in the range of 0.5–0.6 in the Chernobyl fallout in 1986 (Masson et al., 2011).
203 The $^{137}\text{Cs}/^{134}\text{Cs}$ ratio itself is not conclusive enough in order to determine the origin of the
204 radioactive contamination. However, the presence of a significant ^{134}Cs activity suggests a
205 recent input into the atmosphere, and it is very probably associated with Fukushima
206 radioactive release. Finally, we also report ^7Be activities on the filters (^7Be being a natural
207 cosmogenic radioisotope which was not associated with the radioactive cloud). The ^7Be

208 activities that we determined also remained in the same order of magnitude as the activities
209 reported during the same time interval by Loaiza et al. (submitted), thereby confirming the
210 relevance of our values.

211

212 ***3.2 Samples collected in the Seine River basin (Northern France)***

213 Overall, the spring of 2011 was exceptionally dry in France. Extremely low rainfall
214 (total amount < 5 mm) was recorded in Paris at the end of March and early in April. Then, the
215 weather remained almost completely dry until early June (Fig. 2). Masson et al. (2011)
216 showed that airborne concentrations of ^{131}I have increased in western Europe until 28–30
217 March. Then, a second peak of similar magnitude was observed between 3–5 April. Both
218 fallout periods coincided with periods of low rainfall (≤ 5 mm) in the Paris region (Fig. 2).

219 ***3.2.1 Grass samples***

220 Presence of radioiodine (^{131}I) was detected in grass between 30 March and 10 April.
221 The highest ^{131}I activity (11 ± 1 Bq kg^{-1} ; fresh weight – FW) was found in the sample collected
222 in Palaiseau on 5 April. This observation is consistent with the measurements conducted in
223 Great Britain, where Beresford et al. (in press) reported peaks of ^{131}I activities in grass
224 samples collected between 4–6 April 2011. Radioiodine activities measured in grass samples
225 from the Parisian Basin remained in the activity range reported by IRSN for samples collected
226 across France (i.e., 0.5–15 Bq kg^{-1} FW; IRSN, 2011), but they were generally higher than the
227 activities measured on grass collected in southern France (0.1– 4 Bq kg^{-1} FW, with the
228 exception of one grass sample collected in Agen showing an activity of 9 Bq kg^{-1} FW;
229 Parache et al., 2011).

230 Presence of ^{134}Cs (0.2–1.6 Bq kg⁻¹ FW) and ^{137}Cs (0.9–1.6 Bq kg⁻¹ FW) was only
231 detected in the grass samples collected between 31 March and 10 April. This result is
232 consistent with the observation that ^{134}Cs and ^{137}Cs were detected later than ^{131}I , because they
233 exceeded the detection criterion later than radioiodine (Masson et al., 2011). The activities
234 measured in the Seine River basin remained in the same order of magnitude but they were
235 slightly higher than those reported for three grass samples collected in Northwestern Germany
236 (0.1–0.3 Bq kg⁻¹ FW of ^{134}Cs and 0.2–1.6 Bq kg⁻¹ FW of ^{137}Cs ; Pittauerová et al., 2011).

237 Values of the $^{131}\text{I}/^{137}\text{Cs}$ ratio varied within a range of 1.7 ± 0.4 – 9.0 ± 3.8 , and were
238 maximal between 31 March and 5 April. These values are similar to the ones observed in
239 Northwestern Germany during the same period (Pittauerová et al., 2011). Similar values were
240 obtained for $^{131}\text{I}/^{134}\text{Cs}$ ratio. The $^{137}\text{Cs}/^{134}\text{Cs}$ ratio varied indeed between 0.7 ± 0.4 and 5.0 ± 3.0 .
241 Masson et al. (2011) showed that values of this ratio in the Fukushima fallout were close to 1,
242 whereas it was in the range 0.5–0.6 in the Chernobyl fallout in 1986. Furthermore, it must be
243 noted that both ^{134}Cs and ^{137}Cs from previous sources (i.e., global fallout from nuclear
244 weapon tests; Chernobyl fallout) are normally not detected anymore in grass samples in this
245 region, as indicated by the activities below detection limits obtained before 31 March and
246 after 10 April (Table 2). These observations therefore indicate that ^{134}Cs and ^{137}Cs detected in
247 grass of the Parisian Basin were very likely released by the Fukushima accident.

248 Fresh grass density in the field (about 1.2 kg m⁻²) was calculated in the vicinity of
249 LSCE laboratory in order to determine the inventory of ^{134}Cs in the soils. This inventory
250 remained very low (about 2 Bq m⁻²) by 10 April 2011. This inventory is about three orders of
251 magnitude smaller than the ^{137}Cs inventories in soils of the region (about 1500–2000 Bq m⁻²
252 by 2011; recalculated after Sogon et al., 1999). This estimation is consistent with the
253 estimations made for German samples by Pittauerová et al. (2011).

254 *3.2.2 Soil and suspended sediment samples*

255 A low ^{131}I activity was detected in soils on 30 March ($0.4\pm 0.1 \text{ Bq kg}^{-1}$; Table 3).
256 Presence of ^{131}I was only detected in two suspended sediment samples collected in small
257 tributaries of the Seine River basin (between 4.5 ± 1.7 and $60\pm 30 \text{ Bq kg}^{-1}$) between 30 March
258 and 12 April (Table 3). Radioiodine was exclusively detected in upstream river reaches,
259 where sediment can be directly supplied by runoff from agricultural fields. The fine fraction
260 of sediment probably concentrated ^{131}I (having a partition coefficient or K_D of 10^4 L kg^{-1} ;
261 Fischer et al., 2009), explaining why activities could reach up to $60\pm 30 \text{ Bq kg}^{-1}$ at Grange
262 Allard station. However, the ^{131}I activity level remained very low in potential sediment
263 sources and a dilution effect probably led to the decrease of ^{131}I activities when sediment was
264 transferred downstream. Because of its rapid decay and the low level of activities measured
265 upstream, we were not able to detect this radioisotope at downstream river stations (i.e., < 50
266 Bq kg^{-1} at Ribeaucourt and Vitry-le-François stations). Values of the $^7\text{Be}/^{210}\text{Pb}_{\text{xs}}$ ratio indicate
267 that these results remained valid whatever the “age” of sediment, i.e. before and after the
268 delivery of fresh sediment to the river as reflected by the increase in this ratio after rainfall
269 (e.g., on 12 April and on 7 June at Grange Allard station).

270 The presence of ^{134}Cs could never be detected in suspended sediment throughout the
271 study period. This result was confirmed by additional analyses conducted on the very low
272 background detectors available at the *Laboratoire Souterrain de Modane* (Reyss et al., 1995).
273 In contrast, ^{137}Cs from previous sources (i.e., global fallout from nuclear weapon tests;
274 Chernobyl fallout) is continuously detected in soils and suspended sediment in this area (1.2 –
275 8 Bq kg^{-1} ; Table 2). This is reflected by the low values obtained for the $^{131}\text{I}/^{137}\text{Cs}$, showing
276 that the contribution of “old” ^{137}Cs is dominant.

277 **4. Conclusions**

278 It was possible to detect radionuclides emitted by the Fukushima power plant accident in
279 environmental samples collected in France. Radioactivity measurements conducted on
280 aerosols collected using air filters at the *Pic du Midi* observatory showed that the radioactive
281 cloud reached France between 22 – 29 March 2011 as deduced by the presence of a ^{131}I
282 activity of about $200 \pm 6 \mu\text{Bq m}^{-3}$ on the filter collected during this period. These air filter
283 samples provide, to our knowledge, the data obtained at the highest altitude in France and thus
284 complement those measured at lower altitudes and reported by other studies. The presence of
285 ^{134}Cs was also detected in air filters between 29 March and 4 April. Calculation of a
286 $^{137}\text{Cs}/^{134}\text{Cs}$ ratio close to 1 in this sample suggested that it was associated with Fukushima
287 radioactive release. The ^{134}Cs , ^{137}Cs and ^{131}I activities reported in aerosols remained in the
288 same order of magnitude as the activities determined in aerosols in the French Alps over the
289 same period. The ^{131}I activities were detected in grass, soil and river suspended sediment
290 samples collected between 30 March – 12 April in the Seine River Basin. In contrast, ^{134}Cs
291 activities were only reported in grass samples between 31 March – 10 April. Low values of
292 the $^{137}\text{Cs}/^{131}\text{I}$ ratio calculated in suspended matter outlined the contribution of “old” ^{137}Cs
293 from the global nuclear weapon tests and Chernobyl fallout in suspended sediment conveyed
294 by the rivers in the Seine River basin. A total ^{134}Cs input of 2 Bq m^{-2} was measured in grass of
295 the Parisian Basin by 10 April 2011. Overall, this fallout radionuclide input is very low. It is ~
296 400 times lower than the Chernobyl accident input in 1986 (i.e., 800 Bq m^{-2} in the Paris
297 region). ^{131}I and ^{134}Cs have not been detected in environmental samples anymore since the
298 end of April 2011, because of the rapid decay and dilution of ^{131}I and the very low ^{134}Cs
299 activities measured in France.

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305

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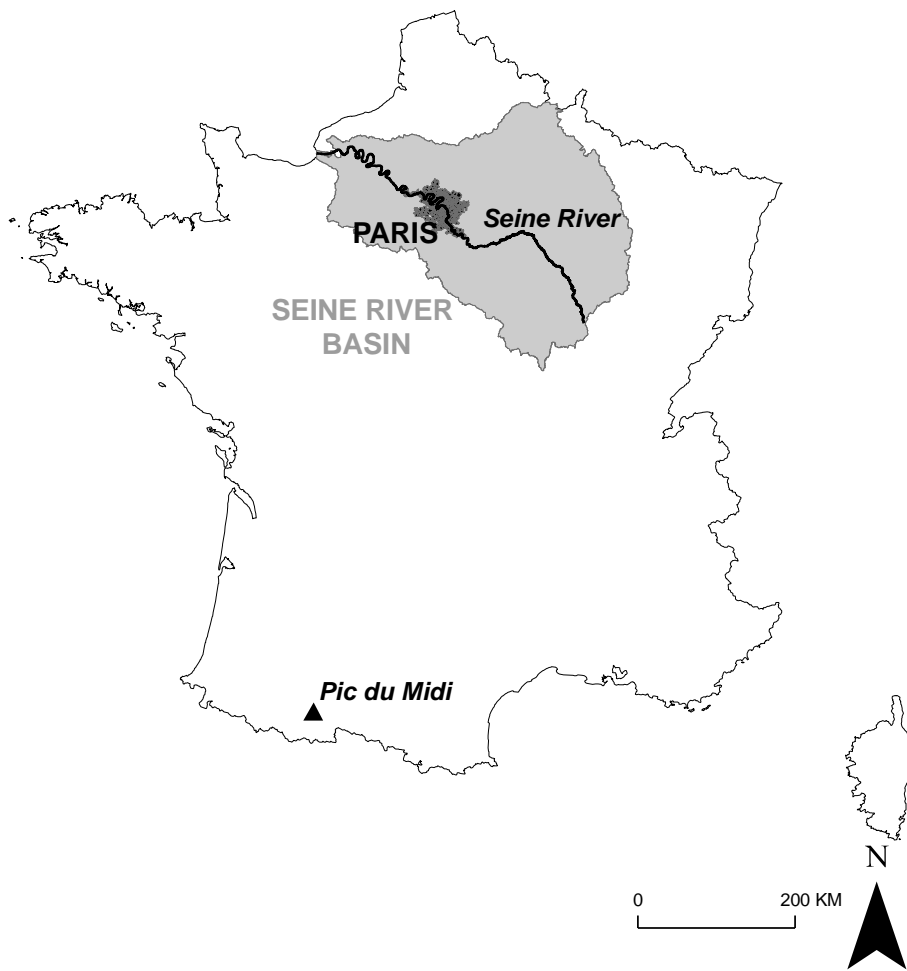
384 **Figure captions**

385 Figure 1. (a) Location of the Seine River basin and the *Pic du Midi* observatory in France; (b)
386 detailed map of the main Seine River tributaries and the grass, soil and suspended sediment
387 sampling sites.

388 Figure 2. Rainfall recorded at Paris during the study period.

Fig. 1

(a)



(b)

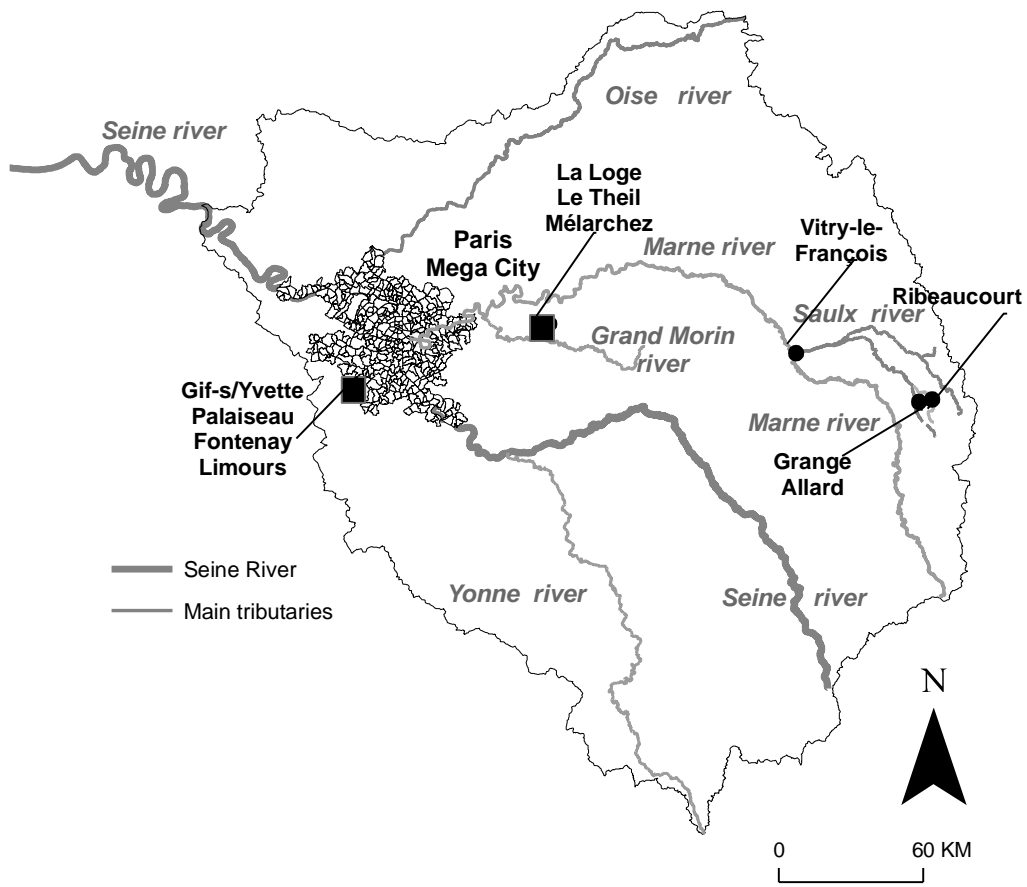


Figure 2

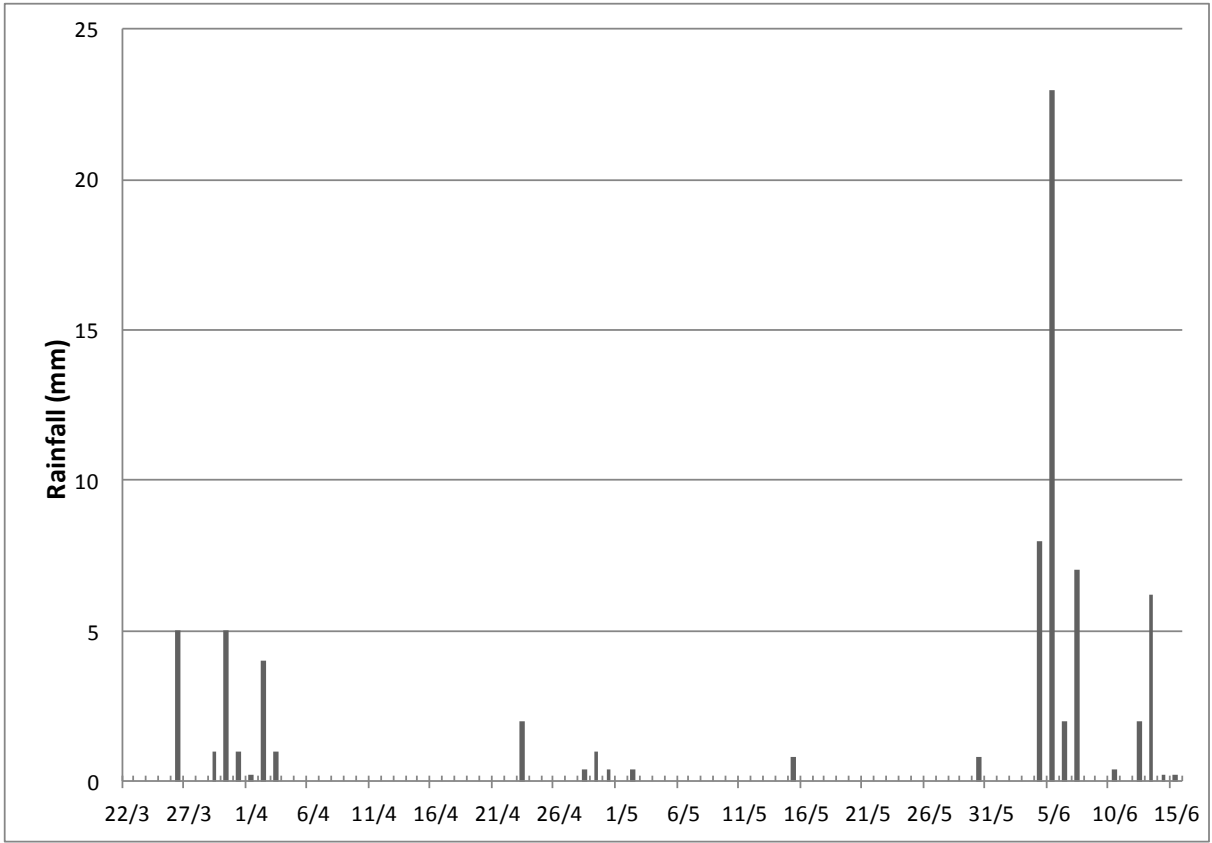


Table 1.

^{131}I , ^{137}Cs and ^{134}Cs activities ($\mu\text{Bq m}^{-3}$) determined in the filters collected on top of *Pic du Midi*, French Pyrénées. The uncertainties are one-sigma error associated with counting statistics.

Sample	Sampling period	^{131}I	^7Be	^{137}Cs	^{134}Cs
# 123	15/3 – 22/3	<MDA	1620±40	<MDA	<MDA
# 124	22/3 – 29/3	200±6	1650±30	<MDA	<MDA
# 125	29/3 – 5/4	<MDA *	1490±60	10±2	7±2
# 126	5/4 – 12/4	<MDA *	2890±80	<MDA	<MDA
# 127	12/4 – 19/4	<MDA *	2160±70	<MDA	<MDA
# 128	19/4 – 26/4	<MDA *	1470±50	<MDA	<MDA

MDA : Minimum Detectable Activity (< 10 $\mu\text{Bq m}^{-3}$ for ^{131}I ; < 4 $\mu\text{Bq m}^{-3}$ for ^{137}Cs and ^{134}Cs ; given at 2-sigma level).

* These samples were analyzed several weeks after sample collection, which prevented the potential detection of ^{131}I activities.

Table 2.

Fission product radionuclide activity (Bq kg^{-1}) in grass samples collected in the Parisian Region between 30 March and 14 June, 2011.

Sampling date	Sampling site	^{131}I	^{134}Cs	^{137}Cs	$^{137}\text{Cs}/^{134}\text{Cs}$	$^{131}\text{I}/^{137}\text{Cs}$	$^{131}\text{I}/^{134}\text{Cs}$	^7Be
30/03/2011	Les Avenelles	< 3.6	< 2.5	< 2.5				45±10
	Mélarchez	4.3 ±1.4	< 2.9	< 2.9				140±20
	La Loge	4.0±0.6	< 1.0	< 1.1				140±10
	Le Theil	2.9±0.4	< 0.7	< 0.8				15±5
31/03/2011	Gif-s/Yvette	6.6±0.4	1.3±0.3	0.9±0.3	0.7±0.4	7.3±2.9	5.1±1.5	90±5
05/04/2011	Palaiseau	11.0±1.0	1.6±0.4	1.2±0.4	0.7±0.4	9.0±3.8	6.9±2.3	170±5
09/04/2011	Fontenay-les-Briis	1.7±0.1	0.2±0.1	1.0±0.1	5.0±3.0	1.7±0.3	8.5±4.8	35±5
10/04/2011	Limours	4.5±0.4	1.4±0.3	1.6±0.3	1.1±0.4	2.8±0.8	3.2±1.0	55±5
28/04/2011	Mélarchez	< 0.2	< 0.1	< 0.1				10±5
	Le Theil	< 0.2	< 0.1	< 0.1				10±5
14/06/2011	Mélarchez	< 0.3	< 0.2	< 0.2				30±5
	Le Theil	< 0.1	< 0.1	< 0.1				25±5

Table 3.

Fission product radionuclide activity (Bq kg^{-1}) in soil and sediment samples collected in the Seine River basin between 16 March and 14 June, 2011. The error values shown are 2-sigma uncertainties.

Sampling date	Sampling site/matrix	^{131}I	^{134}Cs	^{137}Cs	$^{131}\text{I}/^{137}\text{Cs}$	$^{210}\text{Pb}_{\text{XS}}$	^7Be	$^7\text{Be}/^{210}\text{Pb}_{\text{XS}}$
30/03/2011	La Loge (soil)	<0.2	<0.2	3.2±0.1		10±1	5±1	0.5±0.2
	Mélarchez (soil)	0.4±0.1	< 0.2	1.6±0.1	0.2±0.1	10±1	10±1	1.0±0.2
	Mélarchez (sediment)	4.5±1.7	< 2.2	6.0±0.6	0.7±0.3	50±5	55±5	1.1±0.2
12/04/2011	Grange Allard (sediment)	60±30	< 2.7	3.6±0.8	15±11	30±5	70±10	2.3±0.7
	Ribeaucourt (sediment)	< 50	<4.3	6.4±1.2		60±10	110±10	1.8±0.5
	Vitry-le-François (sediment)	<50	<2.9	6.1±0.8		90±10	45±10	0.5±0.2
28/04/2011	Mélarchez (sediment)	< 44	<1.2	3.8±0.7		35±5	55±5	1.6±0.4
17/05/2011	Mélarchez (sediment)	< 44	< 2.2	6.0±0.7		50±5	55±5	1.1±0.2
07/06/2011	Grange Allard (sediment)	<57	<3.1	3.3±0.8		50±10	160±10	3.2±0.8
	Ribeaucourt (sediment)	<3	<0.8	8.0±0.6		75±5	160±5	2.1±0.2
	Vitry-le-François (sediment)	<13	<2.7	2.2±0.7		35±10	75±5	2.1±0.8
14/06/2011	Mélarchez (sediment)	< 21	< 2.2	1.2±0.4		20±5	35±5	1.8±0.7