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Atmospheric Direct Uptake and Long-term Fate of Radiocesium in Trees after the Fukushima Nuclear Accident

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Large areas of forests were radioactively contaminated by the Fukushima nuclear accident of 2011, and forest decontamination is now an important problem in Japan. However, whether trees absorb radioactive fallout from soil via the roots or directly from the atmosphere through the bark and leaves is unclear. We measured the uptake of radiocesium by trees in forests heavily contaminated by the Fukushima nuclear accident. The radiocesium concentrations in sapwood of two tree species, the deciduous broadleaved konara (*Quercus serrata*) and the evergreen coniferous sugi (*Cryptomeria japonica*), were higher than that in heartwood. The concentration profiles showed anomalous directionality in konara and non-directionality in sugi, indicating that most radiocesium in the tree rings was directly absorbed from the atmosphere via bark and leaves rather than via roots. Numerical modelling shows that the maximum ¹³⁷Cs concentration in the xylem of konara will be achieved 28 years after the accident. Conversely, the values for sugi will monotonously decrease because of the small transfer factor in this species. Overall, xylem ¹³⁷Cs concentrations will not be affected by root uptake if active root systems occur 10 cm below the soil.

The Fukushima nuclear disaster, caused by the great earthquake and tsunami of 11 March 2011, contaminated vast forest areas with radiocesium: ¹³⁷Cs at 1.5×10^{16} Bq with ¹³⁴Cs/¹³⁷Cs approximately equal to 1.0¹. Forest decontamination is an urgent issue. Although the internal contamination of trees has been roughly estimated to be small using simple compartment models and a set of parameters² that takes into account the properties of Japanese forest soil, accurate understanding of how trees absorb fallout ¹³⁷Cs is required for quick and effective decontamination. To obtain a better understanding of the uptake mechanisms of fallout radionuclides, heavy metals and volatile organic matter in the xylem of trees, we reviewed past studies on radionuclides from the Chernobyl nuclear accident^{3–6}, nuclear weapons tests^{7–9} and the Nagasaki A-bomb^{10–12}, in addition to studies on heavy metals¹³ and organic material¹⁴. A few studies^{10–14} have suggested the possibility of direct uptake (i.e. not via soil) of atmospheric radionuclides, heavy metals and organic material into the xylem of trees from bark and leaves; however, most studies^{3–9} on the Chernobyl accident and nuclear weapons testing have argued that fallout radionuclides are absorbed through the roots from the soil (i.e. root uptake). If atmospheric direct uptake is the main route for absorption of ¹³⁷Cs into the xylem of trees in Japan, the radiocesium concentration within a tree will decrease with time after the Fukushima nuclear accident. Conversely, if the roots are the main route for radiocesium uptake, ¹³⁷Cs concentration might gradually increase with a large time lag after the accident, because ¹³⁷Cs reaches the active zone of roots in the forest soil after a long delay. Herein, we discuss estimates of the possibility and magnitude of direct uptake of atmospheric ¹³⁷Cs into trees, and long-term prediction of changes in ¹³⁷Cs concentration in tree xylem by root uptake after atmospheric direct uptake.

Results

We measured the vertical profiles of stable and radioactive Cs and K in soil of two tree-harvesting yards at Koriyama, Fukushima, in 2013 (Supplementary Fig. S1). Both ¹³⁴Cs and ¹³⁷Cs in the soil are strongly fixed by clay minerals in soil, as shown in Figs. 1a and 1b. More than 99% of the ¹³⁴Cs and ¹³⁷Cs deposited from the nuclear accident were trapped in the litter layer and the top 2.5 cm of soil 2.5 years after the accident; their concentrations decrease rapidly with increasing depth. In contrast, ⁴⁰K and the stable isotopes of Cs and K are almost evenly distributed through the soil profile, except stable K at two depths in the sugi yard.

We harvested two Japanese common trees (konara: *Quercus serrata* Murray, a deciduous broadleaf tree and sugi: *Cryptomeria japonica* D. Don, an evergreen coniferous tree) to investigate the distribution of ¹³⁷Cs, ¹³⁴Cs and

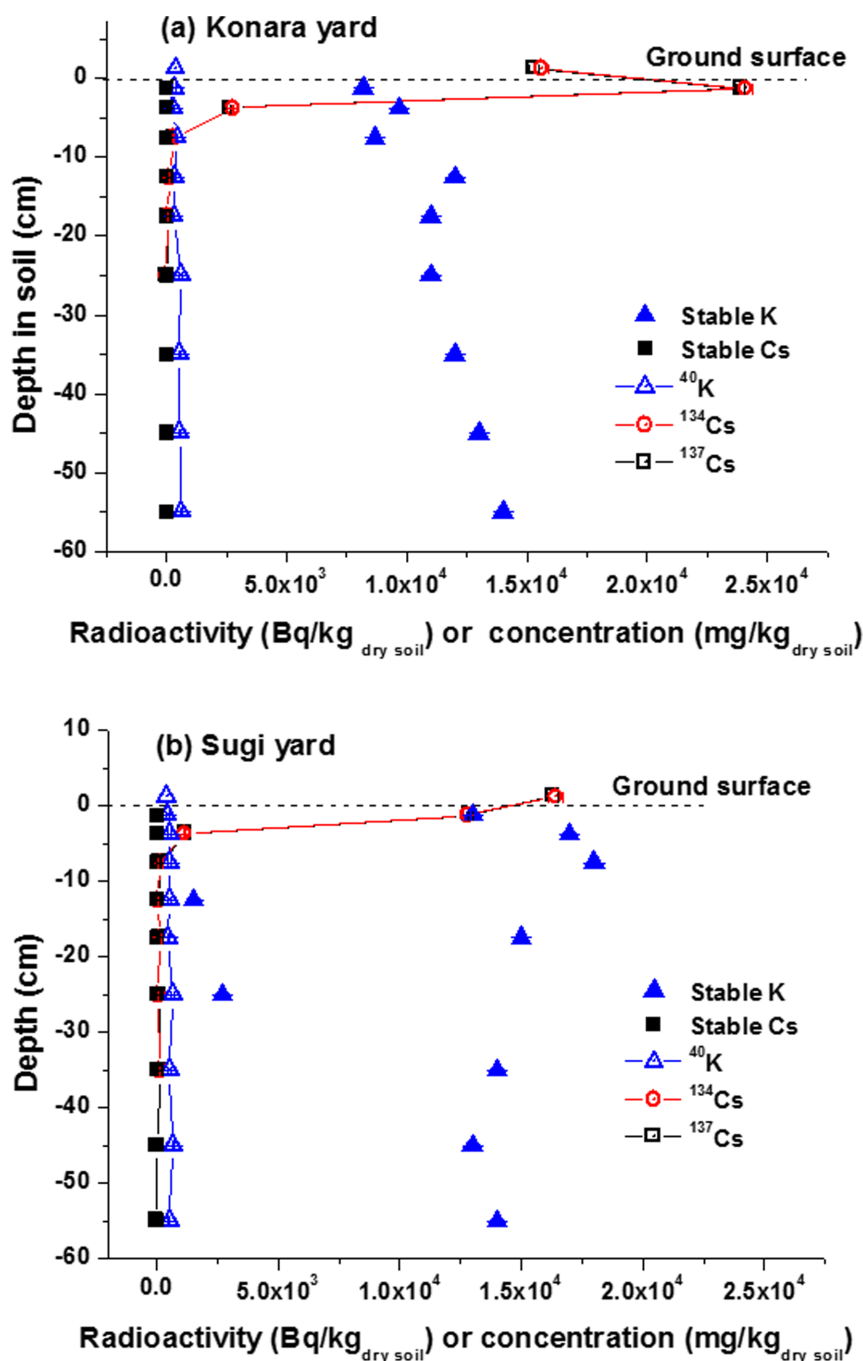


Figure 1 | Vertical concentration profiles of ¹³⁷Cs, ¹³⁴Cs, ⁴⁰K, stable isotopes of Cs and K in soil, 2.5 years after the Fukushima nuclear accident. (a) the konara yard and (b) the sugi yard. Open blue triangles, red circles, black squares and solid squares and triangles indicate the concentration of ⁴⁰K, ¹³⁴Cs, ¹³⁷Cs, and stable isotopes of Cs and K in soil, respectively.

⁴⁰K in tree rings in 2012 (Supplementary Table S1). The distributions of ¹³⁷Cs and ¹³⁴Cs are almost the same in the rings of both trees. We also measured the concentrations of stable isotopes of Cs and K in tree rings (Supplementary Table S2). We show the distribution of ¹³⁷Cs and ⁴⁰K in the xylem in trees divided into four cardinal compass directions in Figs. 2a and 2b. We also demonstrate a directional distribution of ¹³⁷Cs concentration compared with ⁴⁰K concentration for konara (Fig. 3a) and a non-directional distribution for sugi (Fig. 3b). In addition, we summarize the directional distribution of radiocesium concentration in the sapwood region of trunk discs in four different directions, verified using the χ^2 -test, in Table 1.

Using the model proposed in this study, we predict changes in the concentration of ¹³⁷Cs in tree rings of konara and sugi up to 120 years

after the nuclear accident. We show changes in the ¹³⁷Cs concentration in the xylem with time from the accident (Figs. 4a and 4b), varying the magnitude of the retardation factor¹⁵ controlling the migration of ¹³⁷Cs in forest soil.

Discussion

To confirm the occurrence of atmospheric direct uptake into trees, we hypothesized that radionuclides could be absorbed via the following two pathways. First, radionuclides absorbed through the bark could be directly transported into the sapwood, possibly along the ray tissue^{13–14}; if this hypothesis is correct, the radionuclide concentration in tree rings would depend on differences in the directional

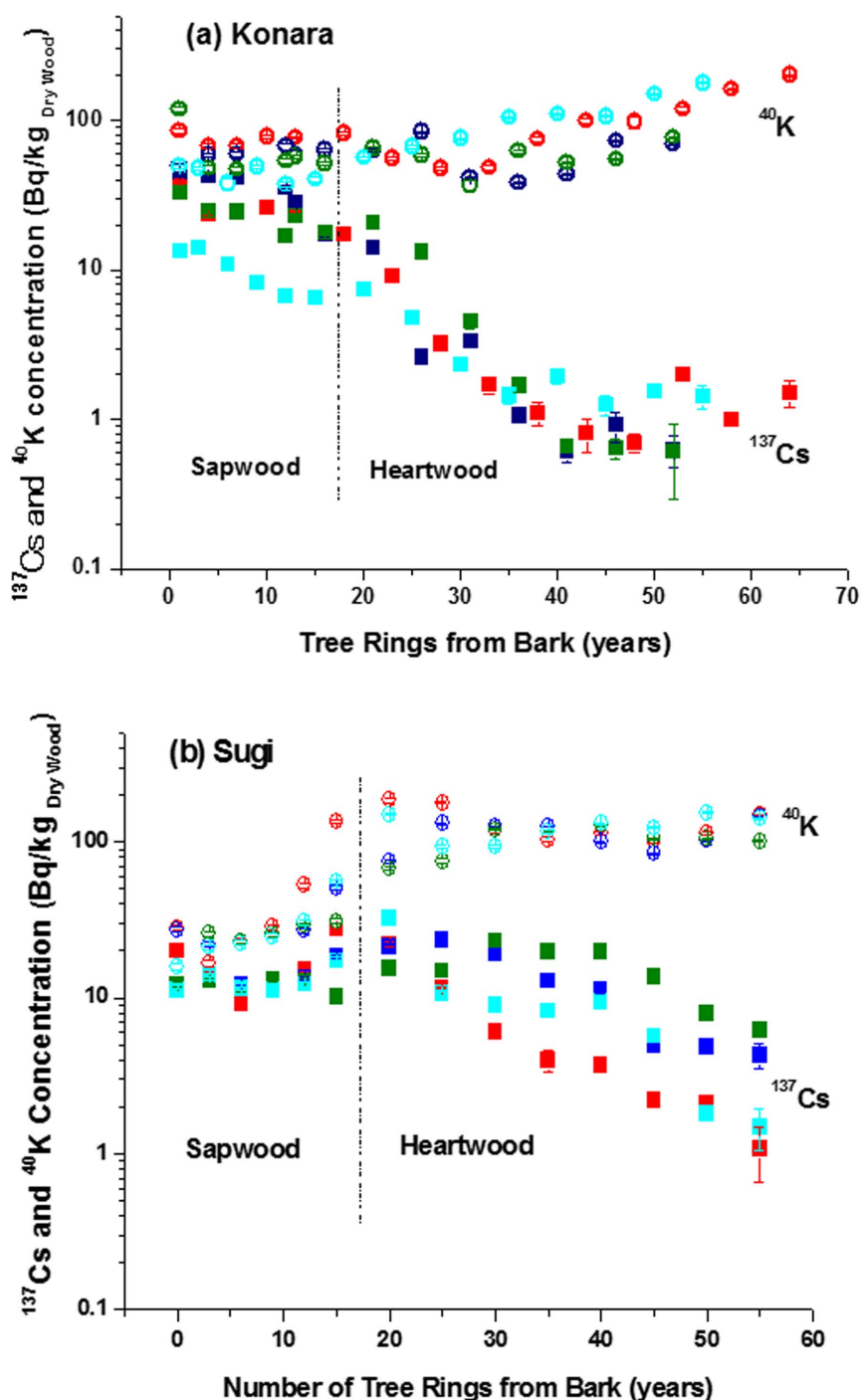


Figure 2 | Radial distribution of ^{137}Cs and ^{40}K measured in annual tree rings in the trunk disc for each of the four different cardinal compass directions. Results for (a) konara and (b) sugi. Solid squares represent the ^{137}Cs concentration and open circles the ^{40}K concentration. Red, navy blue, olive and turquoise blue indicate the sides of the trunks facing east, north, west and south, respectively.

abundance of radionuclides deposited on the bark surface. In contrast, direct uptake through the leaves of an evergreen coniferous tree would probably show no directionality, because radionuclides transported from multi-directional leaves through phloem sieve tubes would be homogenized by mixing in the sapwood^{15,13–14}. In general, the large canopy of leaves of evergreen coniferous trees effectively traps fallout radionuclides in rain before they reach the bark, and the total surface area of leaves is larger than that of bark. If the bark facing a certain compass direction is heavily contaminated with radionuclides before the spring leaf flush, it will be possible to detect significant directionality in the radionuclide concentration in the tree rings of a deciduous tree.

In March 2011, konara in the Fukushima area did not have leaves but sugi exhibited full foliage. Large radio-plumes were released from the accident in the middle of March; later, much radiocesium was transported to the harvesting sites along the mountain range by north-easterly wind and rain during 21–23 March¹⁶ (Supplementary Fig. S1). To test our hypothesis, we measured the radial variation of radionuclide and stable isotope concentrations in tree rings in each of the four different cardinal compass directions. The average concentrations of radiocesium in the sapwood of konara and sugi were markedly higher than those in the heartwood region. The radiocesium concentration in the heartwood of konara decreased dramatically from the

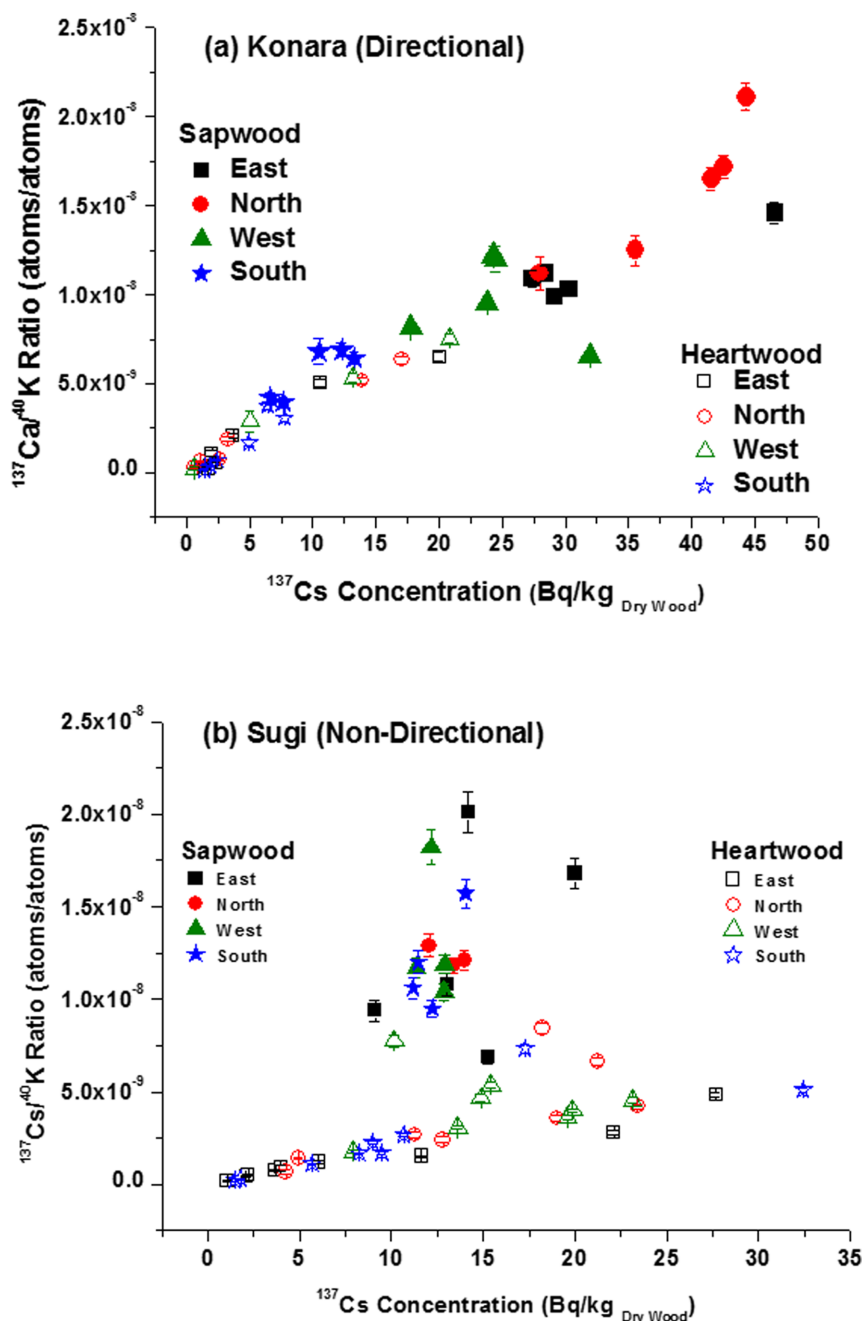


Figure 3 | Correlation between the ^{137}Cs concentration and the $^{137}\text{Cs}/^{40}\text{K}$ ratio in the annual tree rings of (a) konara and (b) sugi. The trunks were divided into four parts, facing north, south, east and west.

boundary between the sapwood and the heartwood towards the pith. The concentration of ^{137}Cs in the heartwood appears to lie approximately along a straight line with a steep slope on a semi-logarithmic scale, showing the effect of diffusion (Fig. 2a). However, as shown in Fig. 2b, the profile for ^{137}Cs concentration in sugi was not the same as the pattern in konara. We interpret this as indicating that radiocesium in the tree rings of sugi diffused faster than in konara, possibly because sugi contains more water and has a lower woody density in the xylem than in konara¹⁷.

If K, an indispensable element for tree growth, is absorbed primarily through the roots, ^{40}K should also be absorbed by roots together with stable isotopes of K and Cs from the soil. We measured the concentrations of stable isotopes of K and Cs in ash samples from the sapwood and the heartwood of both species (Supplementary Table S2). The measured concentration of ^{40}K is generally low in

the sapwood and high in the heartwood. The ^{40}K concentration of both species was uniform and constant in each of the sapwood and heartwood regions in the four different compass directions (Figs. 2a and 2b; Table 1).

For the konara samples, we estimated four ratios ($^{137}\text{Cs}_{\text{north}}/^{137}\text{Cs}_{\text{south}}$, $^{134}\text{Cs}_{\text{north}}/^{134}\text{Cs}_{\text{south}}$, $^{40}\text{K}_{\text{north}}/^{40}\text{K}_{\text{south}}$ and stable $\text{Cs}_{\text{north}}/\text{Cs}_{\text{south}}$) for the same annual tree rings in sapwood regions facing north and south. Similarly, we also estimated the ratios of $^{137}\text{Cs}_{\text{east}}/^{137}\text{Cs}_{\text{west}}$, $^{134}\text{Cs}_{\text{east}}/^{134}\text{Cs}_{\text{west}}$, $^{40}\text{K}_{\text{east}}/^{40}\text{K}_{\text{west}}$ and stable $\text{Cs}_{\text{east}}/\text{Cs}_{\text{west}}$ in sapwood facing east and west. These estimates showed that the radiocesium concentrations in the north-facing tree rings are anomalously higher than those facing south. A clear directionality of radiocesium concentration in the sapwood was observed along the north–south section. The contrast between the heterogeneous concentration profiles of radiocesium and the homogeneous



Table 1 | Ratios (north/south and east/west) of the concentrations of ^{137}Cs , ^{134}Cs , ^{40}K and stable Cs in equivalent annual tree rings in the sapwood of konara and sugi, and the χ^2 -test. (Results of the χ^2 -test with a significance level of 0.01 and minimum four degrees of freedom compared with the completely homogeneous concentration with a ratio of 1.0 in the same band of the annual tree rings.)

	Tree Rings (years)	^{137}Cs		^{134}Cs		^{40}K		Stable Cs		
		Ratio	error	Ratio	error	Ratio	error	Ratio	error	
Konara	North/South	1995–1997	4.22	0.22	6.09	0.88	1.58	0.12	1.66	1.4E-04
		1998–2000	4.64	0.22	4.47	0.37	1.47	0.09	1.59	1.1E-04
		2001–2003	3.96	0.15	3.98	0.25	1.63	0.16	1.32	8.7E-05
		2004–2006	3.46	0.11	5.27	0.39	1.39	0.06	1.10	7.1E-05
		2007–2009	3.34	0.10	3.39	0.24	1.02	0.04	1.40	8.6E-05
		$P(\chi^2)$	43.9 > 13.28		70.8 > 13.28		1.11 < 13.28		0.58 < 13.28	
	East/West	1995–1999	1.27	0.03	1.35	0.05	1.23	0.05	0.81	4.5E-05
		2000–2002	1.79	0.04	1.59	0.05	1.29	0.05	1.16	6.3E-05
		2003–2005	1.17	0.03	1.23	0.04	1.27	0.06	2.17	1.8E-04
		2006–2008	1.21	0.04	1.11	0.05	1.24	0.07	0.68	2.8E-05
	2009–2011	1.41	0.04	1.62	0.08	0.63	0.03	0.99	6.2E-05	
	$P(\chi^2)$	0.94 < 13.28		0.92 < 13.28		0.35 < 13.28		1.53 < 13.28		
Sugi	North/South	1998–2000	1.06	0.03	1.03	0.05	0.92	0.04	0.97	4.6E-04
		2001–2003	1.10	0.04	0.95	0.06	0.88	0.04	1.38	1.3E-03
		2004–2006	no measurement						1.06	8.6E-04
		2007–2009	1.05	0.05	1.09	0.09	0.97	0.05	1.12	8.8E-04
		2010–2012	1.00	0.04	1.01	0.07	1.30	0.07	2.71	4.1E-03
		$P(\chi^2)$	0.016 < 11.34		0.012 < 11.34		0.112 < 11.34		3.09 < 13.28	
	East/West	1998–2000	1.19	0.04	1.35	0.08	1.80	0.09	1.10	7.4E-05
		2001–2003	1.01	0.04	1.08	0.07	1.11	0.06	1.20	7.8E-05
		2004–2006	0.80	0.04	0.88	0.07	1.00	0.05	1.50	1.1E-04
		2007–2009	1.10	0.04	1.19	0.07	0.65	0.04	1.28	9.0E-05
	2010–2012	1.64	0.06	1.77	0.10	1.78	0.11	1.45	8.8E-05	
	$P(\chi^2)$	0.496 < 13.28		0.772 < 13.28		1.383 < 13.28		0.58 < 13.28		

concentration profiles of ^{40}K and stable Cs in the sapwood region strongly indicate that the uptake route of fallout radiocesium differs from those of ^{40}K and stable Cs, which is absorbed from the soil via roots (Fig. 3a; Table 1).

Conversely, the ratios of $^{137}\text{Cs}_{\text{north}}/^{137}\text{Cs}_{\text{south}}$, $^{134}\text{Cs}_{\text{north}}/^{134}\text{Cs}_{\text{south}}$, $^{137}\text{Cs}_{\text{east}}/^{137}\text{Cs}_{\text{west}}$, $^{134}\text{Cs}_{\text{east}}/^{134}\text{Cs}_{\text{west}}$, $^{40}\text{K}_{\text{north}}/^{40}\text{K}_{\text{south}}$, $^{40}\text{K}_{\text{east}}/^{40}\text{K}_{\text{west}}$, stable $\text{Cs}_{\text{north}}/\text{Cs}_{\text{south}}$ and $\text{Cs}_{\text{east}}/\text{Cs}_{\text{west}}$ in the same annual tree rings in the sapwood of sugi were almost constant (Table 1). There are no significant differences among these ratios in the sapwood. We could not observe a clear directionality in the radiocesium concentration even if it was present in the sapwood (Fig. 3b), because as previously mentioned, sugi probably had two different routes for direct atmospheric uptake: through the bark and through the leaves. In the case of sugi, the directionality in the radiocesium concentration absorbed through the bark thus appears to have been diluted and concealed by the effect from the multi-directional leaves in the sapwood. These results support our hypotheses: much fallout radiocesium was directly absorbed into tree xylem through short-term atmospheric uptake in 2011.

We observed anomalous directionality in profiles of mobile radiocesium concentration in tree rings of the deciduous tree konara using precise measurements of radioactivity of radiocesium in the xylem. However, this clear directional trace in konara will soon disappear as a result of rapid transport and diffusion throughout the tree. Directional distribution of radiocesium detected using the imaging plate (IP) method, i.e. autoradiography, in 2011 was undetectable in May 2012¹⁸ due to it being less than the detection limit of the method.

We predicted the ^{137}Cs concentration in the xylem of both types of tree at Koriyama, Fukushima, using the proposed model (Supplementary Information), the average transfer factors of stable Cs estimated for the sapwood region ($9.59 \pm 2.56 \times 10^{-3}$ for konara

and $8.34 \pm 2.55 \times 10^{-4}$ for sugi; Supplementary Table S2), and direct uptake from the atmosphere (11 Bq/kg for konara and 13 Bq/kg for sugi) in 2011 as the initial ^{137}Cs concentration (Supplementary Tables S3 and S4). Our predictions of root uptake at a depth of 0 cm are that the maximum ^{137}Cs concentration in konara tree rings will reach 120 Bq/kg 28 years after the nuclear accident (Fig. 4a). Conversely, the ^{137}Cs concentration of sugi will decrease monotonously due to the small transfer factor of Cs from the accident. Thus, the atmospheric direct uptake value of 13 Bq/kg plays a vital role in the ^{137}Cs concentration in sugi (Fig. 4b).

Consequently, the absorption effect of ^{137}Cs via root uptake is greater for konara than for sugi. The ^{137}Cs concentration in the xylem of a tree strongly depends on the magnitude of the transfer factor of Cs, deposition rate of radiocesium on the ground surface, and depth of the active root system in soil, although the initial effect of atmospheric direct uptake has controlled the ^{137}Cs concentration in the coniferous tree sugi, which has been planted throughout Japan. Our predictions indicate that the ^{137}Cs concentration in the xylem of trees will not be affected by root uptake, if konara does not have an active root system 10 cm below the ground surface. However, further studies of the depths of active root systems in soil are required to estimate the effects of root uptake more precisely.

Methods

Material. Konara was harvested prior to leaf fall at the end of September 2012, and sugi was harvested at the end of October 2012. We used a harvest time of 1.5 years after the nuclear accident to obtain clear data on ^{137}Cs diffusion into xylem. This timing is based on our previous study¹⁰ on ^{137}Cs diffusion in French white fir observed at Nancy, France, three years after the Chernobyl accident. We precisely marked each of the four compass directions on the trunks prior to cutting down the trees. After cutting the trees at a height of 1.0 m from the ground surface, we carefully and completely stripped off all bark from the tree trunks over a distance of 45 cm to prevent contamination of the xylem. We cut the trunk vertically into four quarters

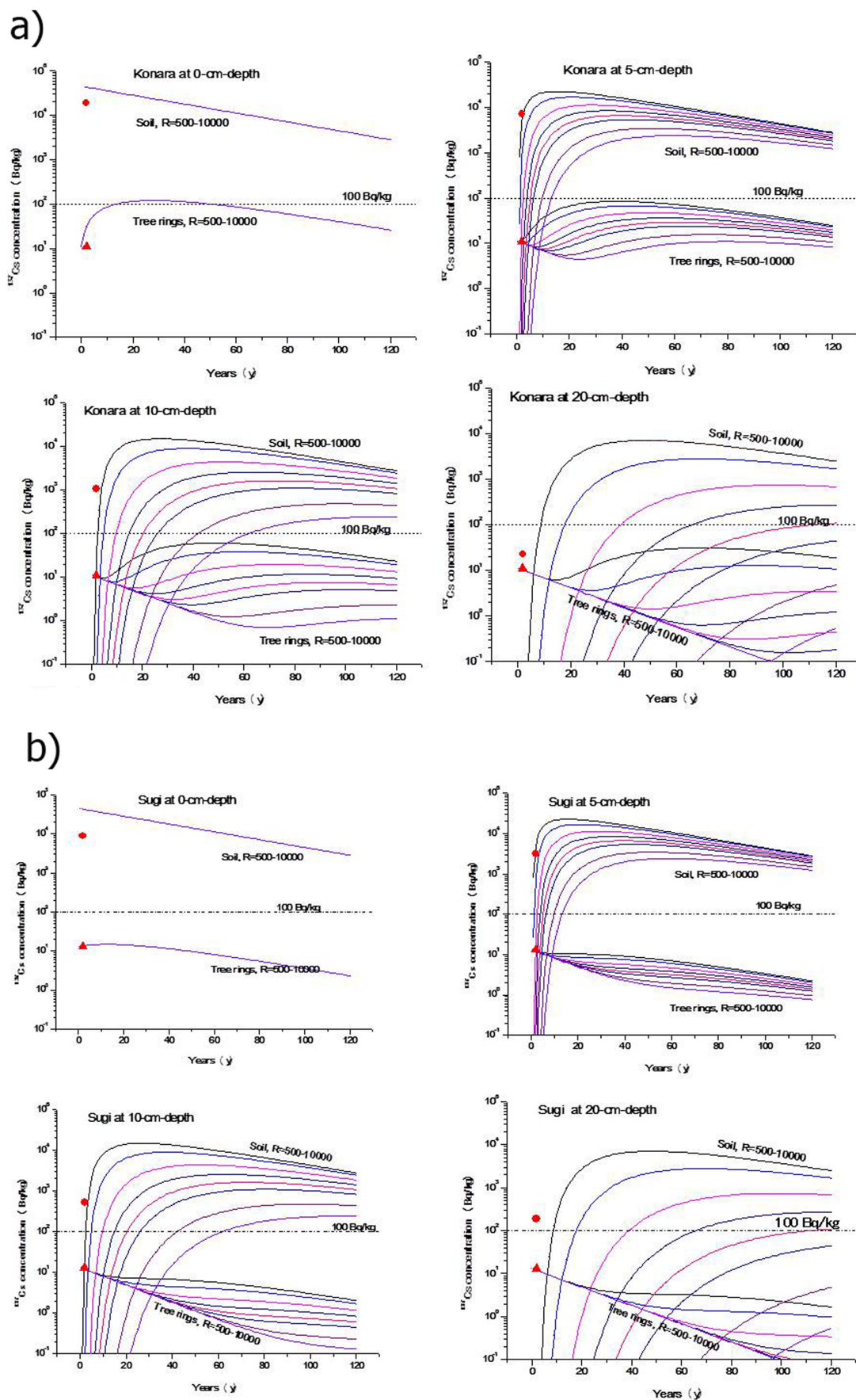


Figure 4 | Correlations between the ^{137}Cs concentration in soil and tree rings, and the elapsed time to 120 years after the Fukushima nuclear accident. (a) konara and (b) sugi. ^{137}Cs concentration in tree rings estimated when all ^{137}Cs was absorbed from depths of 0 to 20 cm by root uptake alone. Red solid circles and triangles indicate the ^{137}Cs concentration in soil and tree rings, respectively. Coloured lines (black, blue, red, dark blue, pink, navy blue, purple and violet) indicate the magnitude of retardation factors (500, 1000, 2000, 3000, 4000, 5000, 7500 and 10000). The unit of radioactivity is Bq/kg of dry wood or soil.



and then sliced each quarter horizontally into 15 quarter discs, each with a thickness of 2–3 cm. We then separated the tree rings in each quarter disc using a clean chisel for each ring separation to prevent contamination. The separated tree rings were ashed for 20 h at 450°C in an electric oven.

Measurement. The ash samples were then measured for radiocesium and ^{40}K by γ -ray spectrometry using a low-background p-type high-purity Ge-detector (GMX40P4-76, ORTEC, Tennessee, U.S.A.) with 40% relative efficiency, and a multichannel analyzer (MCA 7600-000, Seiko EG&G, Tokyo, Japan) with a pulse height analyzer. The instruments were calibrated using the ^{40}K (KCl) method¹⁹, and all samples were measured within a period of five days. The detection limits are 0.032 Bq/kg to 661 keV for ^{137}Cs , 0.035 Bq/kg to 604 keV for ^{134}Cs , and 0.045 Bq/kg to 1460 keV for ^{40}K . The concentrations of the stable isotopes of K and Cs were measured using an ICP-MS (Agilent 7500cx, Agilent Technology, California, U.S.A.) after the ashed samples were completely dissolved with nitric acid (1:10) using the preparation method in reference 20.

Model. We proposed a model to estimate the ^{137}Cs concentration in the xylem of trees using a root-uptake model that considers the vertical ^{137}Cs concentration in soil and the stable Cs transfer factor from soil to tree xylem with atmospheric direct uptake as an initial condition. We estimate the average ^{137}Cs concentration ($C_{\text{tree}}(n)$) in the specified entire disc of tree trunk at a height of 1 m above the ground surface using the following assumptions:

The radius of a trunk of tree can be approximated by a cylinder with radius r_n (time) = n). The elapsed time from the nuclear accident is n .

The radius of the trunk is expressed by the term $r_n = \alpha \times r_{n-1}$ after n years from the accident. The growth rate of trunk α is expressed by the following term $\alpha = r_n/r_{n-1}$ ($\alpha \geq 1$). The magnitude of α is conservatively 1.013 (Supplementary information and Supplementary Fig. S2). $\Delta r = r_n - r_{n-1}$ is the width of a tree ring formed in year n .

The initial ^{137}Cs concentration in the xylem of a tree in 2011 is given only by the atmospheric direct uptake; additional ^{137}Cs is provided every year from soil only by root uptake. The ^{137}Cs concentration (C_a) added in the newly formed tree ring is calculated by using the ^{137}Cs concentration ($C_{\text{soil}(n,x)}$) in soil at a depth of x and the magnitude of the ^{137}Cs transfer factor ($T_{f(i)}$) from soil to tree as follows: $C_a = C_{\text{soil}(n,x)} \times T_{f(i)}$. We used the average transfer factors estimated from the ratio ($C_{\text{stree}}/C_{\text{soil}}$) of stable Cs concentration in tree rings (C_{stree}) of the sapwood region to stable Cs concentration (C_{soil}) in soil (Supplementary Table S2). The active root system that absorbs ^{137}Cs from soil is only at a specific depth x in the soil. The ^{137}Cs concentration at depth x in soil can be estimated using the one-dimensional diffusion–advection model^{21–23}.

The new addition of ^{137}Cs through root uptake is homogeneous every year in the entire xylem of tree as a result of complete mixing with the ^{137}Cs concentration in the tree from the previous year ($n-1$). The ^{137}Cs concentration in the specified disc of the trunk decreases only by the radioactive decay with the decay constant λ : 0.023 y^{-1} .

The ^{137}Cs concentration ($C_{\text{tree}}(n)$) is expressed by the following equation:

$$C_{\text{tree}}(n) = (C_{\text{tree}}(n-1) \cdot \text{Exp}(-\lambda) + (\alpha^2 - 1) \cdot C_{\text{soil}(n,x)} \cdot T_{f(i)}) / \alpha^2 \quad (1)$$

As the initial ^{137}Cs concentration ($C_{\text{tree}}(0)$) is given by the atmospheric direct uptake, the ^{137}Cs concentration ($C_{\text{tree}}(1)$) at the first year ($n = 1$) can be expressed as follows:

$$C_{\text{tree}}(1) = (C_{\text{tree}}(0) \cdot \text{Exp}(-\lambda) + (\alpha^2 - 1) \cdot C_{\text{soil}(1,x)} \cdot T_{f(i)}) / \alpha^2 \quad (2)$$

We verified that the proposed model could reasonably predict the ^{137}Cs concentration ($C_{\text{tree}}(n)$) in the xylem of a tree using data on ^{137}Cs concentration of sugi harvested at Nagasaki in 1988¹⁰ and that of soil^{21,22} (Supplementary information, Table S3 and S4). Sugi harvested at Nagasaki showed the effects of both atmospheric deposition from the local fallout after the Nagasaki A-Bomb and global fallout from nuclear weapons testing in the 1950s and 1960s.

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Author contributions

Y.M. designed this study and performed the field studies, analysed the data and wrote the paper; T.O. performed the experiments, measured the radioactivity, analysed the data and discussed the results; H.O. and A.K. managed and performed the field studies and pre-treated the tree and soil samples.

Additional information

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