Radiocaesium partitioning in Japanese cedar forests following the “early” phase of Fukushima fallout redistribution

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Our study focused on radiocaesium (137Cs) partitioning in forests, three vegetation periods after the Fukushima Daiichi nuclear power plant accident. 137Cs distribution in forest components (organic and mineral soil layers as well as tree compartments: stem, bark, needles, branches and roots) was measured for two Japanese cedar stand ages (17 and 33 years old). The results showed that around 85% of the initial deposit was found in the forest floor and topsoil. For the youngest stand almost 70% of the deposit is present in the forest floor, whereas for the oldest stand 50% is present in the 0–3 cm mineral soil layer. For trees, old and perennial organs (including dead and living needles and branches, litter fall and outer bark) directly exposed to the fallout remained the most contaminated. The crown concentrated 61–69% of the total tree contamination. Surprisingly the dead organs concentrated 25 ± 9% (young cedars) to 36 ± 20% (mature cedar) of the trees’ residual activity, highlighting the importance of that specific compartment in the early post-accident phase for Japanese cedar forests.

Although the stem (including bark) represents the highest biomass pool, it only concentrates 3.3% and 4.6% of the initial 137Cs deposit for mature and young cedars, respectively.

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident in March 2011 released between 380 and 800 PBq of volatile radionuclides (iodine, tellurium and caesium) into the atmosphere, 20% of which were spread across the Japanese terrestrial environment¹. To date, caesium radioisotopes 137Cs and 134Cs dominate in contaminated ecosystems due to their physical half-lives, (30.2 y and 2.1 y, respectively). Forest ecosystems composed of evergreen species, mainly Japanese cedar (Cryptomeria japonica), Japanese Cypress (Chamaecyparis obtusa) and deciduous species cover almost 75% of the highest contaminated area (>5 mSv y⁻¹)². Since the Chernobyl accident, the importance of forest radioactive contamination as a significant source of local radiation exposure has been recognised³, and several studies have shown a clear and persistent high level of contamination in different forest products⁴. Owing to its longevity and high standing biomass and turnover, forest vegetation can efficiently trap and recycle radioactive fallout, resulting in the potential for enhanced external and internal exposures over timescales spanning from decades to centuries⁵. After the initial deposit, the observed 137Cs (or 134Cs) contamination of the tree components is in fact the result of various processes such as interception, foliar absorption, internal translocation, root uptake and immobilisation in perennial parts⁶. Over a timescale of decades or longer, it is expected that long-lived radionuclides will be incorporated into natural elemental cycles, especially in elements such as strontium and caesium due to their chemical analogy with major nutrients (i.e. Ca and K, respectively). In multi-layered forest soils, the possible long-lasting availability of radiocaesium in particular is due to its slow vertical migration and its persistence in surface organic layers⁷–⁹. In 137Cs contamination dynamics, two stages can be distinguished: (1) the “early” phase lasting 4–5 years, in which the depuration processes of the tree crown
predominate, characterised by the rapid redistribution of the initial deposits between trees and soil (via weathering, throughfall and litterfall), and (2) an apparent “steady state” phase characterised by the stabilisation of radioactivity transfers between soil and tree, with root uptake mainly controlling the extent of further 137Cs cycling. Hence, a good understanding of the short- and long-term dynamics of radiocaesium in forests is essential to predicting its behaviour. The type of 137Cs deposits (dry and/or wet) and the environmental conditions (climate, species, edaphon, topography) found near the site of the Chernobyl and Fukushima disasters are different and are likely to generate a distinctive contamination redistribution within the forest environment. For instance, given the predominance of mountainous land in Fukushima, the role of the forest as a potential secondary source of contamination dissemination to other parts of the environment by runoff and erosion processes was examined. In this context, monitoring contaminated forest stands is essential to providing local datasets that will allow flows between forest ecosystem compartments and driving parameters to be quantified.

Many studies in the Fukushima contaminated area related to early 137Cs distribution in the Japanese forest ecosystems. Most of these studies began immediately after the accident (main data from 2011 to 2012) and showed that for the evergreen species, interception by canopies was very high (60–90%) due to high canopy closure. Depending on the tree species and on the renewal of the canopies, contamination return to the forest floor was mainly supported by litterfall and rainfall. According to those observations, canopy self-decontamination is now expected to stabilise. However, there are few detailed budgets on activity partitioning for this transition period, especially those taking into account the soil-tree system as a whole.

The scientific programme launched in November 2013 aims to provide relevant parameters for modelling radiocaesium transfers in forest ecosystems at different timescales, including short to long term. Monitoring of contaminated forest stands in Japan was initiated to cover the whole forest cycle of radiocaesium over 6 years. This study describes the radiocaesium inventories taken in two Japan forest stands (i.e. three vegetative periods after the fallout) at stand scale. Its objective is to provide an initial estimate of the 137Cs distribution between soil and tree and also within the tree using a robust sampling method.

**Materials and Methods**

**Site description.** The characteristics of the monitoring sites have been detailed in previous studies. Briefly, the stands are located approximately 40 km northwest of FDNP in the Yamaki District (Kawamata Town) and consist of two contiguous even-aged Japanese cedar stands: one young (YC, 17 years in 2013) and one more mature (MC, 33 years in 2013). The main characteristics of the forest plots are provided in Table 1.

**Field sampling and measurements.**

**Trees sampling.** The two stands (YC (MC)) were characterised in November 2013 by measuring the diameters at breast height (DBH, 1.30 m above the ground) within the perimeter of defined plots (n = 160 trees for MC, n = 214 trees for YC) and the densities were calculated per hectare. The normality of the DBH distributions of the two stands was statistically tested under R environment (Shapiro-Wilke test, Development Core Team) and showed a normal distribution for the YC stand (p-value = 0.64) and a log-normal distribution for MC (p-value = 0.25). The populations of each stand were divided into nine DBH classes of equal size (i.e. the same number of trees in each class, n = 18 for MC, n = 24 for YC) and one tree was selected per size class to be cut and sampled. The height of each tree was measured after tree felling. The following methodology was adapted from Picard et al. Branches were separated from the trunk and the needles from the branches (living and dead organs). The crown compartments of the three median class trees (classes 4, 5 and 6) were all weighed (living needles and branches) to obtain the total biomass per compartment. A representative aliquot of tree compartments (i.e. organs) was then sampled: living branches (branches > 1 year), twigs (current-year living branches), old living needles (cohorts 2 years and more), current-year living needles and dead organs (branches and needles). The stems were then cut into 1 m logs to be weighed. All the logs were weighed (fw) and biometrics (diameters at each end and lengths of the 1 m logs) were measured to calculate the stem volumes according to a successive truncated cone model.

Along each stem, 2–4 cm wood discs were cut from the cutting plane to the top every 4 m for MC trees and every 2 m for YC trees. Each disc was then weighed, and the biometrics were measured according to two axes (thickness, diameter, width of outer bark, inner bark, sapwood and heartwood) and the age of each tree was checked (cutting plane of the wood disc). Then, from one quarter of each wood disc, each previously mentioned compartment was pooled for each tree. The volume of a weighed aliquot (fw: fresh weight and dw: dry weight) from each sampled organ was measured and densities were determined.

Lastly, the roots of the cut trees from the three median classes of each stand were sampled. The attached soil was removed by gently rinsing with water and they were then separated into fine (<2 mm) and coarse (>2 mm) roots. Understory roots were fully distinguished from tree roots. Samples were taken until a depth of 40–50 cm below the litter layer which contains more than 80% of the fine roots. All fresh weights were determined just after sampling and all samples were dried at 70°C until constant weight (dw) was reached. The dry samples were then homogenised and crushed in a blender (Waring® CB-15T) and stored in sealed bags for further analyses.

**Soil sampling.** Five-layer sampling was performed (n = 7/plot): litter layer, organic fragmented layer and three soil layers according to depth (soil 0–3 cm, soil 3–8 cm and soil 8–20 cm). The organic humified layer was not clearly identified, and was therefore not distinguished from the organic fragmented layer. The litter and organic fragmented layers were all sampled using a 545 cm² aluminium frame (19.4 × 28.1 cm). This sampling method was repeated twice for each sampling location (total sampling area 1,090 cm²), then the litter and organic fragmented materials were kept in sealed plastic bags to be weighed. Below the litter layer which contains more than 80% of the fine roots, an undisturbed soil profile and two open plastic boxes (720 cm³ volume, 9 × 4 × 20 cm each) were placed in the soil, with the top of the box corresponding to the top of soil 0–3 cm. When the boxes were removed the soil...
0–3 cm, soil 3–8 cm and soil 8–20 cm heights were measured, then carefully separated, weighed (fw) and homogenised for each corresponding layer. The collected samples were dried at 105 °C until constant weight was reached. The fresh and dry surface densities (kg m\(^{-2}\)) for the five layers and the volumetric densities (kg dm\(^{-3}\)) for the three soil layers were determined. The dry samples were crushed with a mortar for further analyses.

**Radiocaesium measurements.** An aliquot of the crushed samples was transferred into polystyrene containers for gamma spectrometry analyses. One part of the radiocaesium measurements (\(^{134}\)Cs and \(^{137}\)Cs) was taken at Fukushima University while the other part was taken at the Institute for Radiological Protection and Nuclear Safety (IRSN) in Cadarache. At Fukushima University, the samples were packed into 100-mL polystyrene containers (U-8 geometry) and analysed using an ultra-pure germanium gamma spectrometer (ORTEC GEM40–76, P type, relative efficiency 44%). At IRSN, the samples were packed into 17-mL polystyrene containers (K geometry) and analysed using an ultra-pure germanium gamma spectrometer (Canberra EGPC 42-190-R, P type, relative efficiency 41%). The accuracy of the \(^{137}\)Cs measurement intercomparisons carried out on 30% of samples was less than 5%, which is below the measurement uncertainties (6–8%). All the activities were decay corrected to the 11th of March 2011.

**Calculation of the biomass and its distribution in tree compartments.** To obtain the total biomass at stand scale, the allometric equation and parameters published by Lim et al.\(^{27}\) for living organs were applied to all trees in the plots after checking the correlation between the predicted values and the measured biomass of the organs of the cut trees (stem total (n = 18), foliage and branches (n = 6) and the values for the bark (n = 18) and the stem wood (n = 18) based on their respective biomass distribution measured in wood discs) (Fig. 1).

The distributions of the stem compartment (outer bark, inner bark, sapwood and heartwood) biomasses were calculated using the successive truncated cone model (circular section), calibrated by specific biometrics on wood discs. These distributions were validated by comparing aggregated modelled biomasses of stem organs with measured stem biomasses (mean difference of 5%, maximum difference of 12%) (Fig. 1).

The biomasses of the root compartments were calculated using the allometric equations reported by Fujimaki et al.\(^{26}\) for total biomass (W = a + bD\(^{2}\), where W is the dry biomass in g, D is the DBH in cm, a = 1,198.17 and b = 85.044), and the distribution between fine (<2 mm) and coarse (>2 mm) roots was derived from Fujimaki et al.\(^{26}\) data according to the age of the stand.

With regard to the dead organs (branches and needles) of the MC stand, the allometric equations published by Yoshida and Hijii\(^{28}\) were used (W = a (D\(^{3}\))\(^{b}\), where W is the dry biomass in g, D is the DBH in cm, a = 95.7 and b = 0.728 for dead needles, a = 16.1 and b = 0.970 for dead branches). The parameters of these allometric equations were obtained from a specific stand with dendrometric characteristics that are very similar to those of the MC stand. For the dead organs (branches + needles: dead material in-crown) of the YC stand, exhibiting different dendrometric characteristics, we used a regression model, plotting the log-transformed literature values reported

| Site          | Altitude, m | Annual precipitation (mm) | Mean daily temperature (min, max) (°C) | Nature of soil Type | \(^{137}\)Cs contamination deposit (kBq m\(^{-2}\)) |
|---------------|-------------|----------------------------|----------------------------------------|---------------------|-----------------------------------------------|
| Japanese cedar | 550         | 1,248a                     | 12.4 (−9.7/37)$^a$                     | Andosol             | 442 ± 30$^b$                                  |

**Table 1. Main characteristics of sampling plots.** \(^{a}\)from JMA, Nihonmatsu station, year 2013, \(^{b}\)from Loffredo et al.\(^{23}\), \(^{c}\)raw values without clumping correction measured with a Li-Cor Plant Canopy Analyzer LAI2000.
A mean value regression curve was obtained from fitted value regression: dead material in-crown = aH^b, where a = 0.0361, b = 2.2, R^2 = 0.80, F-statistic on 1 and 7 DF, p-value = 0.001. The relative abundance of dead needles versus dead branches (1.4) was calculated according to Yoshida and Hijii. Details of the dead biomass calculation are given in Supplementary information online.

Calculation of the radiocaesium inventory in the forest ecosystem compartments. The 137Cs inventories (Bq m^{-2}) in the compartments of each forest plot were calculated by multiplying the measured 137Cs concentrations (Bq kg_{dw}^{-1}) in the different tree organs or soil layers by their corresponding biomass or surface density (kg \text{ dw m}^{-2}) respectively. For tree compartments, 137Cs concentrations were not found to be dependent upon DBH (data not shown), and the mean values of the 137Cs concentrations were used.

Only 137Cs data are presented in this paper because the average ratio between 137Cs and 134Cs measurements was calculated, which did not differ from 1 at the date of reference (1.06 ± 0.15, data series not shown) and corresponded well to previous reported values. The delta method was used to allow for the propagation of error in the calculation chains.

Results and Discussion

Distribution of tree biomass and soil surface densities. The obtained tree biomass and soil surface densities (kg_{dw} m^{-2}) are reported in Table 2. As expected for tree compartments, the stem is the main one representing 69–71% of the above-ground biomass for YC and MC respectively. The living foliage (current and old needles) and living branch biomasses respectively represent 9% and 8% for MC and 12% and 9% for YC. This slight difference between stands is due to the growth dynamics associated with recent canopy closure for YC. Biomass and heartwood contributions are naturally different between both stands (MC > YC), and the contribution of the bark compartment is evidently lower for MC. It is interesting to note that the biomass of dead needles and dead branches remaining in the crown is high, accounting for almost 43% (MC) and 22% (YC) of the biomass of the respective living organs (dead needles: 4% and 3%; dead branches: 4% and 2% of the above-ground biomass for MC and YC, respectively). The roots represented 20% and 23% of the total tree biomass for MC and YC respectively. For the soil layers, the surface density of the organic layers is 60% higher in the YC stand compared with the MC stand (2.5 compared with 1.5 kg_{dw} m^{-2}), whereas the mineral soils have the same densities. This difference between stands is probably correlated to annual litterfall amounts as suggested by the stands’ in-crown dead biomass turnover of stands (0.35 y^{-1} and 0.88 y^{-1} for MC and YC, respectively).

137Cs massic activities in the forest compartments. The mean values of 137Cs activities (±1SD) for the various forest compartments are tabulated in Table 3 and displayed in Fig. 2. In a first approximation, no obvious difference in contamination distribution was detected between MC and YC stands. As regards trees organs, Fig. 2 depicts the ranges of measured values and shows that three vegetation periods after the Fukushima fallout the organs directly exposed to 137Cs deposits, even partially, (litterfall, dead needles, old needles, living branches, dead branches and outer bark) still exhibit the highest concentrations in both plots. The tree crown remains the most contaminated compartment compared with other tree parts (i.e. stem and roots) suggesting that the early phase,
involving predominant crown depuration processes, is not complete three vegetation periods after the accident and may even persist for several years, with litterfall being a major source of $^{137}$Cs transfer. In our context, the contamination of the other organs not directly exposed to the initial fallout or which grew after the fallout (i.e. needles young, twigs, inner bark, sapwood, heartwood and roots) can be considered a consequence of internal transfers which were shown to be very efficient for $^{137}$Cs or processes that diffuse the contamination within/down the canopy (throughfall, rain splash, dripping, etc.). It is therefore possible than $^{137}$Cs redistribution in cedar trees through internal transfers is not yet stabilised.

### Table 2. Mean values of biomass and forest floor surface densities (kg dw m$^{-2}$) of the MC and YC stands components (n = 7 for soil layers).

| Components | MC | YC |
|------------|----|----|
| **Tree components** | | |
| Needles | | |
| Living needles | 2.77 ± 0.96 | 3.31 ± 1.16 |
| Dead needles | 1.19 ± 0.34 | 0.71 ± 0.13 |
| Branches | | |
| Living branches | 2.34 ± 0.94 | 2.44 ± 0.97 |
| Dead branches | 1.09 ± 0.42 | 0.50 ± 0.09 |
| Bark | | |
| Total | 1.29 ± 0.37 | 1.38 ± 0.39 |
| Outer bark | 0.73 ± 0.29 | 0.64 ± 0.26 |
| Inner bark | 0.56 ± 0.23 | 0.73 ± 0.29 |
| Stem wood | | |
| Total | 20.89 ± 7.41 | 18.15 ± 6.73 |
| Sapwood | 12.98 ± 6.25 | 13.57 ± 6.32 |
| Heartwood | 7.91 ± 3.98 | 4.59 ± 2.32 |
| Roots | | |
| Total | 7.20 ± 2.82 | 7.72 ± 2.94 |
| <2 mm | 0.44 ± 0.41 | 0.64 ± 0.56 |
| >2 mm | 6.76 ± 2.68 | 7.08 ± 2.74 |
| **Forest floor** | | |
| Organic layers | | |
| Litter | 0.60 ± 0.26 | 0.88 ± 0.29 |
| Fragmented layer | 0.93 ± 0.33 | 1.58 ± 0.52 |
| Mineral soil layers | | |
| Soil 0–3 cm | 6.96 ± 1.60 | 8.41 ± 1.47 |
| Soil 3–8 cm | 19.17 ± 3.13 | 20.6 ± 2.54 |
| Soil 8–20 cm | 52.36 ± 3.75 | 49.95 ± 3.08 |

### Table 3. Mean values of $^{137}$Cs concentrations measured in the samples (kBq kg dw$^{-1}$) (activities corrected to 2011/03/11, n = 9 for trees organs except n = 3 for Roots, n = 7 for soil layers).

| Components | MC | YC |
|------------|----|----|
| **Tree components** | | |
| Needles | | |
| Living needles | 7.12 ± 0.40 | 4.41 ± 0.40 |
| Old needles (>1 year) | 8.40 ± 2.90 | 4.84 ± 1.39 |
| Current needles (<1 year) | 4.48 ± 1.24 | 3.59 ± 1.18 |
| Dead needles | 21.52 ± 9.67 | 20.63 ± 3.42 |
| Branches | | |
| Living branches | 3.94 ± 1.75 | 5.81 ± 2.34 |
| Dead branches | 5.37 ± 1.35 | 10.39 ± 3.88 |
| Bark | | |
| Total | 4.57 ± 2.69 | 5.83 ± 1.32 |
| Outer bark | 6.56 ± 4.69 | 9.88 ± 2.62 |
| Inner bark | 1.96 ± 0.59 | 2.26 ± 0.68 |
| Stem wood | | |
| Total | 0.63 ± 0.43 | 0.64 ± 0.43 |
| Sapwood | 0.47 ± 0.13 | 0.56 ± 0.13 |
| Heartwood | 0.90 ± 0.32 | 0.90 ± 0.30 |
| Roots | | |
| Total | 1.11 ± 0.04 | 1.48 ± 0.06 |
| <2 mm | 1.63 ± 0.45 | 2.09 ± 0.52 |
| >2 mm | 1.08 ± 0.17 | 1.42 ± 0.31 |
| **Forest floor** | | |
| Organic layers | | |
| Litter | 39.99 ± 6.57 | 50.26 ± 5.21 |
| Fragmented layer | 88.92 ± 23.03 | 123.69 ± 25.62 |
| Mineral soil | | |
| Soil 0–3 cm | 36.66 ± 13.77 | 8.26 ± 3.56 |
| Soil 3–8 cm | 4.60 ± 2.93 | 1.08 ± 0.57 |
| Soil 8–20 cm | 0.91 ± 0.73 | 0.42 ± 0.16 |
As already observed in other monitoring studies for Japanese cedar\textsuperscript{17,18} both heartwood and sapwood compartments are contaminated. Although the contamination levels are of the same order of magnitude, a positive gradient from external (sapwood) rings to internal rings (heartwood) can be observed as a trend, implying that Cs\textsuperscript{+} cation has a strong radial mobility\textsuperscript{37}. This positive gradient of concentration from sapwood to the pith was previously observed for potassium and caesium for Japanese cedar\textsuperscript{38–40} and their accumulation in heartwood was associated with the formation of these tissues\textsuperscript{39}. This peculiarity was not observed in post-Chernobyl studies on pine trees or birches, however, for which the reverse trend gradient prevailed\textsuperscript{6,41}, i.e. an increasing concentration from the pith to the external rings.

The similar 137Cs root concentrations found in the stands, but different 137Cs vertical distributions in the soil profile, and the 137Cs inner bark concentrations suggest a potential 137Cs downward flow following foliar uptake through phloemic pathways. This assumption was corroborated by the fact that root 137Cs concentration is very similar at the two sites, even though 137Cs vertical distribution is different. While the foliar incorporation pathway is most probably predominant at this stage\textsuperscript{35,42–44} our results do not make it possible to assess the relative contributions of each pathway, foliar or root uptake, and only the 137Cs/analogue (K) or stable isotope (133Cs) ratios could be used to address this issue in future studies.

In the soil compartments, the fragmented layer was the most contaminated compartment in the investigated soil profile, at 89 and 124 kBq kg\textsuperscript{-1} for MC and YC stands, respectively (Table 3 and Fig. 2). The litter layer was less contaminated (40–50 kBq kg\textsuperscript{-1}) than the fragmented layer suggesting a decrease in 137Cs concentration within litterfall over time, as already observed\textsuperscript{22}. This assumption is also supported by comparing litter and dead needles (20 kBq kg\textsuperscript{dry-1}) activities. Although the activities are in the same order of magnitude, dead needles is less contaminated than the litter, suggesting that litter with a higher activity than dead needles is present in the litter layer. For the mineral soil layers (soil 0–3 cm, soil 3–8 cm and soil 8–20 cm), the 137Cs activities classically decreased with the sampling depth (Table 3 and Fig. 2) as has already been observed in other studies\textsuperscript{45,46}. However, a significant difference was observed between the two plots. In the MC plot, litter layer was less contaminated (40–50 kBq kg\textsuperscript{dry-1}) than the fragmented layer suggesting a decrease in 137Cs concentration within litterfall over time, as already observed\textsuperscript{22}. This assumption is also supported by comparing litter and dead needles (20 kBq kg\textsuperscript{dry-1}) activities. Although the activities are in the same order of magnitude, dead needles is less contaminated than the litter, suggesting that litter with a higher activity than dead needles is present in the litter layer. For the mineral soil layers (soil 0–3 cm, soil 3–8 cm and soil 8–20 cm), the 137Cs activities classically decreased with the sampling depth (Table 3 and Fig. 2) as has already been observed in other studies\textsuperscript{45,46}. However, a significant difference was observed between the two plots. In the MC plot, litter layer was less contaminated (40–50 kBq kg\textsuperscript{dry-1}) than the fragmented layer suggesting a decrease in 137Cs concentration within litterfall over time, as already observed\textsuperscript{22}. This assumption is also supported by comparing litter and dead needles (20 kBq kg\textsuperscript{dry-1}) activities. Although the activities are in the same order of magnitude, dead needles is less contaminated than the litter, suggesting that litter with a higher activity than dead needles is present in the litter layer. For the mineral soil layers (soil 0–3 cm, soil 3–8 cm and soil 8–20 cm), the 137Cs activities classically decreased with the sampling depth (Table 3 and Fig. 2) as has already been observed in other studies\textsuperscript{45,46}.

Kato \textit{et al.}\textsuperscript{22} reported that the initial deposit on the forest floor was lower in YC than in MC. They observed that contamination of the forest floor a few months after the accident (July 2011) was two times higher for MC than YC. This observation might be explained by a higher initial interception of fallout in the YC stand characterised by a higher LAI value (Table 1). As a result, for YC, the 137Cs concentration profiles, as depicted in Table 3, seem...
Below ground* 488.0
Above ground 79.5
Total stand 567.5

Table 4. 137Cs inventories (kBq m−2) and distribution (%) in the forest compartments (activities corrected to 2011/03/11). Including roots.

...to reflect a delayed downward transfer of 137Cs from the crown to the forest floor with a lower massic activity for soil 0–3 cm compared with MC.

Inventories of 137Cs in the forest compartments. The 137Cs inventories (kBq m−2) and corresponding relative distributions for each forest compartment are tabulated in Table 4 and depicted in Fig. 3. When considering the whole forest, the total 137Cs inventories were estimated at 568 ± 92 and 432 ± 98 kBq m−2 for the MC stand and YC stand, respectively. With consideration of uncertainties resulting from spatial variability and in the estimation of the stand-level inventories in each compartment, these results are in close correlation with the total deposit of 442 ± 30 kBq m−2 already calculated for this area23 using airborne monitoring data in May 201147.

Tree compartments. The 137Cs inventories in the above-ground parts of MC and YC stands (80 and 68 kBq m−2, respectively) contribute to 14% and 16% of the total 137Cs inventories respectively (Table 4 and Fig. 4). Not including the dead material (i.e. dead needles and dead branches), the 137Cs inventory in the above-ground parts of MC stands represents around 9% and 11% for MC and YC stands, respectively. This values are consistent with those obtained in 2013 by Kajimoto et al.17 for the above-ground parts of four Japanese cedar monitored stands (discarding dead material), with the respective 137Cs fallout deposit values released from Komatsu et al.13 the contributions varied from 6–11% of the total amount of 137Cs deposit, highlighting the importance of the dead material present in the crowns, at least during the early post-accident phase. Similar ranges of values for the same time period after 137Cs release were also published for forests contaminated by the Chernobyl accident (6–8%), but without distinguishing evergreen and deciduous species48–49.

The crown (foliage and branches) contributes to 76% (MC) and 71% (YC) of the above-ground inventories (Fig. 4). The contributions of current needles to crown inventories (5–6%) are the same for both stands (Fig. 4). Differences between stands are mostly due to respective 137Cs concentrations (vs. biomass). When focusing on dead materials, these compartments represent 17% (YC) to 31% (MC) of the total crown biomass (dw). However, contamination of these compartments alone contributes to more than 40% of the crown 137Cs inventories: 52% of crown 137Cs stocks for MC and 41% for YC. The difference of 137Cs inventory/biomass ratios for dead materials between stands (2.4 and 1.7 for YC and MC, respectively) suggests a higher interception of the FDNPP releases in YC stand. It can be explained by a higher crown biomass and LAI (Table 1) in the YC stand in parallel with its tree density, recent canopy closure and related growth dynamic. Yoshida and Hijii24, and references therein, reported that for Japanese cedar the turnover of crown dead material varied within a range of 1 order of magnitude from 0.17 y−1 to 1.9 y−1. They also reported that although needle lifespan physiologically reaches 6 years for Japanese cedar, the
Figure 3. $^{137}$Cs distribution in the forest compartments (%) of the forest plot YC and MC.

Figure 4. $^{137}$Cs relative contribution in the Japanese cedars aboveground parts of each stands (a) and in the belowground material (b).
dynamic of litterfall was mostly affected by physical factors (snow, wind). The litterfall biomass amounts (g m⁻²) recorded for the stands of this study from May 2011 to May 2012 and from October 2011 to October 2012, respectively, reached a factor of 1.5 (YC) to 1.9 (MC) between the two recording periods and accurately depict the variability of this flow. Litterfall dynamics is a sensitive parameter for modeling 137Cs redistribution during the early post-accident phase, and its inter-annual variability, especially for Japanese cedar stands, remains an issue that should be addressed, at least until foliage renewal becomes effective.

The stem wood compartments (sapwood and heartwood) account for the majority of stand biomass (from 69% for YC to 71% for MC for the above-ground biomass dw, including dead materials), but related 137Cs inventories contribute to only 15% of the above-ground 137Cs inventories. The relative contributions of heartwood and sapwood to stem 137Cs inventories differ according to the stand, since heartwood’s contribution to stem biomass is naturally smaller for younger forest stands. When comparing our 137Cs inventory data with that obtained by Kajimoto et al. for the same compartments sampled in September 2013 in Japanese cedar stands exhibiting the same biomass distribution, the same trends, i.e. a major contribution of crown contamination, can be observed. The 137Cs distribution between crowns (living needles and branches) and stems (sapwood, heartwood, outer bark and inner bark) ranges from 76–93% to 7–24%, respectively.

Although roots constitute 20% (MC) and 23% (YC) of stand biomass, this compartment accounted for only 9% and 14% of the 137Cs inventory in the trees (1% and 3% of the total 137Cs inventory) for MC and YC, respectively.

Soil compartments. The soil compartments are the largest pool of 137Cs in the forest ecosystem (85% and 82% for MC and YC, respectively, 86% and 84% if root compartments are included) and there was some difference between the two plots. Despite the fact that the fragmented layer has the highest mass concentrations in both stands (Table 2), its 137Cs inventory in MC is much smaller than that of the soil 0–3 cm layer (Table 4). This result is consistent with a higher 137Cs initial deposit on the MC forest floor and a thinner organic layer for the MC plot (1.5 vs. 2.5 kg dry m⁻² soil, Fig. 3, Table 2). The difference in organic layer thickness between the two plots generates a lower 137Cs residence time in the MC plot, either resulting from organic material degradation or leaching, as has already been mentioned. In addition, the higher initial 137Cs interception for YC generated a delayed downward transfer of 137Cs from the crown to the forest floor, and also from the organic layer to soil 0–3 cm. In the next decade, the importance of forest floor thickness is expected to play a key role in further 137Cs migration and resulting vertical distribution as evidenced in post-Chernobyl studies. Focusing on the soil 137Cs profile, we observed that for the MC plot, 22% of the 137Cs soil inventory is present in the organic layer and around 68% in the 0–8 cm mineral layers (Fig. 4). These results correlate with those obtained by Fujii et al. in 2012 for stands of the same age (18–26% in the organic layer and 68–69% in the 0–5 cm mineral layers). As shown in Fig. 4 for the YC plot, due to the higher initial interception generating a delay in 137Cs transfer from the crown to the soil compared to MC plot, the 137Cs profile (68% in the organic layer, 26% in the 0–8 cm mineral layers) is quite similar to the distribution observed immediately after the accident. Although the main stock of 137Cs is concentrated in the upper forest floor layers, we observed that 137Cs has migrated deeper than 8 cm, with 10% and 6% of the soil 137Cs budget measured in the 8–20 cm layer for MC and YC respectively (Fig. 4). In forest soils 137Cs can be highly concentrated in organic layers, or immobilised in the first surface soil layers. However, a small fraction of 137Cs is highly mobile and could quickly migrate to deeper mineral layers. These last authors concluded that the high content of organic matter vs. clay mineral compared with other land soils could be responsible for this higher mobility. This higher availability in organic soils has also already been observed. However, the impact of organic matter content on 137Cs behaviour depends on the concentration and nature of the clay mineral. In our soils, only kaolinite was found in the mineral layers (Table 1), a clay mineral known to sorb caesium to a lesser degree than illite or vermiculite. This observation corresponds with the high mobility of part of the 137Cs in our forest soils.

Even if a difference was observed between the two plots, the 137Cs remains concentrated in the organic layer or the upper mineral soil layer. Higher levels of 137Cs contamination in the organic layer compared with the mineral layers were also observed after the Chernobyl disaster, even a long time after the accident.

Conclusion

This monitoring, carried out three vegetation periods after the FDNPP accident, produced a detailed dataset on 137Cs concentrations and inventories in forests (trees and soil components). The sampling strategy (9 trees and 7 forest floor locations) was implemented to integrate the spatial variability occurring in two forest stands of different ages. Our results showed that the forest floor exhibited around 85% of the 137Cs inventories and is the major contamination reservoir three years after the accident. For the young stand (YC) with the highest initial interception generating a delay in 137Cs transfer from the crown to the soil compared to MC (18–26% in the organic layer and 68–69% in the 0–5 cm mineral layers). As shown in Fig. 4 for the YC plot, due to the higher initial interception generating a delay in 137Cs transfer from the crown to the soil compared to MC plot, the 137Cs profile (68% in the organic layer, 26% in the 0–8 cm mineral layers) is quite similar to the distribution observed immediately after the accident. Although the main stock of 137Cs is concentrated in the upper forest floor layers, we observed that 137Cs has migrated deeper than 8 cm, with 10% and 6% of the soil 137Cs budget measured in the 8–20 cm layer for MC and YC respectively (Fig. 4). In forest soils 137Cs can be highly concentrated in organic layers, or immobilised in the first surface soil layers. However, a small fraction of 137Cs is highly mobile and could quickly migrate to deeper mineral layers. These last authors concluded that the high content of organic matter vs. clay mineral compared with other land soils could be responsible for this higher mobility. This higher availability in organic soils has also already been observed. However, the impact of organic matter content on 137Cs behaviour depends on the concentration and nature of the clay mineral. In our soils, only kaolinite was found in the mineral layers (Table 1), a clay mineral known to sorb caesium to a lesser degree than illite or vermiculite. This observation corresponds with the high mobility of part of the 137Cs in our forest soils.
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Author Contributions
F.C. and P.H. wrote the first version of the manuscript. F.C., P.H., C.S. and Y.T. designed the sampling strategy and participated in the sampling campaign. N.L., A.J. and M.A.G. participated in the sampling campaign. Y.O. and F.C. and P.H. wrote the first version of the manuscript. F.C. and P.H. designed the sampling strategy and participated in the sampling campaign. All authors reviewed the manuscript and improved it.

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