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ABSTRACT

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

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

1. Introduction

Boiling water reactor (BWR) nuclear power plants, such as the Fukushima Dai-ichi Nuclear Power Plant (FDNPP), contribute to a release of ^{14}C into the atmosphere in the gaseous form of $^{14}CO_2$ during routine operations [1]. The excess ^{14}C is well recorded in vegetation, such as tree-ring and leaves, in the vicinity of such BWR nuclear plants [2; 3-7]. Monitoring this excess ^{14}C is of biological significance because of the rapid exchange of the $^{14}CO_2$ within the terrestrial carbon reservoirs and its long residence time (T=5730 years) [1].

Significant leakage of radionuclides occurred at the time of the FDNPP accident on 2011, March 11th. Among these radionuclides, post-accidental research has focused on the gamma-emitting radionuclides [8], and mainly ^{137}Cs. A heavily contaminated area by the ^{137}Cs fallout, ~70% of which is covered by mixed forests, was identified downwind of the power plant in the prevailing northwestward wind direction in a variety of environments, including lakes, rivers, agricultural soils and forest ecosystems [9-17]. In contrast, the ß-emitter ^{14}C has been understudied after the FDNPP accident [8], although it has provided clues on the dispersion patterns of atmospheric $^{14}CO_2$ release from nuclear fuels reprocessing plants in Japanese context [18-20] and on the long-term fate of Fukushima–derived ^{137}Cs in forest soils similar to those found in the Fukushima prefecture from the comparative analyses of bomb-produced ^{14}C and ^{137}Cs migration in soils [21]. High ^{14}C activities were measured from tree ring analyses of a Japanese cedar in the local environment (~2 km far from FDNPP), although revealing a reverse pattern between the early and late wood of year 2011 [4,7]. The activity of ^{14}C was indeed higher in the late wood than in the early wood, while the seasonal transition commonly occurs between mid-June and mid-July [7]. The ^{14}C activities decreased to background levels at 14 km northwest of the FDNPP in the highly contaminated region while the dispersion pattern of ^{14}C in the southward direction remained unclear [3-5, 7]. A small enrichment of ^{14}C, linked to a ^{14}C release during the accident, has been noted in the late wood of the 2011 tree-ring from the Iwaki area located at 50 Km south of the FDNPP [3].

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

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

2. Material and Methods

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

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

Prior to AMS-^{14}C analyses, the bulk litter samples were first crushed (≤1mm). The classical Acid-Alkali-Acid (AAA) pre-treatment of routine samples of organic matter was applied to eliminate potential contamination. Transformation into CO_2 was described elsewhere [27] and was performed at the Gif-LSCE Radiocarbon Laboratory. Graphite targets were prepared and measured at the French National AMS-ARTEMIS-LMC14 facility [28]. The % C in litter is obtained from the mass of sample and from laboratory calibration between pressure of CO_2 of burned sample and the mass of carbon. Individual deciduous leaves and two different pine needles picked into litter LFb were also analyzed. Contiguous samples were cut on one of the pine needle, one sample including fungi and the other representing *clean* leaf. Samples of deciduous leaves were obtained from one single free-fungi leaf and from several pieces of deciduous leaves including fungi. After the classical AAA pre-treatment, aliquots containing about 1mg of carbon were weighted into tin boats and processed using the automated graphitization equipment (AGE3) [29]. Measurements were performed on the MICADAS equipment at the Gif-LSCE Radiocarbon Laboratory [30, 31]. The $^{14}C/^{12}C$ activity of the samples is $\delta^{13}C$-normalized and expressed as Fraction modern [32, 33]. The ^{14}C content of litter material were compared with those of *clean* air values of $^{14}CO_2$ as measured at the Schauinsland/Jungfrau stations (SIL) [34,35] and with the stack of atmospheric ^{14}C data of the Northern Hemisphere Zone 2 (NHZone2) [36] to analyze the timing of deposition of leaf material in the two litter fractions.

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

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

3. Results

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

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

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

3. Discussion

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

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

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

3.1 Disturbance of the leaf fall deposits

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

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

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

3.2 The carbon paths during deciduous leaf growth

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

We estimated the ^{14}C values of the deciduous leaves when accounting for a turnover time of stored carbon reserves during the growth season and their use for deciduous leaves growth during the following year using:

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

with t the duration of the growth season and τ the turnover time in days, and A/A_o the ratio of the atmospheric ^{14}C activity during the considered years.

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

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

4. Conclusion

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

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

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

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

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Table Captions

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

Plates

Plate 1**:** Microscope Photographs of the leaves accumulated in litter layer L (A-C) and layer F (D-F) showing no visible sign of degradation of the leaves. The scales (white bar) are 1cm in A, B and D; 0.5 cm in E; 30µm in C and 50µm in F. On image F, minerals are, magnetite, hematite and feldpars.

Fig. Captions

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

Fig. 2: 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}C (Fraction modern, $F^{14}C$) of the litter material and those of the seasonal (May-June) atmospheric ^{14}C at the Schauinsland/Jungfrau stations (SIL) [34, 35] (green triangles). $P_{0.05} \leq 5\%$ were not taken into account. Values of $F^{14}C$ are also reported for the stack of atmospheric ^{14}C data (May-June) of the Northern Hemisphere Zone 2 (NHZone2) [36] (blue circles) and for the annual tree-ring record at Yamakiya, nearby Kawamata Town (violin diamonds) [4]. Colors on the probability diagrams referred to those of the time intervals (rectangles) of ^{14}C calibration of the litter fractions LFa and LFb (A,B), of the mean ^{14}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).

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

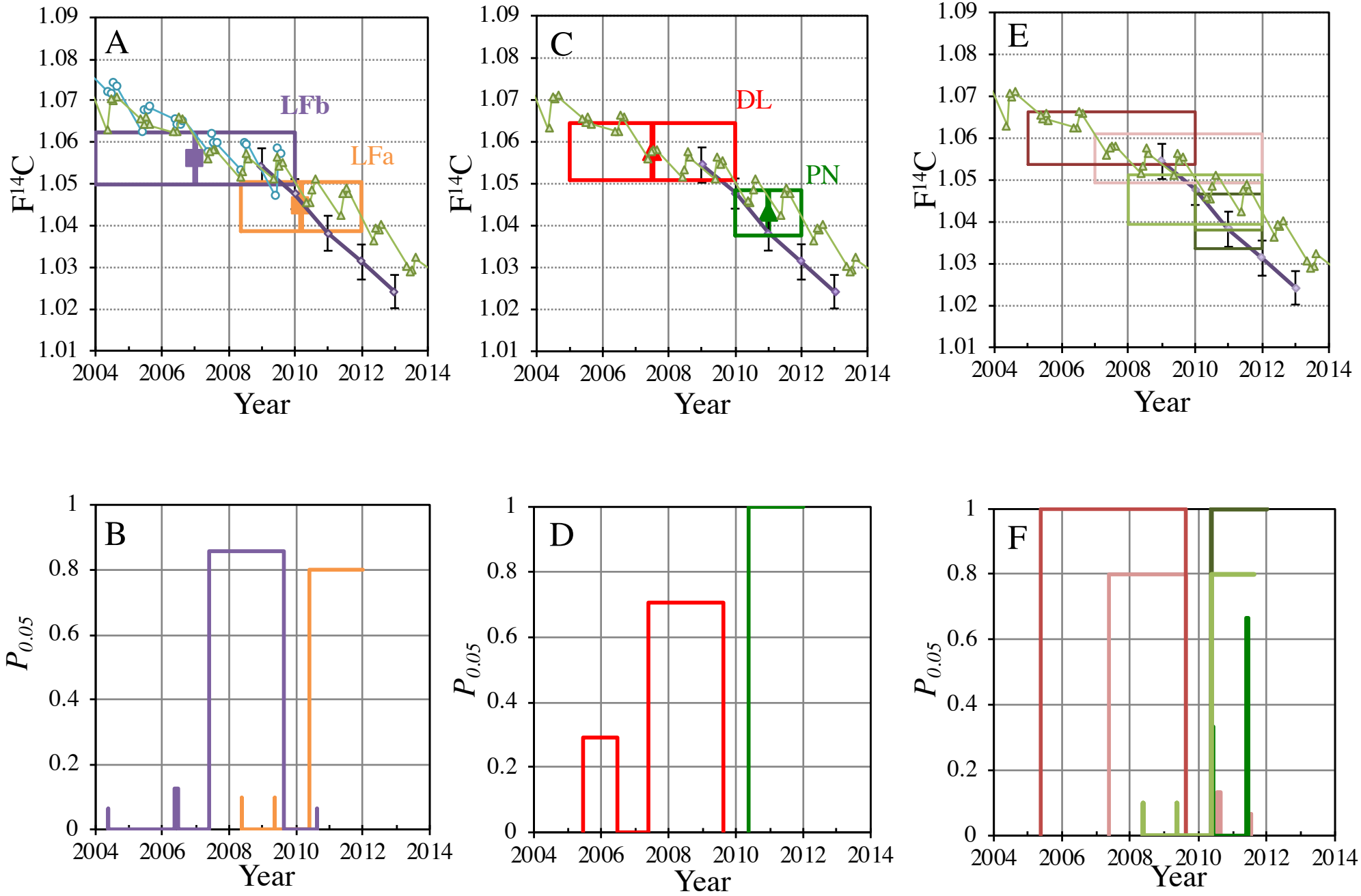

Figure 2

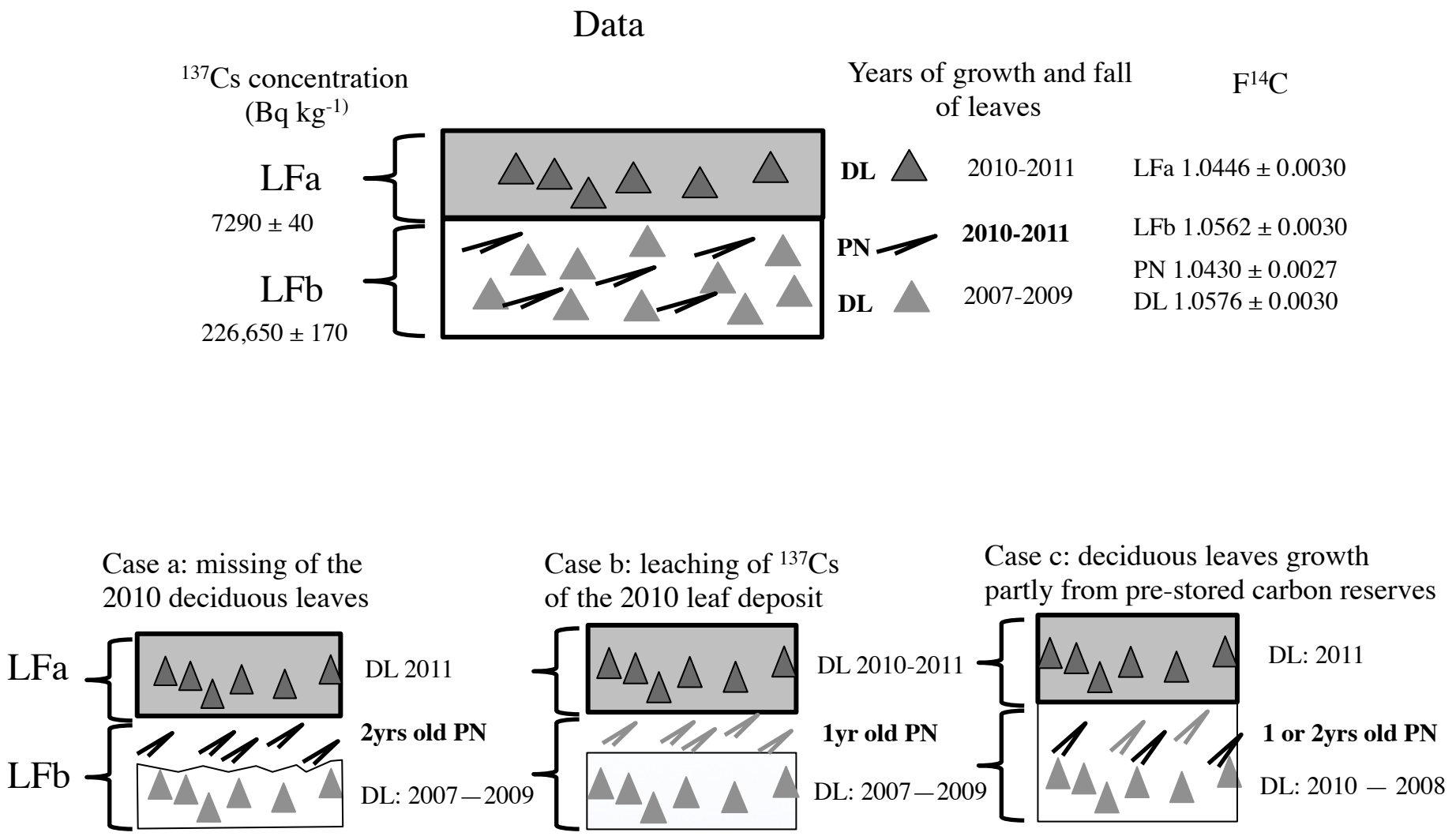

Figure 3

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

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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 ^{14}C in 2010 and the mean value of the May-June SIL atmospheric ^{14}C between 2007 and 2009, which are very similar (Figure 2). The time interval and probability of calibrated ^{14}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).

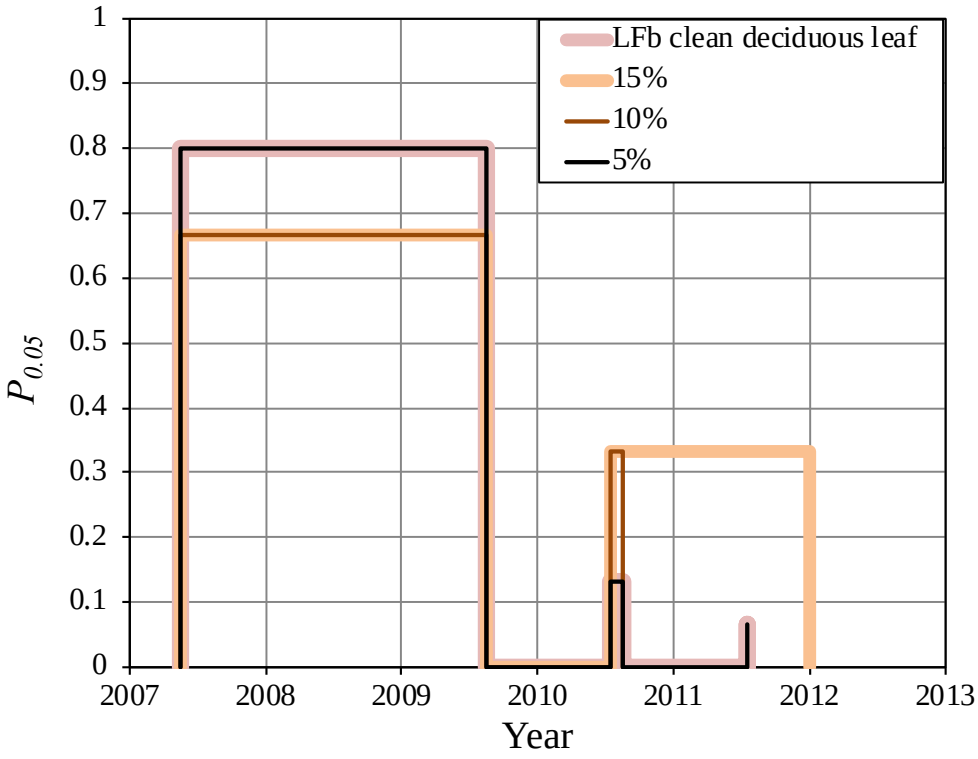

Fig. S1: Probability ($P_{0.05}$) of the calibrated age intervals of the clean deciduous leaf in LFb compared to those of deciduous leaves growing and falling between 2007 and 2009 mixed with 5%, 10% and 15% of deciduous leaves of year 2010. The calculation is based on Chi-square tests between the mean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

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

The May-August age calibration of ^{14}C values of leaf material, using the May-August atmospheric ^{14}C of the *clean* air $^{14}CO_2$ 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).

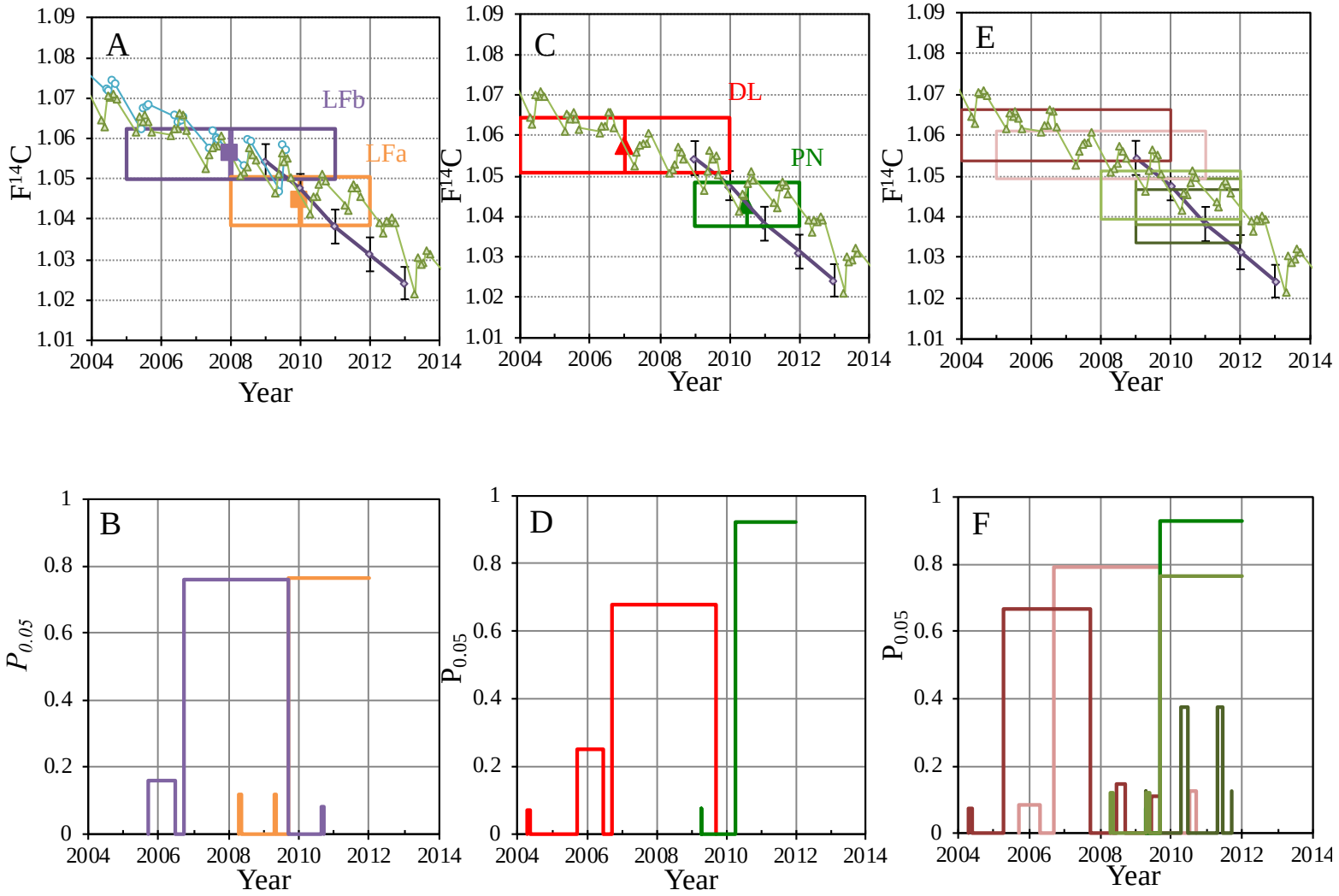
A
B
C
D
E
F
LFb
LFa
DL
PN
F14C
P0.05
Year
1.09
1.08
1.07
1.06
1.05
1.04
1.03
1.02
1.01
1
0.8
0.6
0.4
0.2
0
2004
2006
2008
2010
2012
2014

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}C (Fraction modern, $F^{14}C$) of the litter material and those of the seasonal (April-September) atmospheric ^{14}C at the Schauinsland/Jungfrau stations (SIL) [34, 35] (green triangles). $P_{0.05} \leq 5\%$ were not taken into account. Values of $F^{14}C$ are also reported for the stack of atmospheric ^{14}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}C calibration of the litter fractions LFa and LFb (A,B), of the mean ^{14}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).