



**HAL**  
open science

## **Radiocarbon and radiocesium in litter fall at Kawamata, to 45 km NW from the Fukushima Dai-ichi nuclear power plant (Japan)**

Martine Paterne, O. Evrard, Christine Hatté, Patrick Laceby, Julius Nouet, Yuichi Onda

► **To cite this version:**

Martine Paterne, O. Evrard, Christine Hatté, Patrick Laceby, Julius Nouet, et al.. Radiocarbon and radiocesium in litter fall at Kawamata, to 45 km NW from the Fukushima Dai-ichi nuclear power plant (Japan). *Journal of Radioanalytical and Nuclear Chemistry*, 2019, 319 (3), pp.1093-1101. 10.1007/s10967-018-6360-9 . hal-02087713

**HAL Id: hal-02087713**

**<https://hal.science/hal-02087713v1>**

Submitted on 15 May 2020

**HAL** is a multi-disciplinary open access archive for the deposit and dissemination of scientific research documents, whether they are published or not. The documents may come from teaching and research institutions in France or abroad, or from public or private research centers.

L'archive ouverte pluridisciplinaire **HAL**, est destinée au dépôt et à la diffusion de documents scientifiques de niveau recherche, publiés ou non, émanant des établissements d'enseignement et de recherche français ou étrangers, des laboratoires publics ou privés.

1 Radiocarbon and radiocesium in litter fall at Kawamata, ~45 km NW from the Fukushima  
2 Dai-ichi nuclear power plant (Japan)

3 Martine Paterne (corresponding author)<sup>1</sup>, Olivier Evrard<sup>1</sup>, Christine Hatté<sup>1</sup>, J. Patrick  
4 Laceby<sup>1,2</sup>, Julius Nouet<sup>3</sup>, Yuichi Onda<sup>4</sup>

5 <sup>1</sup>Laboratoire des Sciences du Climat et de l'Environnement, LSCE/IPSL, CEA-CNRS-UVSQ,  
6 Université Paris- Saclay, Domaine du CNRS, F-91198 Gif-sur-Yvette,  
7 [martine.paterne@lsce.ipsl.fr](mailto:martine.paterne@lsce.ipsl.fr), [olivier.evrard@lsce.ipsl.fr](mailto:olivier.evrard@lsce.ipsl.fr), [christine.hatte@lsce.ipsl.fr](mailto:christine.hatte@lsce.ipsl.fr),

8 <sup>2</sup> Environmental Monitoring and Science Division, Alberta Environment and Parks, 3115 – 12  
9 Street NE, Calgary, Alberta, Canada, [Patrick.Laceby@gov.ab.ca](mailto:Patrick.Laceby@gov.ab.ca)

10 <sup>3</sup> UMR GEOPS 8148 - Géosciences Paris Sud, Université Paris Sud/CNRS, Rue du Belvédère,  
11 F-91400 Orsay, [julius.nouet@u-psud.fr](mailto:julius.nouet@u-psud.fr)

12 <sup>4</sup> Center for Research in Isotopes and Environmental Dynamics (CRIED), University of  
13 Tsukuba, Tsukuba, Japan, [onda@geoenv.tsukuba.ac.jp](mailto:onda@geoenv.tsukuba.ac.jp)

14

15

## 16 **ABSTRACT**

17 Radiocarbon and radiocesium were measured on litter fractions (LF) collected on  
18 November 19<sup>th</sup>, 2011 at 40 km NW of the FDNPP. The <sup>137</sup>Cs concentration is much higher in  
19 the lower fraction LFb at  $226,650 \pm 170$  Bq kg<sup>-1</sup> than in the upper fraction LFa at  $7290 \pm 40$   
20 Bq kg<sup>-1</sup>. From leaf-air <sup>14</sup>C comparison, no excess <sup>14</sup>C due to the FDNPP accident is detected  
21 in LFa deposited in 2010-2011. A significant <sup>14</sup>C difference of 1.4% exists between pine  
22 needles and deciduous leaves in LFb, which may be due either to post-depositional processes  
23 or to a turnover time of 0.5-1yr of stored carbon for deciduous leaves growth.

24

25 Keywords: Radiocarbon. Radiocesium. Leaf Litter. Deciduous and evergreen Japanese trees.  
26 Fukushima nuclear accident.

27

## 28 1. Introduction

29 Boiling water reactor (BWR) nuclear power plants, such as the Fukushima Dai-ichi  
30 Nuclear Power Plant (FDNPP), contribute to a release of  $^{14}\text{C}$  into the atmosphere in the  
31 gaseous form of  $^{14}\text{CO}_2$  during routine operations [1]. The excess  $^{14}\text{C}$  is well recorded in  
32 vegetation, such as tree-ring and leaves, in the vicinity of such BWR nuclear plants [2; 3-7].  
33 Monitoring this excess  $^{14}\text{C}$  is of biological significance because of the rapid exchange of the  
34  $^{14}\text{CO}_2$  within the terrestrial carbon reservoirs and its long residence time ( $T=5730$  years) [1].

35 Significant leakage of radionuclides occurred at the time of the FDNPP accident on  
36 2011, March 11<sup>th</sup>. Among these radionuclides, post-accidental research has focused on the  
37 gamma-emitting radionuclides [8], and mainly  $^{137}\text{Cs}$ . A heavily contaminated area by the  
38  $^{137}\text{Cs}$  fallout, ~70% of which is covered by mixed forests, was identified downwind of the  
39 power plant in the prevailing northwestward wind direction in a variety of environments,  
40 including lakes, rivers, agricultural soils and forest ecosystems [9-17]. In contrast, the  $\beta$ -  
41 emitter  $^{14}\text{C}$  has been understudied after the FDNPP accident [8], although it has provided  
42 clues on the dispersion patterns of atmospheric  $^{14}\text{CO}_2$  release from nuclear fuels reprocessing  
43 plants in Japanese context [18-20] and on the long-term fate of Fukushima-derived  $^{137}\text{Cs}$  in  
44 forest soils similar to those found in the Fukushima prefecture from the comparative analyses  
45 of bomb-produced  $^{14}\text{C}$  and  $^{137}\text{Cs}$  migration in soils [21]. High  $^{14}\text{C}$  activities were measured  
46 from tree ring analyses of a Japanese cedar in the local environment (~2 km far from FDNPP),  
47 although revealing a reverse pattern between the early and late wood of year 2011 [4,7]. The  
48 activity of  $^{14}\text{C}$  was indeed higher in the late wood than in the early wood, while the seasonal  
49 transition commonly occurs between mid-June and mid-July [7]. The  $^{14}\text{C}$  activities decreased  
50 to background levels at 14 km northwest of the FDNPP in the highly contaminated region  
51 while the dispersion pattern of  $^{14}\text{C}$  in the southward direction remained unclear [3-5, 7]. A  
52 small enrichment of  $^{14}\text{C}$ , linked to a  $^{14}\text{C}$  release during the accident, has been noted in the late  
53 wood of the 2011 tree-ring from the Iwaki area located at 50 Km south of the FDNPP [3].

54 New annual leaves of oaks and beeches are preformed in winter buds before budburst  
55 in spring [22], and the  $^{14}\text{C}$  analyses of these leaf species may provide new insights on the  
56 storage of the released  $^{14}\text{C}$  from the damaged FDNPP in March 2011 by the vegetation  
57 samples. We report here the results of radiocarbon ( $^{14}\text{C}$ ) analyses of two leaf litter fractions  
58 collected on November 19<sup>th</sup>, 2011 in a temperate deciduous broad-leaved (mixed) forest  
59 (DBF) in Kawamata Town located at ~45 km downwind of the FDNPP (Fig. 1). The

60 Kawamata mixed forest is mainly composed of deciduous trees (oaks and beeches) and  
61 Japanese red pine trees [22] and it is located in the heavily contaminated area by the  $^{137}\text{Cs}$   
62 fallout in the northwestward wind direction from FDNPP [9, 13 *and references therein*]. The  
63 uppermost fraction (LFa) is composed of mostly entire deciduous leaves of beeches and oaks  
64 and the immediately underlying fraction (LFb) includes fragmented deciduous leaves of the  
65 same species and pine needles. The leaves in LFa are associated to the 2011 leaf fall, which  
66 commonly occurs between October and November [22, 24,25].  $^{14}\text{C}$  was measured on the bulk  
67 litter and on individual leaves and pine needles picked in the lowermost fraction LFb. In  
68 addition to  $^{14}\text{C}$  analyses, radiocesium ( $^{137}\text{Cs}$ , Bq kg $^{-1}$ ) concentration was measured on bulk  
69 litter samples. We analyzed the timing of deposition of leaf material in the two litter fractions,  
70 that we discussed considering either a post-depositional disturbance of litter fall deposits or a  
71 turnover time of carbon (C) reserves in the growing vegetation.

72

## 73 **2. Material and Methods**

74 The sampling site of the litter is located close to the Kawamata Town elementary  
75 school (37°60'N; 140°68E; 562m asl) at around 45 km NW downwind of FDNPP (Fig. 1).  
76 Litter deposited on a flat floor parcel of a mixed forest composed of oaks (*Quercus aliena*),  
77 beeches (*Fagus crenata*) and Japanese red pines (*Pinus densiflora*) with a population density  
78 of 2,500 trees/ha [23]. The two samples of litter fall were taken one plot from the surface over  
79 a depth of 4 cm well above the soil. After sampling, the litter fractions have been stored in  
80 hermetically closed plastic bags. Samples have been dried at 40°C. The uppermost fraction of  
81 the litter (LFa) is approximately 1cm thick and contains mostly *quasi* entire deciduous leaves  
82 and very few pine needles (1% dry weight (d.w.)) (Plate 1). The immediately underlying 3 cm  
83 thick fraction (LFb) includes 27% d.w. pine needles as an average. The size of the deciduous  
84 leaves varies from 0.5 cm to 2-3 cm and the pine needles may have sizes up to 3-4 cm. The  
85 two litter fractions are characterized by macroscopically recognizable material of non-  
86 degraded leaves.

87 Dominant ground level winds from the Weather Station at Iitate (Japanese  
88 Meteorological Agency, RSMC Tokyo, Japan), close to Kawamata Town, are westward in  
89 summer and eastward in winter. At the time of FDNPP accident in March 2011, the eastward  
90 winds dominated (Fig. 1). The amount of annual precipitation was higher in 2010 than in  
91 2011 at Iitate [26].

92 Prior to AMS-<sup>14</sup>C analyses, the bulk litter samples were first crushed ( $\leq 1\text{mm}$ ). The  
93 classical Acid-Alkali-Acid (AAA) pre-treatment of routine samples of organic matter was  
94 applied to eliminate potential contamination. Transformation into CO<sub>2</sub> was described  
95 elsewhere [27] and was performed at the Gif-LSCE Radiocarbon Laboratory. Graphite targets  
96 were prepared and measured at the French National AMS-ARTEMIS-LMC14 facility [28].  
97 The % C in litter is obtained from the mass of sample and from laboratory calibration between  
98 pressure of CO<sub>2</sub> of burned sample and the mass of carbon. Individual deciduous leaves and  
99 two different pine needles picked into litter LFb were also analyzed. Contiguous samples  
100 were cut on one of the pine needle, one sample including fungi and the other representing  
101 *clean* leaf. Samples of deciduous leaves were obtained from one single free-fungi leaf and  
102 from several pieces of deciduous leaves including fungi. After the classical AAA pre-  
103 treatment, aliquots containing about 1mg of carbon were weighted into tin boats and  
104 processed using the automated graphitization equipment (AGE3) [29]. Measurements were  
105 performed on the MICADAS equipment at the Gif-LSCE Radiocarbon Laboratory [30, 31].  
106 The <sup>14</sup>C/<sup>12</sup>C activity of the samples is  $\delta^{13}\text{C}$ -normalized and expressed as Fraction modern [32,  
107 33]. The <sup>14</sup>C content of litter material were compared with those of *clean* air values of <sup>14</sup>CO<sub>2</sub>  
108 as measured at the Schauinsland/Jungfrau stations (SIL) [34,35] and with the stack of  
109 atmospheric <sup>14</sup>C data of the Northern Hemisphere Zone 2 (NHZone2) [36] to analyze the  
110 timing of deposition of leaf material in the two litter fractions.

111 In addition, <sup>13</sup>C/<sup>12</sup>C ratio of the bulk litter was measured using an online continuous  
112 Elementary Analyzer (Flash EA 1112) coupled with an Isotopic Ratio Mass Spectrometer  
113 (Finigan Delta+XP). The results are expressed in  $\delta^{13}\text{C}$  per mil (‰) against the international  
114 standard V-PDB (Vienna Pee Dee Belemnite). Uncertainty is defined according to the source  
115 linearity checked for each run based on internal standard.

116 <sup>137</sup>Cs activities were determined by gamma spectrometry using coaxial N- and P-type  
117 HPGe detectors (Canberra/Ortec). The samples were first dried at 40 °C for ~48 h and pressed  
118 into 11 mL polyethylene containers ( $\varnothing$  45mm, h 7mm). <sup>137</sup>Cs activities were measured at the  
119 662 keV emission peak. Counting efficiencies and reliability were quantified using certified  
120 International Atomic Energy Agency (IAEA) standards (IAEA-156, IAEA-372) prepared in  
121 the same containers as the samples. All activities were decay-corrected to March 14, 2011, the  
122 date of the main radionuclide fallout deposition [37].

123

### 124 3. Results

125 The carbon content (%) in the bulk litter at ~45% is close to that measured in fresh  
126 leaves and litters [38, 39] (Table 1). The values of  $\delta^{13}\text{C}$  (‰) are similar in the two bulk litter  
127 samples at around -31‰. They are in agreement with the values of litter leaves from  
128 temperate broadleaved forests [40, 41] and these very depleted  $\delta^{13}\text{C}$  values would indicate a  
129 pre-decomposition stage of the leaves [41]. The macro- and microscopic observations show  
130 indeed no visible to moderate signs of degradation of the leaves attesting a recent deposition  
131 of the two litter samples (Plate 1).

132 The  $^{14}\text{C}$  activity is significantly higher in the lower litter fraction LFb at  $1.0562 \pm$   
133  $0.0031$  than in the uppermost fraction LFa at  $1.0446 \pm 0.0030$  at the 95 % confidence level  
134 using the statistical Chi-square test (Fig. 2; Table 1). No significant statistical difference  
135 exists between the *clean* leaves and needles and those including fungi. In LFb, the mean value  
136 of  $^{14}\text{C}$  in deciduous leaves at one standard deviation is significantly higher at  $1.0576 \pm 0.0034$   
137 than in pine needles at  $1.0430 \pm 0.0027$ .

138 The litter fractions have much higher concentrations of  $^{137}\text{Cs}$  than background values,  
139 measured at  $5.0 \pm 1.2 \text{ Bq kg}^{-1}$  in a litter fall collected in 2007 in a Japanese deciduous forest  
140 [11]. The  $^{137}\text{Cs}$  concentration is 30 times higher in the litter fraction LFb at  $226,650 \pm 170 \text{ Bq}$   
141  $\text{kg}^{-1}$  than in the uppermost fraction LFa at  $7290 \pm 40 \text{ Bq kg}^{-1}$  (Table 1).

142

### 143 3. Discussion

144 The seasonal growth patterns of Kawamata forest vegetation are overall mostly similar  
145 among the studied species. Field observations in a northern Japanese DBF indicate that leaves  
146 of beeches and oaks, as those found in the Kawamata litter, appear as a flush in about one or  
147 two weeks between late April and mid-May without late leaf production and they fall between  
148 October and November [22, 24, 25]. Needles of the Japanese red pines elongated rapidly in  
149 June-July and the old needles usually fall between September and November [42]. The  $^{14}\text{C}$  of  
150 leaves of evergreen and deciduous trees of same ages do not show significant differences and  
151 they are similar to that of the atmosphere in which they grow [6, 43-45]. Foliar  $\delta^{13}\text{C}$  in  
152 terrestrial vegetation vary as a function of vegetation species and of environmental parameters,  
153 such as, among others, light, temperature and water availability. The  $\delta^{13}\text{C}$  values are more  
154 negative by about 1 to 4‰ in deciduous leaves than in pine needles [46, 47]. The  $^{14}\text{C}$

155 activities of the deciduous leaves and of the pine needles are  $^{13}\text{C}$ -corrected for carbon  
156 fractionation due to biological processes and to the analytical  $^{14}\text{C}$  procedures. Moreover, a  
157 1‰ change in  $\delta^{13}\text{C}$  results in a difference of the  $^{14}\text{C}/^{12}\text{C}$  activity of 0.002 that is very much  
158 lower than the measured  $^{14}\text{C}$  difference of 1.4% between the deciduous leaves and pine  
159 needles in LFb. The  $^{14}\text{C}$  discrepancy between deciduous leaves and needles in LFb cannot be  
160 due to seasonal variations of atmospheric  $^{14}\text{C}$ , nor to plant specific differences.

161 To analyze such a  $^{14}\text{C}$  difference, we compared the  $^{14}\text{C}$  activity of the Kawamata  
162 samples to that of the SIL clean air during the growth season of leaves (May-June) [34, 35],  
163 which is very similar to that of NHZone 2 [36] (Fig. 2a). The  $^{14}\text{C}$  of the deciduous leaves in  
164 LFa are compatible with the atmospheric  $^{14}\text{C}$  of year 2011, but also of year 2010, and the  
165 deciduous leaves in LFa overlay the older deciduous leaves in LFb mainly deposited between  
166 2007 and 2009 (Fig. 2). LFb may contain a part of fallen leaves in 2010. This contamination  
167 cannot however exceed 5% as estimated from mixing equations using the very similar values  
168 of atmospheric  $^{14}\text{C}$  between 2007 and 2009 (Fig. 2) and those of year 2010 and from the  
169 comparison of these estimates with the  $^{14}\text{C}$  of the *clean* leaf in LFb (supplementary material,  
170 Fig. S1). The pine needles in LFb would have grown during one or two years and fallen down  
171 either in 2010 or 2011. They have the same ages than those of the deciduous leaves in LFa,  
172 although sampled below, and they are significantly younger than the deciduous leaves though  
173 mixed in the same fraction LFb. Extending the seasonal age calibration of the  $^{14}\text{C}$  of leaf  
174 material from April to September does not change significantly the timing of leaf deposits  
175 (Supplementary material, Fig. S2).

176

### 177 **3.1 Disturbance of the leaf fall deposits**

178 The  $^{14}\text{C}$  discrepancy between the deciduous leaves and pine needles in LFb may be  
179 due to a vertical transfer and mixing of pine needles from the uppermost fraction LFa to the  
180 immediately underlying fraction LFb. The litter fractions were however sampled on a flat  
181 forest parcel. The  $^{14}\text{C}$  of the bulk litter LFa and LFb are significantly different as well as the  
182  $^{14}\text{C}$  of deciduous leaves in LFa and in LFb (Fig. 2). Moreover, although the number of  
183 analyses is small, very similar values of  $^{14}\text{C}$  are measured in two different pine needles picked  
184 into two different subsamples of LFb and they significantly differ from those of deciduous  
185 leaves (Table 1; Fig. 2). A vertical mixing of pine needles between the two litter fractions is  
186 very unlikely without having at the same time a mixing of deciduous leaves.

187 Pine needles may seasonally fall down immediately before the deciduous leaves in  
188 LFa and they would have been mixed with the lowermost fraction while sampling LFb (Fig.  
189 3). Needle longevity on shoots of *Pinus densiflora* may vary between ~1 and ~3 years  
190 depending on the more or less polluted environments in which they grow [42]. The pine  
191 needles in LFb may have lifetimes of 1 or 2 years, consistent with field observations (Fig. 2).  
192 A fall of the 2yr old pine needles (2010-2011) prior to that of the deciduous leaves in 2011 in  
193 LFa would have covered the deciduous leaves of year 2010 which are not observed below in  
194 LFb (Fig. 3, case a). A fall of the 1yr old pine needles (2010) prior to the deciduous leaves fall  
195 in 2010 and no fall of pine needles in 2011 in LFa (Fig. 3, case b) conflicts with the measured  
196 <sup>137</sup>Cs concentration which is 30 times lower in LFa than in LFb. At the time of the FDNPP  
197 accident in March 2011, <sup>137</sup>Cs deposited directly on the forest floor due to the absence of  
198 leaves on trees and directly on the 2010 litter fall and preceding fall, which were therefore  
199 highly contaminated.

200 The <sup>14</sup>C discrepancy between the pine needles and the deciduous leaves in LFb may be  
201 relevant to disturbance of forest litter deposit, admitting either the lack of the 2010 deciduous  
202 leaves fall, or a *quasi*-complete leaching of the <sup>137</sup>Cs fallout on the 2010 leaf fall deposit at our  
203 sampling site, eight months after the FDNPP accident.

204

### 205 **3.2 The carbon paths during deciduous leaf growth**

206 Leaf buds that develop in winter and expanding leaves, such as those of oaks and  
207 beeches in the Japanese DBF [22], would grow from carbon (C) reserves stored in previous  
208 years [48]. Using an inadvertent huge spike of <sup>14</sup>C release near a Californian temperate  
209 deciduous oak forest and numerical modeling, Gaudinski et al. [2009] estimated that the mean  
210 age of the stored C used for leaf-bud and leaf growth of oak and red maple tree species was  
211 about 0.5 to 1 year.

212 We estimated the <sup>14</sup>C values of the deciduous leaves when accounting for a turnover  
213 time of stored carbon reserves during the growth season and their use for deciduous leaves  
214 growth during the following year using:

$$215 \frac{A}{A_0} = \exp\left(-\frac{t}{\tau}\right)$$



216 with  $t$  the duration of the growth season and  $\tau$  the turnover time in days, and  $A/A_0$  the  
217 ratio of the atmospheric  $^{14}\text{C}$  activity during the considered years.

218 The contribution of the stored carbon during the preceding year in newly formed  
219 leaves would be 51% (70%) with a turnover time  $\tau$  of 0.5 year (1year). From a simple mixing  
220 equation using the SIL atmospheric  $^{14}\text{C}$  in years 2010 and 2009 at  $1.0477 \pm 0.0028$  and  
221  $1.05427 \pm 0.0022$  respectively [34, 35] and the proportion given above 51% (70%) of carbon  
222 reserves from year 2009 and 49% (30%) of atmospheric  $^{14}\text{C}$  from year 2010, then the  $^{14}\text{C}$   
223 values of the deciduous leaves growing in year 2010 would be estimated at  $1.0510 \pm 0.0025$   
224 ( $1.0524 \pm 0.0025$ ). Those values are compatible with the mean  $^{14}\text{C}$  value of the deciduous  
225 leaves in LFb at  $1.0576 \pm 0.0034$  at the 95% confidence level. Consequently, the deciduous  
226 leaves in LFb may have grown and fallen in year 2010, and they would have a higher  $^{14}\text{C}$   
227 value than the contemporaneously living pine needles. Similarly, the  $^{14}\text{C}$  value of the  
228 deciduous leaves in LFa at  $1.04459 \pm 0.00296$  is compatible with the estimated value at  
229  $1.0471 \pm 0.0028$  for leaf growth and fall in 2011 with the use of 51% stored C during year  
230 2010. Extending the growth season from April to September does not change significantly  
231 such values.

232 The timing of the litter fractions at our site of sampling and the  $^{14}\text{C}$  discrepancy  
233 between the pine needles and deciduous leaves in the lower litter sample LFb are well  
234 explained when accounting for a turnover time of carbon for deciduous leaves growth (Fig. 3,  
235 case c). Fraction LFa would result from deciduous leaf fall in 2011 and fraction LFb from  
236 those of year 2010 and previous years. The pine needles in LFb would have fallen down  
237 either contemporaneously with the deciduous leaves in 2010 and mixed with older leaves, or  
238 immediately before the deciduous leaves in 2011 and inadvertently mixed with the lower  
239 fraction. No excess  $^{14}\text{C}$  due to the FDNPP accident is detected in the preformed deciduous  
240 leaves of oak and beeches in winter buds, that is in agreement with the  $^{14}\text{C}$  value of cedar  
241 tree-ring in the nearby site of Yamakiya [3].

242

#### 243 **4. Conclusion**

244  $^{14}\text{C}$  analyses were performed on leaf material of two contiguous litter fractions LFa  
245 and LFb deposited in the Kawamata Town mixed forest and collected on November 19<sup>th</sup>,  
246 2011. Compared with the atmospheric  $^{14}\text{C}$  records, they revealed that deciduous leaves of oak

247 and beeches deposited in 2010 and 2011 in the uppermost fraction LFa and between 2007 and  
248 2009 in the underlying fraction LFb. No excess  $^{14}\text{C}$  due to the FDNPP is detected in the fallen  
249 leaves in LFa. The presence of pine needles of younger ages than those of the associated  
250 deciduous leaves in the lowermost fraction LFb may be explained by post-depositional  
251 processes such as either a lack of the 2010 deciduous leaves deposit if LFa is uniquely  
252 composed of the 2011 deciduous leaf fall or a *quasi*-complete leaching of the  $^{137}\text{Cs}$  fallout on  
253 the 2010 leaf fall deposit at our site of sampling in November 2011 if LFa is composed of  
254 deciduous leaf fall of year 2010 and 2011. The ~30 times higher  $^{137}\text{Cs}$  concentration in the  
255 lower litter fraction LFb compared to that found in the upper fraction LFa is in agreement  
256 with these two assumptions, as the  $^{137}\text{Cs}$  fallout from the FDNPP accident in March occurred  
257 before the canopy extension associated with the growth of the deciduous leaves.

258 Alternately, deciduous leaves of oak and beeches preformed in winter buds would  
259 partly grow from stored carbon reserves during the preceding year. The previously published  
260 values of  $^{14}\text{C}$  of fresh leaves of deciduous trees and pine needles [6, 43-45] are also  
261 compatible within the uncertainties with the atmospheric  $^{14}\text{C}$  of the preceding year. A  
262 turnover time of carbon of 0.5-1yr, consistent with previous observations [48], may be  
263 estimated from the values of  $^{14}\text{C}$  of leaf material deposited in the Kawamata Town mixed  
264 forest. Estimating the turnover time of carbon in vegetation is likely more complex owing to  
265 the different paths of non-structural carbon in trees [49]. Further  $^{14}\text{C}$  analyses of leaf material  
266 in different stands in broadleaved forest would help in discriminating the causes of the  
267 observed  $^{14}\text{C}$  discrepancy among different species.

268

269 **Acknowledgements:** This work received the financial support of CEA-CNRS-UVSQ and of  
270 the French National Research Agency (ANR) in the framework of the TOFU (ANR-11-  
271 JAPN-001) and the AMORAD (ANR-11-RSNR-0002) research projects. Thanks are due to  
272 the teams of Gif-ECHOMICADAS Radiocarbon Laboratory and of the French AMS National  
273 facility LMC14. We thanked an anonymous reviewer for very helpful comments. The authors  
274 declare no conflict of interests.

275

276 **References**

- 277 1. IAEA (2004) Management of waste containing tritium and Carbon-14 Technical  
278 reports series n°421 International Atomic Energy Agency, Austria, Vienna 2004,  
279 STI/DOC/010/421, 109pp
- 280 2. Stenström K, Erlandsson B, Mattsson S, Thornborg C, Hellborg R, Kiisk M, Persson P,  
281 Skog G (2000)  $^{14}\text{C}$  emission from Swedish nuclear power plants and its effect on the  
282  $^{14}\text{C}$  levels in the environment. Internal Report LUNDFD6/(NFFR-3079), Lund  
283 University
- 284 3. Xu S, Cook GT, Cresswell AJ, Dunbar E, Freeman SP, Hastie H, Hou X, Jacobsson P,  
285 Naysmith P, Sanderson DC (2015) Radiocarbon concentration in modern tree rings  
286 from Fukushima, Japan. *J Environ Radioact* 146:67–72
- 287 4. Xu S, Cook GT, Cresswell AJ, Dunbar E, Freeman SP, Hou X, Jacobsson P, Kinch  
288 HR, Naysmith P, Sanderson DCW, Tripney BG (2016a) Radiocarbon releases from  
289 the 2011 Fukushima nuclear accident. *Sci Rep* 6:36947
- 290 5. Xu S, Cook GT, Cresswell AJ, Dunbar E, Freeman SP, Hastie H, Hou X, Jacobsson P,  
291 Naysmith P, Sanderson DC, Tripney BG, Yamaguchi K (2016b)  $^{14}\text{C}$  levels in the  
292 vicinity of the Fukushima Dai-ichi Nuclear Power Plant prior to the 2011 accident. *J*  
293 *Environ Radioact* 157:90–96
- 294 6. Xu S, Cook GT, Cresswell AJ, Dunbar E, Freeman SP, Hastie H, Hou X, Kinch H,  
295 Naysmith P, Sanderson DWC, Zhang L (2016c) Carbon, cesium and iodine isotopes in  
296 Japanese cedar leaves from Iwaki, Fukushima. *J Radioanal Nucl Chem* 310:927–934
- 297 7. Chen B, Xu S, Cook GT, Freeman SP, Hou X, Liu CQ, Naysmith P, Yamaguchi K  
298 (2017) Local variance of atmospheric  $^{14}\text{C}$  concentrations around Fukushima Dai-ichi  
299 Nuclear Power Plant from 2010 to 2012. *J Radioanal Nucl Chem* 314:1001–1007
- 300 8. Steinhauser G (2014) Fukushima's Forgotten Radionuclides: A Review of the  
301 Understudied Radioactive Emissions. *Environ Sci Technol* 48:4649–4663
- 302 9. Hashimoto S, Ugawa S, Nanko K, Shichi K (2012) The total amounts of radioactively  
303 contaminated materials in forests in Fukushima, Japan. *Sci Rept* 2: 416
- 304 10. Koarashi J, Atarashi-Andoh M, Matsunaga T, Sato T, Nagao S, Nagai H (2012)  
305 Factors affecting vertical distribution of Fukushima accident-derived radiocesium in  
306 soil under different land-use conditions. *Sci Total Environ* 431:392–401
- 307 11. Koarashi J, Atarashi-Andoh M, Takeuchi E, Nishimura S (2014) Topographic  
308 heterogeneity effect on the accumulation of Fukushima-derived radiocesium on forest

- 309 floor driven by biologically mediated processes. *Sci Rept* 4, 6853
- 310 12. Koarashi J, Atarashi-Andoh M, Matsunaga T, Sanada (2016) Forest type effects on the  
311 retention of radiocesium in organic layers of forest ecosystems affected by the  
312 Fukushima nuclear accident. *Sci Rpt* 6, DOI:101038/srep38591
- 313 13. Evrard O, Laceby JP, Lepage H, Onda Y, Cerdan O, Ayrault S (2015) Radiocesium  
314 transfer from hillslopes to the Pacific Ocean after the Fukushima Nuclear Power Plant  
315 accident: A review. *J Environ Radioact* 148: 92-110
- 316 14. Takahashi J, Tamura K, Suda T, Matsumura R, Onda Y (2015) Vertical distribution  
317 and temporal changes of  $^{137}\text{Cs}$  in soil profiles under various land uses after the  
318 Fukushima Dai-ichi Nuclear Power Plant accident. *J Environ Radioact* 139:351-361
- 319 15. Coppin F, Hurtevent P, Loffredo N, Simonucci C, Julien A, Gonze M-A, Nanba K,  
320 Onda Y, Thiry Y (2016) Radiocaesium partitioning in Japanese cedar forests  
321 following the “early” phase of Fukushima fallout redistribution. *Sc Rep* 6: 37618
- 322 16. Imamura N, Komatsu M, Ohashi S, Hashimoto S, Kajimoto T, Kaneko S, Takano T  
323 (2017) Temporal changes in the radiocesium distribution in forests over the five years  
324 after the Fukushima Daiichi Nuclear Power Plant accident. *Sc Rpt* 7: 8179
- 325 17. Kato H, Onda Y, Hisadome K, Loffredo N, Kawamori A (2017) Temporal changes in  
326 radiocesium deposition in various forest stands following the Fukushima Dai-ichi  
327 Nuclear Power Plant accident. *J Environ Radioact* 166: 449-457
- 328 18. Ota M, Nagai H, Koarashi J (2016) Modeling dynamics of  $^{137}\text{Cs}$  in forest surface  
329 environments: application to a contaminated forest site near Fukushima and  
330 assessment of potential impacts of soil organic matter interactions. *Sci Total Environ*  
331 551–552:590–604
- 332 19. Koarashi J, Fujita H, Watanabe H, Sumiya S (2011) Diverse monitoring approaches  
333 reveal  $^{14}\text{C}$  dispersion pattern and its impact on the environment around the Tokai  
334 reprocessing plant. *J Nucl Sci Technol* 48 :120-129
- 335 20. Koarashi J, Fujita H, Nagaoka M (2016) Atmospheric discharge of  $^{14}\text{C}$  from the Tokai  
336 reprocessing plant: comprehensive chronology and environmental impact assessment.  
337 *J Nucl Sci Technol* 53 :546-553
- 338 21. Koarashi J, Atarashi-Andoh M, Hikaru Amano H, Matsunaga T (2017) Vertical  
339 distributions of global fallout  $^{137}\text{Cs}$  and  $^{14}\text{C}$  in a Japanese forest soil profile and their  
340 implications for the fate and migration processes of Fukushima-derived  $^{137}\text{Cs}$ . *J*  
341 *Radioanal Nucl Chem* 311:473–481
- 342 22. Kikuzawa K (1983) Leaf survival of woody plants in deciduous broad-leaved forests 1

- 343 Tall trees. *Can J Bot* 61: 2133-2139
- 344 23. Loffredo N, Onda Y, Kawamori A, Kato H (2014) Modeling of leachable  $^{137}\text{Cs}$  in  
345 throughfall and stemflow for Japanese forest canopies after Fukushima Daiichi  
346 Nuclear Power Plant accident. *Sci Total Environ* 493:701–707
- 347 24. Kikuzawa K (1988) Leaf survivals of tree species in deciduous broad-leaved forests.  
348 *Pl Sp Biol* 3:67-76
- 349 25. Tateno R, Aikawa T, Takeda H (2005) Leaf-fall phenology along a topography-  
350 mediated environmental gradient in a cool–temperate deciduous broad-leaved forest in  
351 Japan. *J For Res* 10:269–274
- 352 26. Laceby JP, Huon S, Onda Y, Vaury V, Evrard O (2016) Do forests represent a long-  
353 term source of contaminated particulate matter in the Fukushima Prefecture? *J of*  
354 *Environ Manag* 183: 742-753
- 355 27. Hatté C, Poupeau JJ, Tannau JF, Paterne M (2003) Development of an automated  
356 system for preparation of organic samples. *Radiocarbon* 45(3): 421-430
- 357 28. Moreau C, Caffy I, Comby C, Delqué-Količ E, Dumoulin J, Hain S, Vincent J (2013)  
358 Research and Development of the Artemis  $^{14}\text{C}$  AMS Facility: Status Report  
359 *Radiocarbon* 55(2): 331-337
- 360 29. Wacker L, Němec M, Bourquin J (2010) A revolutionary graphitisation system: Fully  
361 automated, compact and simple. *NIMS B* 268: 931-934
- 362 30. Synal HA, Stocker M, Suter M (2007) MICADAS: A new compact radiocarbon AMS  
363 system. *NIMS B* 259:7-13
- 364 31. Tisnérat-Laborde N, Thil F, Synal H-A, Cer soy S, Hatté C, Gauthier C, Massault M,  
365 Michelot J-L, Noret A, Siani G, Tombret O, Vigne J-D, Zazzo A (2015)  
366 ECHOMICADAS: A new compact AMS system to measuring  $^{14}\text{C}$  for Environment,  
367 Climate and Human Sciences. 22nd International Radiocarbon Conference, Dakar,  
368 Senegal
- 369 32. Stuiver M, Pollach, P (1977) Discussion: Reporting of  $^{14}\text{C}$  data. *Radiocarbon* 19:355-  
370 363
- 371 33. Reimer PJ, Brown TA, Reimer RW (2004) Discussion : reporting and calibration of  
372 post-bomb  $^{14}\text{C}$  data. *Radiocarbon* 46(3): 1299-1304
- 373 34. Levin I, Kromer B (2004) The tropospheric  $^{14}\text{CO}_2$  level in mid latitudes of the  
374 Northern Hemisphere. *Radiocarbon* 46(3):1261-1272
- 375 35. Hammer S, Levin I (2017) Monthly mean atmospheric  $\text{D}^{14}\text{CO}_2$  at Jungfrau joch and  
376 Schauinsland from 1986 to 2016. DOI/1011588/data/10100

- 377 36. Hua Q, Barbetti M, Rakowski AZ (2013) Atmospheric radiocarbon for the period  
378 1950-2010. *Radiocarbon* 55:2059-2072
- 379 37. Kinoshita N, Sueki K, Sasa K, Kitagawa J, Ikarashi S, Nishimura T, Wong Y-S, Satou  
380 Y, Handa K, Takahashi T, Sato M, Yamagata T (2011) Assessment of individual  
381 radionuclide distributions from the Fukushima nuclear accident covering central-east  
382 Japan PNAS. 108 (49): 19526-19529
- 383 38. Gloaguen JC, Touffet J (1982) Evolution du rapport C/N dans les feuilles et au cours  
384 de la décomposition des litières sous climat atlantique. Le hêtre et quelques conifères.  
385 *Ann Sci Forest* 39:219-230
- 386 39. Thomas SC, Martin A R (2012) Carbon Content of Tree Tissues: A Synthesis. *Forests*  
387 3:332-352
- 388 40. Sakata T, Ishizuka S, Takahashi M (2007) Separation of soil respiration into CO<sub>2</sub>  
389 emission sources using <sup>13</sup>C natural abundance in a deciduous broad-leaved forest in  
390 Japan. *Soil Science and Plant Nutrition* 53:328–336
- 391 41. Gautam MK, Lee KS, Song BY, Lee D, Bong YS (2016) Early-stage changes in  
392 natural <sup>13</sup>C and <sup>15</sup>N abundance and nutrient dynamics during different litter  
393 decomposition. *J Plant Res* 129:463-476
- 394 42. Kume A, Tsuboi N, Suzuki TSM, Chiwa M, Sakurai KNN, Horikoshi T, Sakugawa H  
395 (2000) Physiological characteristics of Japanese red pine, *Pinus densiflora* Sieb et  
396 Zucc, in declined forests at Mt Gokurakuji in Hiroshima Prefecture. *Japan Trees*  
397 14:305-311
- 398 43. Muraki Y, Masuda K, Arslanov A, Toyozumi T, Kato M, Naruse Y, Murata T,  
399 Nishiyama T (2001) Measurements of radiocarbon content in leaves from some  
400 Japanese sites. *Radiocarbon* 43(2B):695-701
- 401 44. Sakurai H, Tokanai F, Kato K, Takahashi Y, Sato T, Kikuchi K, Inui E, Arai Y,  
402 Masuda K, Miyahara H, Mundia C, Tavera W (2013) Latest <sup>14</sup>C Concentrations of  
403 Plant Leaves at High Altitudes in the Northern and Southern Hemispheres: Vertical  
404 Stability of Local Suess Effect. *Radiocarbon* 55(2-3):1573-1579
- 405 45. Lee JH, Kim CH, Kang J, Song S, Yun MH, Kim JC (2017) Radiocarbon data from  
406 the vicinity of four south Korean nuclear power plants in 2013-2014. *Radiocarbon*, 59  
407 (3):973–984
- 408 46. Garten CT, Taylor GE Jr (1992) Foliar  $\delta^{13}\text{C}$  within a temperate deciduous forest:  
409 spatial, temporal, and species sources of variation. *Oecologia* 90 :1-7
- 410 47. Brooks JR, Flanagan LB, Buchman N, Ehleringer JR (1997) Carbon isotope

- 411 composition of boreal plants: functional grouping of life forms. *Oecologia* 110 :301-  
412 311
- 413 48. Gaudinski J B, Torn M S, Riley W J, Swanston C, Trumbore S E, Joslin J D, Majdi  
414 Dawson H T E, Hanson P J (2009) Use of stored carbon reserves in growth of  
415 temperate tree roots and leaf buds: analyses using radiocarbon measurements and  
416 modeling. *Gl Ch Biol* 15:992–1014
- 417 49. Trumbore S, Czimczik CI, Sierra CA, Muhr J, Xu X (2015) Non-structural carbon  
418 dynamics and allocation relate to growth rate and leaf habit in California oaks *Tree*  
419 *Physiology* 35 :1206–1222
- 420

421 **Table Captions**

422 Table 1: Sample references and types, AMS  $^{14}\text{C}$  Lab codes, and results of carbon C in %,  $\delta^{13}\text{C}$   
423 in ‰, fraction modern  $^{14}\text{C}$  ( $F^{14}\text{C}$ ),  $\Delta^{14}\text{C}$  in ‰ and  $^{137}\text{Cs}$  in  $\text{Bq kg}^{-1}$ . Uncertainties are at  
424 one sigma ( $\sigma$ ).

425 **Plates**

426 Plate 1: Microscope Photographs of the leaves accumulated in litter layer L (A-C) and layer F  
427 (D-F) showing no visible sign of degradation of the leaves. The scales (white bar) are  
428 1cm in A, B and D; 0.5 cm in E;  $30\mu\text{m}$  in C and  $50\mu\text{m}$  in F. On image F, minerals are,  
429 magnetite, hematite and feldpars.

430

431 **Fig. Captions**

432 Fig. 1: Locations (A) of the Fukushima prefecture and (B) of the Kawamata Town  
433 ( $37.602482$ ;  $140.677049$ ;  $562\text{m asl}$ ) and the other sites cited in the text in northeast Japan.

434 Fig. 2: Seasonal age calibration of litter material and probability (in %) at the 95% confidence  
435 level as a function of time (in years). Calibration and probabilities were obtained using  
436 Chi-square tests between the values of  $^{14}\text{C}$  (Fraction modern,  $F^{14}\text{C}$ ) of the litter material  
437 and those of the seasonal (May-June) atmospheric  $^{14}\text{C}$  at the Schauinsland/Jungfrau  
438 stations (SIL) [34, 35] (green triangles).  $P_{0.05} \leq 5\%$  were not taken into account. Values of  
439  $F^{14}\text{C}$  are also reported for the stack of atmospheric  $^{14}\text{C}$  data (May-June) of the Northern  
440 Hemisphere Zone 2 (NHZone2) [36] (blue circles) and for the annual tree-ring record at  
441 Yamakiya, nearby Kawamata Town (violin diamonds) [4]. Colors on the probability  
442 diagrams referred to those of the time intervals (rectangles) of  $^{14}\text{C}$  calibration of the litter  
443 fractions LFa and LFb (A,B), of the mean  $^{14}\text{C}$  of deciduous leaves (DL) and pine needles  
444 (PN) in LFb (C,D), and of individual deciduous leaves (red and rose) and pine needles  
445 (dark, medium and light green) in LFb (E,F).

446 Fig. 3: Schematic description of the litter fall deposition LFa and LFb showing the results of  
447 the  $^{137}\text{Cs}$  concentration, of the time interval of calibrated  $^{14}\text{C}$  of deciduous leaves (DL)  
448 and pine needles (PN) and of  $F^{14}\text{C}$ . The three cases (a, b and c) are discussed in the text.

449



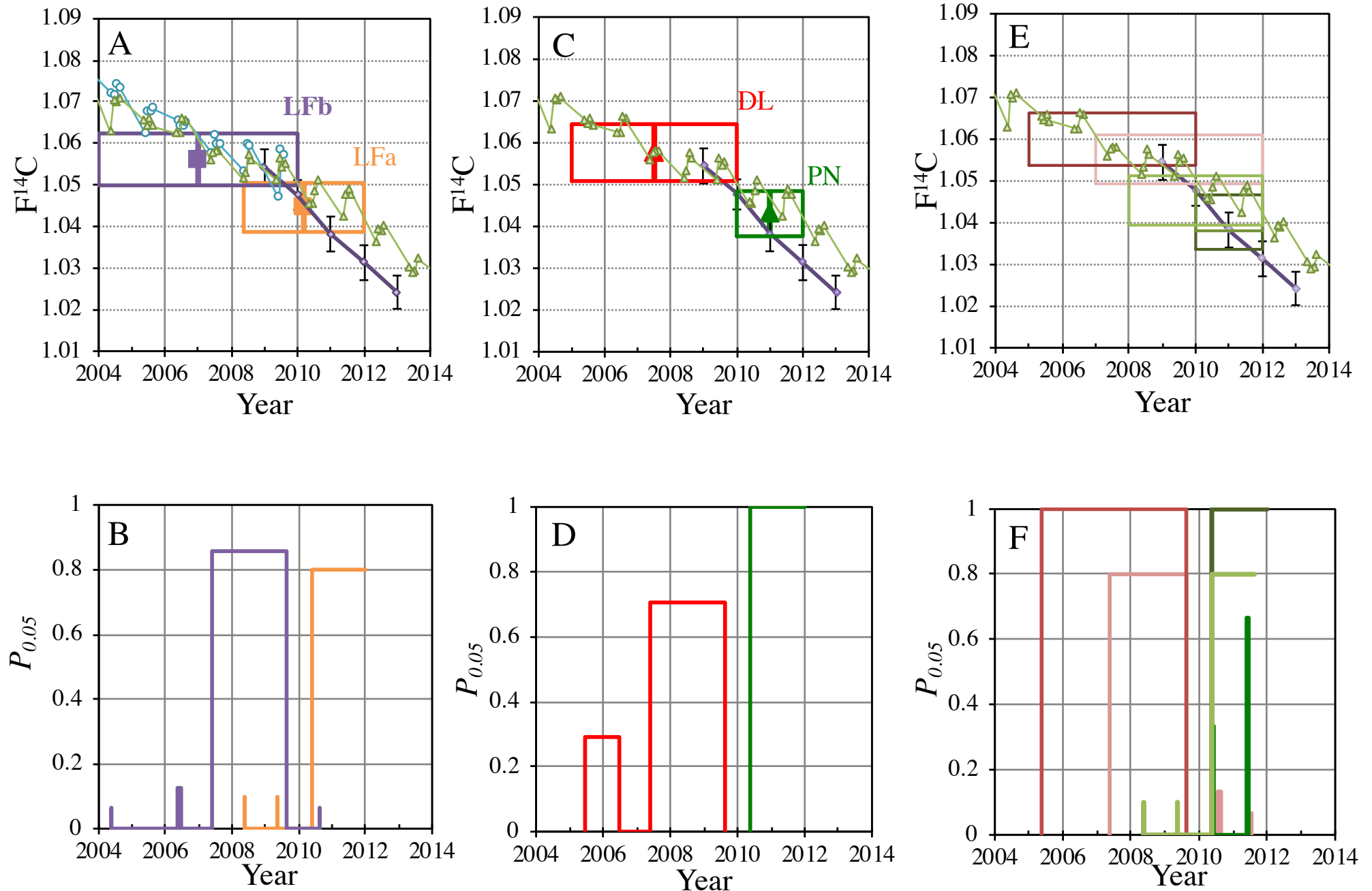


Figure 2

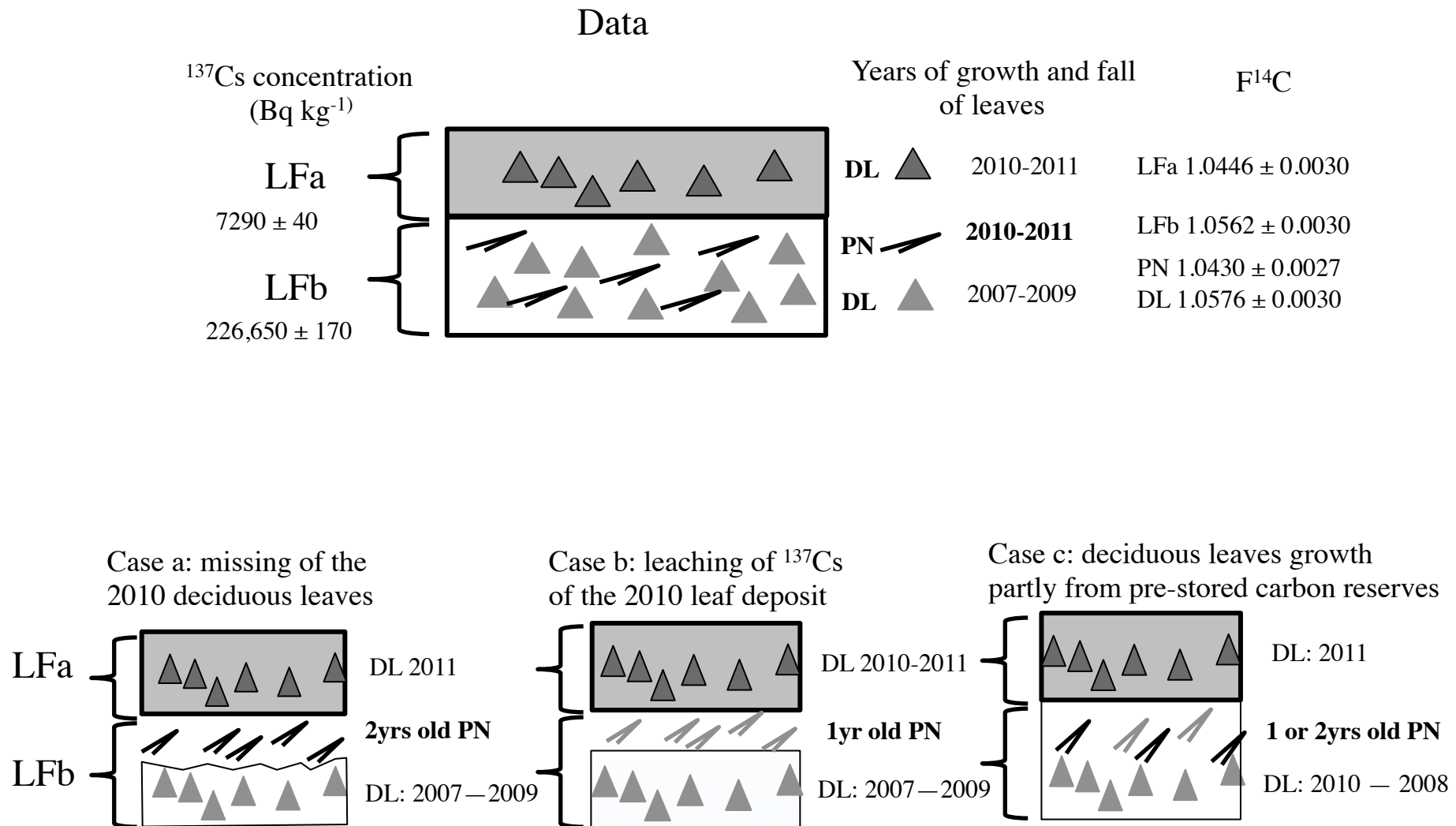


Figure 3

Litter Fraction	Sample Ref.*	Sample type	Lab n° [chemistry/measurement]	C %	$\delta^{13}\text{C}$ [‰]	$\text{F}^{14}\text{C}$ ( $\pm 1\sigma$ )	$^{137}\text{Cs}$ Bq kg <sup>-1</sup>
LFa	FKV-046A-1	upper litter bulk	GifA15936/SacA44607	45.8	$-30.63 \pm 0.15$	$1.0446 \pm 0.0030$	$7290 \pm 40$
LFb	FKV-046B-1	lower litter bulk	GifA15938/SacA44609	43.3	$-31.62 \pm 0.15$	$1.0562 \pm 0.0031$	$226,650 \pm 170$
LFb	FKV-046B-1&2	leave & fungi	GIFA17276/ECHo 1714			$1.0600 \pm 0.0031$	
LFb	FKV-046B-1	clean leave	GIFA17277/ECHo 1715			$1.0551 \pm 0.0029$	
					<i>average</i>	$1.0576 \pm 0.0034$	
LFb	FKV-046B-1	pine needle 1 & fungi	GIFA17274/ECHo 1717			$1.0401 \pm 0.0033$	
LFb	FKV-046B-2	pine needle 2 & fungi	GIFA17361/ECHo 1863			$1.0453 \pm 0.0030$	
LFb	FKV-046B-2	clean pine needle 2	GIFA17360/ECHo 1862			$1.0437 \pm 0.0028$	
					<i>average</i>	$1.0430 \pm 0.0027$	

Table 1

## Supplementary material

Radiocarbon and radiocesium in litter fall at Kawamata, ~45 km NW from the Fukushima Dai-ichi nuclear power plant (Japan)

Martine Paterne Correspondent<sup>1</sup>, Olivier Evrard<sup>1</sup>, Christine Hatté<sup>1</sup>, J. Patrick Lacey<sup>1,2</sup>, Julius Nouet<sup>3</sup>, Yuichi Onda<sup>4</sup>

<sup>1</sup>Laboratoire des Sciences du Climat et de l'Environnement, LSCE/IPSL, CEA-CNRS-UVSQ, Université Paris- Saclay, Domaine du CNRS, F-91198 Gif-sur-Yvette, [martine.paterne@lsce.ipsl.fr](mailto:martine.paterne@lsce.ipsl.fr), [olivier.evrard@lsce.ipsl.fr](mailto:olivier.evrard@lsce.ipsl.fr), [christine.hatte@lsce.ipsl.fr](mailto:christine.hatte@lsce.ipsl.fr),

<sup>2</sup> Environmental Monitoring and Science Division, Alberta Environment and Parks, 3115 – 12 Street NE, Calgary, Alberta, Canada, [Patrick.Lacey@gov.ab.ca](mailto:Patrick.Lacey@gov.ab.ca)

<sup>3</sup> UMR GEOPS 8148 - Géosciences Paris Sud, Université Paris Sud/CNRS, Rue du Belvédère, F-91400 Orsay, [julius.nouet@u-psud.fr](mailto:julius.nouet@u-psud.fr)

<sup>4</sup> Center for Research in Isotopes and Environmental Dynamics (CRIED), University of Tsukuba, Tsukuba, Japan, [onda@geoenv.tsukuba.ac.jp](mailto:onda@geoenv.tsukuba.ac.jp)

### Text S1. Contamination of the deciduous leaves in litter fraction LFb by younger deciduous leaves.

We calculated the effect of contamination (in %) on the time interval of deposition of the litter fraction LFb using 5%, 10% and 15% of leaves from the upper fraction of year 2010. The time interval of age calibration was calculated using the seasonal SIL atmospheric <sup>14</sup>C in 2010 and the mean value of the May-June SIL atmospheric <sup>14</sup>C between 2007 and 2009, which are very similar (Figure 2). The time interval and probability of calibrated <sup>14</sup>C values with a 5% contamination of LFb deciduous leaves by those of year 2010 exactly mirror that of the clean deciduous leaves in LFb at the 95% confidence level. The 10% and 15% contamination obviously enlarge the calibrated age intervals of the LFb deciduous leaves with a greater probability of the young ages (Fig. S1).

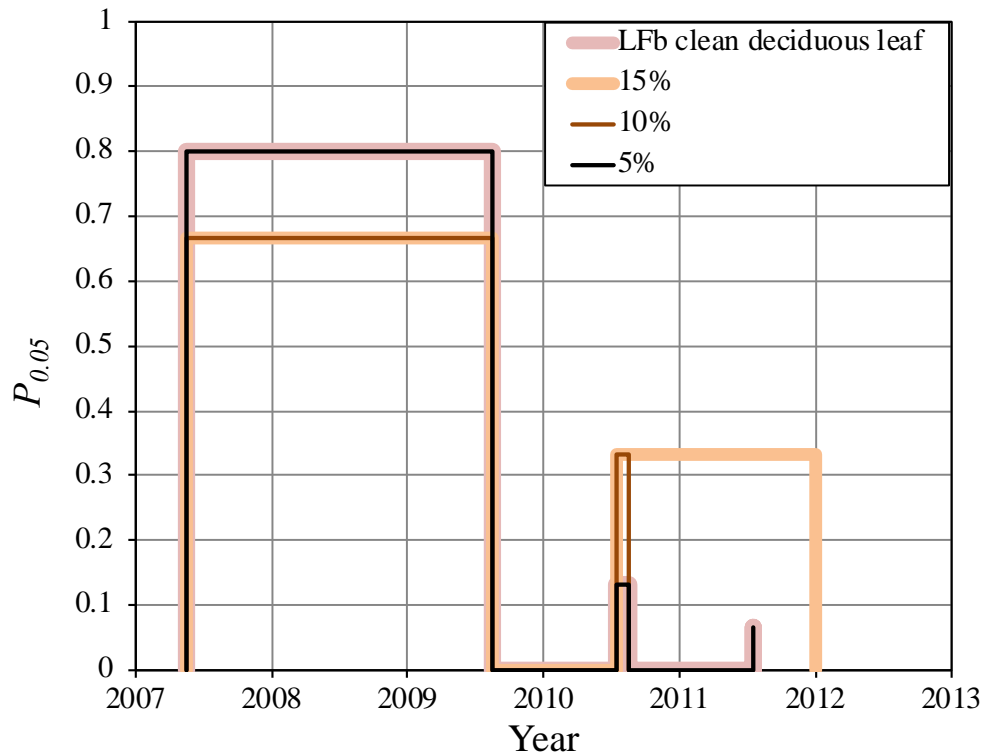


Fig. S1: Probability ( $P_{0.05}$ ) of the calibrated age intervals of the clean deciduous leaf in LfB compared to those of deciduous leaves growing and falling between 2007 and 2009 mixed with 5%, 10% and 15% of deciduous leaves of year 2010. The calculation is based on Chi-square tests between the mean seasonal (May-August) SIL atmospheric  $^{14}\text{C}$  between 2007 and 2009 and that of year 2010 [34, 35].

Text S2. The April-September age calibration of  $^{14}\text{C}$  values of leaf material deposited at Kawamata Town mixed forest

The May-August age calibration of  $^{14}\text{C}$  values of leaf material, using the May-August atmospheric  $^{14}\text{C}$  of the *clean* air  $^{14}\text{CO}_2$  at the Schauinsland/Jungfrau stations (SIL) [34, 35], indicate that the deciduous leaves in LFa and LfB deposited between 2010 and 2011 and between 2007 and 2009, respectively. Extending the seasonal age calibration to April-September does not change significantly the timing of leaf deposit in the litter fractions, and the pine needles and deciduous leaves in LfB have incompatible ages (Fig. S2).

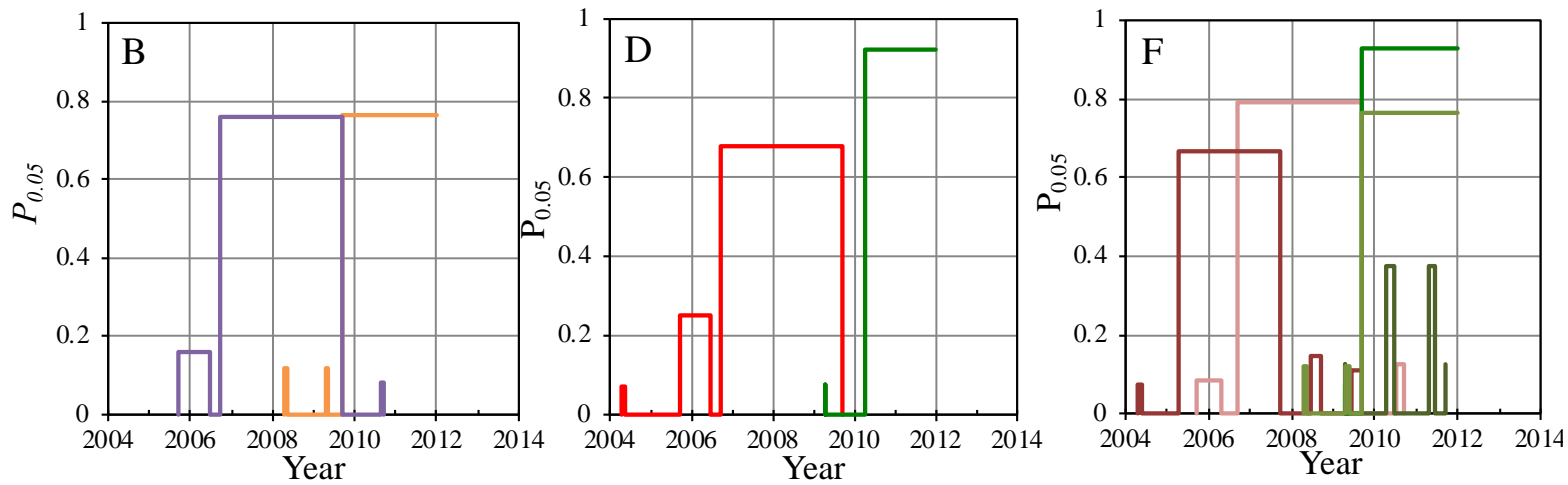
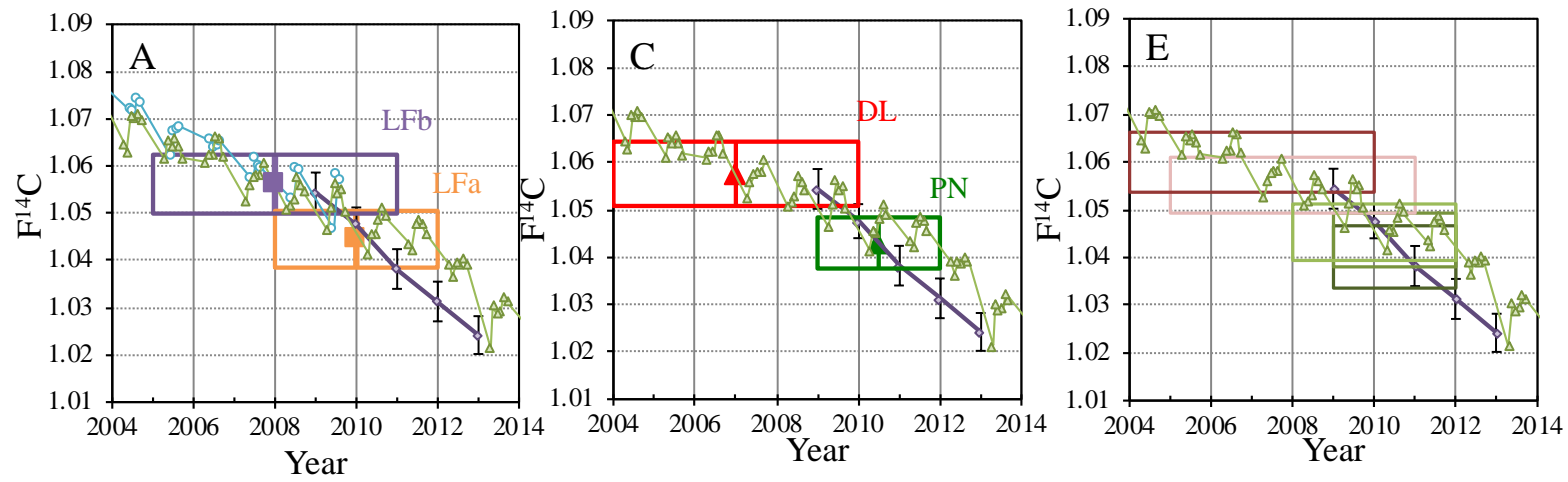


Fig. S2: Seasonal age calibration of litter material and probability (in %) at the 95% confidence level as a function of time (in years). Calibration and probabilities were obtained using Chi-square tests between the values of  $^{14}\text{C}$  (Fraction modern,  $F^{14}\text{C}$ ) of the litter material and those of the seasonal (April-September) atmospheric  $^{14}\text{C}$  at the Schauinsland/Jungfrau stations (SIL) [34, 35] (green triangles).  $P_{0.05} \leq 5\%$  were not taken into account. Values of  $F^{14}\text{C}$  are also reported for the stack of atmospheric  $^{14}\text{C}$  data (May-June) of the Northern Hemisphere Zone 2 (NHZone2) [36] (blue circles) and for the annual tree-ring record at Yamakiya (violin diamonds) [4]. Colors on the probability diagrams referred to those of the time intervals (rectangles) of  $^{14}\text{C}$  calibration of the litter fractions LFa and LFb (A,B), of the mean  $^{14}\text{C}$  of deciduous leaves (DL) and pine needles (PN) in LFb (C,D), and of individual deciduous leaves (red and rose) and pine needles (dark, medium and light green) in LFb (E,F).