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Radionuclides from the Fukushima Daiichi Nuclear Power Plant in terrestrial systems

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Abstract | The 2011 Fukushima Daiichi Nuclear Power Plant (FDNPP) accident, Japan, released the largest quantity of radionuclides into the terrestrial environment since the 1986 accident at Chernobyl. This accident resulted in 2.7 PBq of radiocaesium (^{137}Cs) contaminated forests, agricultural lands, grasslands and urban areas, which subsequently migrated through soil and waterways in the Fukushima Prefecture. In this Review, we synthesize knowledge regarding the deposition, distribution and transport of fallout radionuclides, especially ^{137}Cs, in the terrestrial environment after the FDNPP accident, which were revealed by extensive and continuous environmental monitoring. Anthropogenic activities, high run-off and steep topography led to a rapid decline in the activity concentration of ^{137}Cs in soils and rivers, especially in the first year after the accident. The decline in exposed radioactivity was notably faster than that seen after the Chernobyl Nuclear Power Plant accident, likely related to differences in geography and climate, and the intensive remediation activities in Fukushima. However, forests in Fukushima have retained a notable amount of ^{137}Cs in the upper centimetres of soil and could persist as a source of ^{137}Cs into rivers. For continued understanding of both natural and fallout radionuclide behaviour in the environment, there must be long-term accessibility of the data collected in response to the FDNPP accident.

Fallout
Atmospherically deposited radionuclides or atmospheric deposition of radionuclides onto the Earth surface.

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Nuclear power generated 2,563 TWh of electricity worldwide[1] in 2018, with low greenhouse-gas emissions and little air pollution as by-products. However, environmental pollution by nuclear radioactive materials, especially during uncontrolled-release events, can have devastating consequences. The March 2011 large-scale nuclear accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) in Japan released 520 PBq of radionuclides excluding noble gases[2–4], such as radiocaesium (^{137}Cs, ^{134}Cs), radioiodine (^{131}I), radiostrontium[5] and uranium[6], among others, to the atmosphere. This release spread radioactive fallout onto the ocean and throughout the Fukushima region, including 10 PBq of ^{137}Cs and 120 PBq of ^{131}I (REF.[7]) (FIG. 1a). Of the radionuclides, ^{137}Cs had the most substantial long-term impact on terrestrial contamination and human health released by the FDNPP accident, owing to its high release rate and relatively long half-life (30.1 years)[8,9]. Therefore, it is particularly important to understand the fate and dynamics of the fallout ^{137}Cs (REFS[10–12]).

The atmospheric release[10,13] of ^{137}Cs and direct release of radioactively contaminated water from the power plant into the ocean[14] are both well understood. Similarly, the oceanic transport and redistribution of ^{137}Cs after the FDNPP accident were intensively monitored and modelled[15,16]. In seafloor sediments surrounding Fukushima, the activity concentrations of ^{137}Cs declined rapidly due to sediment transport and dilution in the ocean[17]. Indeed, 16–19 PBq of ^{137}Cs was released to the marine environment[16] through atmospheric deposition and direct discharge, and ^{137}Cs activity concentrations decreased due to the dilution effect from the mixing of the contaminated seawater with low ^{137}Cs oceanic water during the 8 years following the accident[18].

The impact of FDNPP radionuclide fallout in the terrestrial environment is less well known than in the marine environment. Indeed, the terrestrial post-accident 'self-cleaning' (REF.[19]) and rapid reduction of exposed radioactivity has surprised researchers, as it was assumed that, based on observations made after the Chernobyl Nuclear Power Plant (CNPP) accident in Ukraine, the reduction of the exposed radioactivity in the environment would take much longer. The 1986 CNPP accident is the only other accident rated at the

Key points

- Intensive monitoring after the 2011 Fukushima Daiichi Nuclear Power Plant accident in Japan revealed radionuclide, including radiocaesium (^{137}Cs), contamination in the terrestrial environment, in forests, soils, cropland, paddy fields, urban areas and rivers.
- After deposition, fallout radionuclides migrated through soil and water into rivers. Some radionuclides were eventually washed into the ocean, though most of the ^{137}Cs in forests remains in the soil.
- The ^{137}Cs activity concentration declined quickly during the first year after the accident, though the rates of decline varied between land uses. Subsequently, ^{137}Cs activity concentrations continued to decline, but the rates became more similar between land uses.
- Due to the combination of active land use and decontamination efforts, the fallout ^{137}Cs activity concentration in terrestrial environments declined quickly relative to expectations.
- Continued accessibility of data collected post-accident is crucial, as nuclear power is a prevalent energy source.

Throughfall
Rainfall passing through tree crowns.

Stemflow
Rainwater flowing down along tree trunks.

Litterfall
Plant material such as foliage, twigs and branches falling to the ground.

Radioactive decay
When an unstable nucleus changes into a stable nucleus by emitting radiation.

maximum level on the International Nuclear Event Scale. While dissolved ^{137}Cs activity concentrations decreased rapidly in European rivers and lakes within several months after the CNPP accident[19], a second, much slower rate of decline followed[20,21]. This long-term slow decline of ^{137}Cs in catchment soils and water[22] caused ^{137}Cs to persist in freshwater fish in Norway[23] and the United Kingdom[24] for several decades after the accident. However, differences between the radionuclide composition, post-accident environmental recovery efforts and local environmental factors led the terrestrial areas impacted by the FDNPP to recover more quickly than expected[25].

In this Review, we discuss the fate of the radionuclide fallout — notably of ^{137}Cs — on Japan after the FDNPP accident. We examine the downwards migration of ^{137}Cs through forests and soil, remobilization from hillslopes and transport through rivers, and the factors that influenced the radionuclide movement. The environmental recovery of the impacted region post-accident is compared with that after the CNPP accident. Finally, we highlight some of the scientific lessons learned from continual monitoring and emphasize the importance of maintaining these lessons and data for future researchers.

Environmental distribution of ^{137}Cs

Following the FDNPP accident, 2.7 PBq of ^{137}Cs was deposited onto the terrestrial environment, 67% of which fell out onto forests, 10% onto paddy fields, 7.4% onto cultivated areas and grasslands, and 5.0% onto urban areas[12] (FIGS 1b,2). Although wet deposition (associated with rainfall and snowfall) accounted for 98% of the total deposition[26,27], dry deposition (interception of gases phases under gravity, diffusion processes or by turbulent transfer) dominated along the coastline around the FDNPP[28,29]. Approximately 80% of the total ^{137}Cs deposition onto land surface occurred within the combined catchment areas of the Abukuma River (the largest river in the region) and small coastal rivers located in the 80-km radius area from the FDNPP (for a total area of 9,862 km^2)[25]. Of this land, 3,900 km^2 was contaminated by at least 100 kBq m^{-2} of ^{137}Cs fallout and 254 km^2 was subjected to severe ^{137}Cs fallout of at least 1,480 kBq m^{-2} (REF.[12]) (FIG. 1).

Forested areas. Forested areas experienced most of the FDNPP ^{137}Cs fallout in the terrestrial environment — 2,600 km^2 of forest received ^{137}Cs fallout greater than 100 kBq m^{-2} (REFS[12,30]). Some of this fallout was deposited directly onto the forest floor, but in evergreen forests (such as those dominated by Japanese cedar), 60–90% of the deposited ^{137}Cs was intercepted by tree canopies[31–35] (FIG. 3). Washing experiments of the contaminated foliage and branch wood showed that 50–60% of the intercepted ^{137}Cs was firmly fixed on the tree biomass[36]. However, only 10–25% of the ^{137}Cs was intercepted by the canopy in deciduous broadleaf forests (such as those dominated by Japanese oak), as the fallout period occurred prior to canopy regrowth[33,34] (FIG. 3). These variations in radionuclide interception resulted in a wide range of contamination levels in forest components and accumulated radiation doses to human and forest wildlife[37–39].

The distribution of ^{137}Cs within forests changed significantly within the first year after the fallout (referred to here as the first phase)[40,41] (FIG. 3). During this phase, ^{137}Cs was transported from the canopy to the forest floor through hydrological[31,33,35,40–48] and biological processes[33–36,48,49]. Hydrological pathways such as throughfall and stemflow were initially the most important in transporting ^{137}Cs and were responsible for 70% of the total flux from the canopy to the forest floor in the first year, mostly (>80%) in the dissolved form[43]. Whereas ^{137}Cs from the contaminated forest leached efficiently to throughfall during the early phase of contamination, the leaching rate of ^{137}Cs from the canopy to throughfall decreased over time[35], owing mainly to the internalization of surface contamination by the trees[50]. In these early stages of deposition, ^{137}Cs accumulated mainly in the organic matter at the surface of the forest floor, before migrating deeper into the subsoil.

Around one year after the fallout (referred to here as the second phase), litterfall played an increasing role in the migration of ^{137}Cs from the canopy to the forest floor[33,42,48,49]. Owing to the combination of throughfall, stemflow, litterfall and radioactive decay, the contamination of trees as a whole decreased during the 8 years of observation after the accident[35,51,52]. ^{137}Cs activity concentrations in foliage, which was initially the most highly contaminated part of the trees, decreased faster than the contamination of perennial parts of the trees, such as wood and bark[53,54] (FIG. 3). Indeed, in some trees, such as the Japanese oak, wood contamination showed an increasing trend after the accident[53–55]. For Japanese cedar (a coniferous tree), the initial increasing trend in

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Fig. 1 | **^{137}Cs deposition and land-use type in the 80-km radius from the FDNPP.** The Fukushima Daiichi Nuclear Power Plant (FDNPP) is located on the east coast of Fukushima Prefecture, Japan (grey area in the inset map of Japan). **a** | ^{137}Cs deposition density within 80 km of the FDNPP, based on airborne monitoring calibrated by the soil sampling of 2,200 points[11,12]. The area with a deposition above 1,480 kBq m^{-2} is bordered by a thin white line and is mostly to the north-west of the FDNPP. **b** | Land use within 80 km of the FDNPP, including a small urban area (in grey) around the FDNPP, which is surrounded mainly by dense forest (dark green) and paddy fields (light blue). The area surrounded by a red dotted line is designated as a special decontamination area[203]. In this area, decontamination activities were carried out by the Japanese Ministry of the Environment[127]. In both panels, the Abukuma River basin is enclosed by a black dotted line.

^{137}Cs activity concentrations in the wood was followed by stabilization or a slight decrease 4–5 years after the accident[51].

The continuous, intensive monitoring of ^{137}Cs distribution and its transfer in several forests in Fukushima revealed detailed post-depositional redistribution of ^{137}Cs in forest ecosystems. The ^{137}Cs activity concentration of the above-ground vegetation in an evergreen coniferous forest decreased to less than 3 % of initial levels due to the depuration processes in the 8 years after the accident (FIG. 3a,b). Over the same period, the loss of ^{137}Cs from the canopy to the forest floor and uptake from root to tree reached quasi-equilibrium conditions[48] (FIG. 3c). Canopy contamination decreased more slowly in a deciduous forest[35,53,54], where the canopy and tree trunks retained 21% of the total ^{137}Cs inventory 8 years after the accident (FIG. 3d,e). Root uptake of ^{137}Cs from soil to tree organs coincided with deposition onto the forest floor, but the impact of root uptake was masked by the rapid reduction of canopy contamination and direct uptake from foliage and bark surfaces during the first year after the initial deposition (FIG. 3f). Nevertheless, once the ^{137}Cs loss from the contaminated canopy decreased several years after the fallout, the ^{137}Cs inventory in stem wood tended to increase, possibly due to root uptake[53,55]. This later uptake implies that the deciduous trees and ecosystems are slower to reduce their levels of radioactive contamination relative to evergreen forests.

Continuous monitoring of the depth distribution of ^{137}Cs in forest soil showed that ^{137}Cs activity concentrations were greatest in the litter layer for 2–3 years after fallout and then in the uppermost mineral soil layer on the forest floor, relating to the downwards migration of ^{137}Cs from decomposing organic matter on the forest floor[56–59]. The percentages of ^{137}Cs inventory in 0–2 cm of soil increased from 18% in 2011 to 48% in 2015 (REF.[60]). Downwards migration of ^{137}Cs from the uppermost soil layer to subsoil occurs slowly over the timescale of decades[58,60,61], although the continuous input of ^{137}Cs to the forest floor from litterfall can cause the downwards migration of ^{137}Cs in forest soil to appear slower (FIG. 3).

The loss of contaminants deposited on forest catchment occurs through erosion of sediment and organic matter through the groundwater-fed headwater streams[62,63] (FIG. 2), which had an annual discharge flux ^{137}Cs of less than 0.3% of the initial fallout to the catchment area[46,64–66]. Lateral transport of litterfall led ^{137}Cs to also accumulate at the bottom of hillslopes[67] — litter

Depuration
Reduction of radioactivity concentration in plants due to environmental processes.

Fig. 2 | **Migration and monitoring of ^{137}Cs in the terrestrial environment.** After the 2011 nuclear accident in Fukushima, Japan, ^{137}Cs was deposited on forested areas, agricultural land, grassland, paddy fields, and urban and residential areas. After deposition, ^{137}Cs migrated through soil and into rivers. Some of the ^{137}Cs in rivers was discharged into the ocean, including approximately 10 TBq of ^{137}Cs that was discharged from the Abukuma River basin to the Pacific Ocean between June 2011 and March 2017. In the years following the accident, there was extensive monitoring, including soil, groundwater and river water monitoring. The arrows in the figure represent ^{137}Cs transfer.

^{137}Cs activity concentrations along a steep hillslope were found to be 2.4–2.7 times greater than those of deposited organic matter under stream water in both Japanese coniferous and deciduous forests[68]. Such transport of ^{137}Cs along forest hillslopes to riparian areas downhill could enhance ^{137}Cs discharge from the forested area through drainage. In the experimental forested headwater catchment at Yamakiya, Japan, the ^{137}Cs activity concentrations of litter layers were in the range 37–150 kBq kg^{-1} (REF.[48]), with activity concentrations of 0.01 Bq l^{-1} to 0.1 Bq l^{-1} for the mobile ^{137}Cs (REF.[62]) stored in the litter leachate in the upper soil layer of the forest hillslope[69–71]. During storm events, the saturated zone of the soil expands upslope[62] into the area covered by thick, ^{137}Cs-contaminated organic layers, potentially leading to the temporary increase of the activity concentration of dissolved ^{137}Cs in stream water observed at the beginning of a rainstorm[62,72]. Despite the differences between the first and second phases of depuration in the vegetation and litter layer, the efficiency of ^{137}Cs removal from the forested catchment as a whole likely has not changed[66], and forested areas can, thus, act as a long-term source of dissolved ^{137}Cs in river waters[73,74]. Overall, some litter-bound ^{137}Cs was transported downslope[67,70] but as it can be discharged only through stream riparian zones[62,65], the ^{137}Cs fallout in forest ecosystems largely have not been transported out of the system.

Farmlands and grasslands. Approximately 40% of the 900 km^2 of farmlands and grasslands[12] shown in FIG. 1 received over 100 kBq m^{-2} of ^{137}Cs, for a total of 0.16 PBq of ^{137}Cs. The ^{137}Cs was deposited onto both vegetation, where it adhered to leaves[75] and was eventually transferred to surface soils[76,77], and directly onto the soil, where it was first retained in the upper several centimetres and then migrated to deeper layers. In July 2011, for example, 71%, 21% and 7% of ^{137}Cs in a meadow was present in the soil, litter and standing dead material, respectively[77]. Between July 2011 and January 2012, the percentage of ^{137}Cs retained in litter relative to the total ^{137}Cs inventory in soil dropped from 19% to 0.8% in a pasture and from 69% to 14% in a meadow[76]. Notably in farmland, the ^{137}Cs activity concentration in the tillage layer was relatively uniform with the depth of tillage[78]. In general, the ^{137}Cs migration into deeper soil depended on soil characteristics such as porosity, particle-size distribution and ^{137}Cs interception potential[76,79–82].

From the soil, ^{137}Cs was transferred to vegetation via plant uptake, the ratio of which depends on the plant species and soil properties such as soil pH and cation-exchange capacity[83]. The transfer factors (^{137}Cs activity concentration in agricultural products divided by ^{137}Cs activity concentration in dry soil) of various agricultural products have been intensively investigated and published[84,85]. For example, transfer factors of edible

parts in 24 vegetables ranged from 0.0001 to 0.0054 in a series of test cultivations[84]. The application of potassium is an effective measure to regulate the uptake of ^{137}Cs by plants for both farmland and grassland[86–88].

The lateral movement of ^{137}Cs from the soil layer was driven by water and sediment movement[89,90], which was influenced by the erosivity of rainfall, erodibility of soil, slope gradient and length, vegetation cover and vertical distribution of ^{137}Cs in soil. The ^{137}Cs wash-off from farmland and grassland, therefore, differed, and the activity concentration of ^{137}Cs in sediments eroded from farmlands (typically cultivated to the depth of 15 cm) was low and homogeneous compared with those from uncultivated farmland, owing to soil mixing by tillage[89,90]. The tillage resulted in an approximately 60% decrease in ^{137}Cs activity concentrations in eroded sediments in a cultivated farmland compared with an adjacent uncultivated farmland[90] Despite the low ^{137}Cs activity concentration of eroded sediments, soil erosion was enhanced by tillage, so that up to 15% of the initially deposited ^{137}Cs in a cultivated farmland was lost through run-off in 3 years during 2011–2014 (REF.[90]). In contrast, there was generally little lateral movement of ^{137}Cs in grasslands owing to permanent vegetation cover, which reduces surface run-off and soil erosion[89,90]. Annual grassland ^{137}Cs losses due to wash-off ranged from 0.0029 to 0.067% per year, given an annual rainfall of 1,200–1,600 mm (REF.[90]). Overall, the activity concentration in soil lost from grasslands showed huge variability and no significant decreasing trend over the observation period (2011–2014)[90]. ^{137}Cs washed off from farmland and grassland is transported to the rivers, where can be transported into the ocean, though a large part of agricultural land is poorly connected to the rivers[91], so the export into rivers is expected to be smaller.

Paddy fields. Approximately 37% of the paddy fields within the 80-km-radius zone of the FDNPP received over 100 kBq m^{-2} of ^{137}Cs fallout[12] (FIG. 1). At the time of the accident, paddy fields were not irrigated, so most

Fig. 3 | **^{137}Cs distribution and transfer in forests. a** | Initial fallout in cedar forests. During the initial fallout in 2011, 442 kBq m^{-2} ^{137}Cs was deposited onto experimental Japanese cedar forests in Yamakiya district, Kawamata town of Fukushima Prefecture, most of which was intercepted by the cedar needles and branches. The values without units show ^{137}Cs inventories (kBq m^{-2}) in each component, whereas the percentages in parentheses denote inventory ratio to total atmospheric fallout deposited on the forest following the accident. **b** | ^{137}Cs inventory in the cedar forest in 2018. Most of the ^{137}Cs had migrated to the soil via litterfall, throughfall and stemflow. The numbers next to the arrows indicate downwards (from the canopy to the forest floor) and upwards (from the soil to trees) migration flux of ^{137}Cs inventory (kBq m^{-2}), where the percentage values for throughfall, stemflow and litterfall represent the contribution of each pathway to the total downwards transfer of ^{137}Cs. **c** | Temporal changes in the ^{137}Cs inventory of the experimental Japanese cedar forests in kBq m^{-2} after the accident[33,35,40,44,51,57,60]. **d** | Initial fallout in broadleaf forests. In an experimental Japanese broadleaf (oak) forest at the same site, 451 kBq m^{-2} ^{137}Cs was deposited, with most of the ^{137}Cs reaching the litter layer. **e** | ^{137}Cs inventory in the oak forest in 2018. Like in the cedar forest, the most ^{137}Cs was present in the soil layer, with notable activity concentrations in the parts of trees. **f** | Temporal changes in the ^{137}Cs inventory of the experimental Japanese oak forests in kBq m^{-2}. The ^{137}Cs transfer between compartments 'canopy', 'wood' and 'soil' was calculated based on the temporal trends of activity concentrations and inventories in each forest component. The data are derived from the intensive field observations at the experimental forest, regarding the transfer from the canopy to the forest floor via hydrological and biological pathways[31–33,35,40,44], the temporal changes of ^{137}Cs inventories in different parts of tree components[51] and the distribution of ^{137}Cs in the organic and mineral soil layers[57,60].

of the ^{137}Cs deposited on the ground was retained in the upper several centimetres of soil initially[76], as reported in other undisturbed fields[76,92]. However, ^{137}Cs is mixed into the top 20 cm of paddy soil every year during soil puddling (the mixing of soil and irrigation water), rice planting and cultivation under flooding conditions from spring to early autumn[42,93–97], leading to an activity-concentration decrease in the upper centimetre of soil. For example, ^{137}Cs activity concentration in the top 4 cm of soil decreased from 3,200 Bq kg^{-1} to 2,400 Bq kg^{-1} during the observation period from 2012 to 2014 (REF.[93]).

After the initial fallout, there were main two routes for ^{137}Cs movement into and out of paddy fields: irrigation water inflow and discharge. The ^{137}Cs input to the paddy with irrigation water, which is drawn from streams, rivers and ponds, was negligible compared with the ^{137}Cs inventory of the soil, and contributed only 0.03–0.75% of the total ^{137}Cs in the paddy fields[93,98,99]. Discharge of ^{137}Cs from paddy fields was caused by flooding related to puddling of paddy soil and rainfall, enforced drainage before rice planting and harvesting, and wash-off due to rainfall. During the irrigation period, 0.67–7.3% of total ^{137}Cs in the paddy fields in 2011 and 0.0025–0.063% in 2013 and 2014 was discharged[97,98,100], with the puddling of paddy soil sometimes accounting for over 90% of the total annual ^{137}Cs discharge, especially in the first puddling after the fallout[97]. Subsequent ^{137}Cs discharge varied with the rainfall event and paddy maintenance.

Urban areas. Approximately 37% of the impacted urban areas (which were 7.9% of the area within 80 km of the FDNPP) received over 100 kBq m^{-2} of ^{137}Cs deposition[12] (FIG. 1). Urbanized areas comprise diverse components such as paved grounds, roofs, walls and permeable ground, and the activity per unit area varied across these components, owing to the differences in initial run-off and the following wash-off effects. For example, ^{137}Cs inventories of the paved ground and other building surfaces were 20% and less than 1% of that of permeable fields, respectively, 4 years after the FDNPP accident[101]. Indeed, in permeable fields such as a grassland in a residential area, ^{137}Cs migrated into the soil without ^{137}Cs loss due to run-off associated with precipitation[102–104]. In contrast, impermeable surfaces such as paved grounds and roofs retained ^{137}Cs on their surface upon deposition[105,106], but a large proportion of the ^{137}Cs was removed by the initial run-off and the following wash-off associated with precipitation[101], and was transported into river systems directly or via wastewater-treatment plants[107,108] (FIG. 2). The rapid removal of ^{137}Cs from urban surfaces is consistent with those observed after the CNPP accident[109–111].

The ^{137}Cs activity concentration of sludge in wastewater-treatment plants was highest during the initial period. In the case of a catchment in Fukushima city, which had a catchment area of 41 km^2 and ^{137}Cs deposition density of approximately 300 kBq m^{-2}, ^{137}Cs activity in dewatered sludge ranged from 467 to 6,158 Bq kg^{-1} from the end of April to July 2011 (REFS[112,113]). The ^{137}Cs activity concentration decreased over the first year post-accident by about one order of magnitude[112,113] (a rate faster than those observed in other land uses[90]), further demonstrating the rapid initial removal of ^{137}Cs from urban areas[112,113]. Although there is no quantitative information on the rate of ^{137}Cs loss from the various urban compartments or urban catchment, early removal of ^{137}Cs from urban areas implies that there was a large initial migration of ^{137}Cs from the urban area into river systems, with less migration as time proceeded.

Environmental migration of ^{137}Cs

After deposition onto forests and the ground, radionuclides migrated through the soil and to and through rivers, and eventually into the ocean (FIG. 2). Approximately 10 TBq of ^{137}Cs was discharged from the Abukuma River basin to the Pacific Ocean between June 2011 and March 2017, corresponding to 4.8% of the initial deposition[25,114]. As 13% of the initial deposition physically decayed in the period, 82% of the ^{137}Cs was retained in the catchment.

Migration through soil. Most of the fallout ^{137}Cs deposited on the soil surface was present in the upper 5 cm of soil[76,79,92], where it was sorbed by clay minerals[115,116] (FIG. 4a–c). Generally, the initial distribution was influenced by the first rainfall post-accident[117], but was not substantially impacted by subsequent heavy rainfall[61]. However, convective transport by flowing water, diffusive movement within the fluid, physical mixing and erosion of soil particles, and bioturbation caused slight migration of ^{137}Cs in the soils[118,119]. Furthermore, many studies have shown that the chemical form of ^{137}Cs, soil texture, organic matter and water permeability would have strongly influenced the distribution and migration of ^{137}Cs in soils[118,120], although few studies have quantified the downwards migration through continuous field monitoring.

As was observed in the environment, the ^{137}Cs rapidly migrated from the litter layer to the mineral soil surface in forest sites (FIG. 4a). However, the downwards migration of ^{137}Cs from the surface soil layer (0–5 cm) to deeper soil layers was slower in forests than in other land uses, and the relaxation mass depth, β, apparently did not change substantially over time because of continuous input from the litter layer (green line in FIG. 4d; see Supplementary Material for calculations of β)[60]. Downwards migration of fallout ^{137}Cs was fastest in abandoned paddy fields[121] (red line in FIG. 4d), where the ^{137}Cs activity concentration in the upper 10 cm of soil was fairly homogeneous by 3 years after the fallout (FIG. 4b). This increase in β contributes to a decrease in the air-dose rate (as ^{137}Cs in the top few centimetres of soil contributes most of the air dose[122]) but also reduces the efficiency of decontamination efforts, which typically involve scraping off the top 5 cm of soil. For example, approximately a third of the original fallout ^{137}Cs remained after decontamination attempts removed the top 5 cm of soil in an abandoned paddy field in 2015 (REF.[121]). The downwards migration (mixing) of ^{137}Cs in cultivated paddy-field soils also occurred in the years following the accident[93] (FIG. 4c). Indeed, the residence half-life of ^{137}Cs in the

Relaxation mass depth
Parameter quantifying the exponential penetration distribution of radionuclides in the soil profile, defined as the mass depth at which the activity concentration or inventory at the soil surface decreases to 1/e.

Air-dose rate
The amount of radiation energy in the air per unit time or the converted value to evaluate the health effect on the human body.

Fig. 4 | **Temporal changes in the vertical distributions and relaxation mass depth of ^{137}Cs activity concentrations. a** | Temporal changes of ^{137}Cs activity concentrations (kBq kg^{-1} soil) in forest soils plotted by depth (cm) and mass depth (g cm^{-2}) for the years 2011, 2013 and 2015. Litter layers are shown as brown bars. Relaxation mass depths β (g cm^{-2}) are labelled with a black arrow and their corresponding values. **b** | Temporal changes of ^{137}Cs activity concentrations and β in abandoned paddy field soils plotted by depth and mass depth for the years 2011, 2012 and 2014. **c** | Temporal changes of ^{137}Cs activity concentrations and β in cultivated paddy field soils plotted by depth and mass depth for the years 2012, 2013 and 2014. **d** | Scatter diagram of β in lands with various uses in Fukushima post-accident, with values from three soil types in grasslands and abandoned farmlands impacted by the Chernobyl Nuclear Power Plant (CNPP) accident for comparison. With time post-accident, ^{137}Cs generally migrated deeper into the soil and β increased. The migration rates and soil profiles differed between land uses and anthropogenic activities. Blue circles are urban lands[81,82] (number of sites, $n = 55–83$, $\beta = 0.33t + 0.86$, $r = 0.99$, $P < 0.0001$), red diamonds are abandoned paddy fields[121] ($n = 1$, $\beta = 1.3t + 0.72$, $r = 0.93$, $P < 0.01$), orange squares are cultivated paddy fields[93] ($n = 1$, $\beta = 0.56t + 6.95$, $r = 0.82$, $P = 0.18$), yellow squares are abandoned pasture and farmlands[76] ($n = 3$, $\beta = 0.09t + 1.17$, $r = 0.21$, $P = 0.73$), green triangles are forest sites[60] ($n = 3$, $\beta = 0.04t + 0.53$, $r = 0.56$, $P = 0.09$), white squares are CNPP-impacted sandy loam and loamy soils[204–206] ($n = 2–4$, $\beta = 0.37t + 2.40$, $r = 0.72$, $P = 0.28$), black diamonds are CNPP-impacted loamy sand and sandy soils[204] ($n = 1–7$, $\beta = 0.29t + 1.76$, $r = 0.89$, $P < 0.05$) and grey circles are CNPP-impacted organic soils[204,205] ($n = 1–3$, $\beta = 0.36t + 0.10$, $r = 0.87$, $P = 0.06$). Error bars represent standard deviations, solid lines represent significant correlations with time and dotted lines represent non-significant correlations with time. The β values were calculated without litter layers.

Fig. 5 | **Temporal trends in ^{137}Cs S_c values.** S_c ($m^2\,kg^{-1}$) is the normalized ^{137}Cs activity concentration in transported sediment and is obtained by dividing the ^{137}Cs activity concentration in sediment ($Bq\,kg^{-1}$) by the initial ^{137}Cs deposition on the plot or catchment of interest ($Bq\,m^{-2}$). Data for paddy fields until the third year after the accident were derived from REF.[124] and those in subsequent years were derived from REF.[125] based on measurements of ^{137}Cs activity concentration in a turbid water paddy field at paddling periods. Data for cultivated farmland and grassland were derived from REFS[90,126]. The data for forested catchments until 2.5 years after the accident were derived from the Koutaishi site and those in subsequent years were derived from the Setohachi site in REF.[62]. Data for urban catchments were derived from REF.[107]. Temporal trends of S_c at the years since the FDNPP accident (t, year) were expressed by the following equations; $S_c = 12e^{-18t} + 0.030e^{-1.3t} + 0.021e^{-0.031t}$ for paddy fields, $S_c = 0.068e^{-0.25t}$ for forested catchments and $S_c = 0.19e^{-0.40t}$ for urban catchments.

ploughed layer of paddy fields is estimated to be 9–24 years[123]. The values of β in urban sites revealed a linear increase of approximately 0.3 $g\,cm^{-2}$ per year (blue line in FIG. 4d, based on an average of 55–83 sites for each year post-accident).

^{137}Cs continues to migrate downwards in mineral soils, affecting the air dose. However, the percentage of ^{137}Cs inventory in soils deeper than 10 cm is still small, at less than 10% as of 2017 (REF.[60]). As downwards migration is likely continue, long-term future monitoring will be required.

Transport into rivers. Erosion and run-off of sediments and soils transported ^{137}Cs into rivers (FIG. 5), which was measured as S_c ($m^2\,kg^{-1}$), the ^{137}Cs activity concentrations in transported sediments ($Bq\,kg^{-1}$) normalized by local ^{137}Cs inventory at the time of initial deposition ($Bq\,m^{-2}$) after the accident[89,90,124]. The amount of ^{137}Cs transported varied between land-use types and with time after the accident. For example, S_c values for paddy-field experimental plots were 0.15 $m^2\,kg^{-1}$ initially and drastically decreased with time to 0.016 $m^2\,kg^{-1}$ by 2018 (FIG. 5). The decrease was most notable during the first year[97] and slower in subsequent years[124]; recent data[125] show no declining trend. In contrast, the S_c of grasslands and cultivated farmlands did not show a significant decrease with time[90,126]. In the farmlands, the S_c values were low, ranging from 0.0021 to 0.040 $m^2\,kg^{-1}$ (average of 0.015 $m^2\,kg^{-1}$), which is attributed to the redistribution of ^{137}Cs to deeper soils during ploughing. Grassland S_c values were more variable owing to erosion, ranging from 0.0018 to 0.15 $m^2\,kg^{-1}$ (average of 0.031 $m^2\,kg^{-1}$)[90,126], and the overall amount transported was low.

Notably, the S_c values for paddy fields showed larger values (maximum of 0.15 $m^2\,kg^{-1}$) than other land uses in the initial period and decreased one order of magnitude during the initial half-year after the accident (during the first phase) (FIG. 5). The following period (the second phase) showed a slower decrease in S_c with time down to 0.016 $m^2\,kg^{-1}$ in 2018, but the decrease was still at a faster rate than that observed for other land uses[124]. Compared with other land uses, paddy-field discharges had a marked impact on ^{137}Cs in downstream water bodies during the first phase[25,97,124]. Although no substantial discharge into rivers was reported during the second phase[98,100], the suspended sediment discharged from paddy fields is often described as being transported directly to the river (100% sediment connectivity), indicating the continuing importance of paddy fields to the downstream transport of ^{137}Cs through rivers. Decontamination by scraping the surface soil — the most effective measure for reducing the activity contamination of ^{137}Cs discharged[25] — was widely conducted, even within the evacuation zone[127] (BOX 1). In one case study, surface removal reduced the ^{137}Cs activity concentration in paddy soil to about 3% of the initial value[97]. These activities reduced the contribution of paddy fields to ^{137}Cs in river systems[25,97].

Both forested and urban catchments were sources of ^{137}Cs into rivers, with S_c values of 0.0031 to 0.064 $m^2\,kg^{-1}$ (mean of 0.030 $m^2\,kg^{-1}$)[62] and 0.034 to 0.26 $m^2\,kg^{-1}$ (mean of 0.083 $m^2\,kg^{-1}$)[107], respectively, from 1.5 to 4 years post-accident (3.6 and 1.7 times higher than the S_c values of paddy fields). S_c values in the urban catchment decreased with a rate of 0.40 $year^{-1}$ during 2012–2015, and a rapid decrease was found during 2012–2013. By contrast, S_c values in the forested catchment showed a decrease with a rate of 0.25 $year^{-1}$ during 2012–2016.

Wastewater treatment plant sludge values suggest that there was a rapid decline in the run-off S_c from urban areas, similar to that of paddy fields[112,113]. The relatively high S_c in urban areas can be attributed to decontamination activities, such as road-dust sweeping and brushing, resulting in preferential transport of sediment with a high ^{137}Cs activity concentration, though the detailed processes are not fully understood. Similarly, rapid decreases of S_c values in the paddy fields and the urban catchments in the first 2 years and slight decreases in the subsequent years were found. Together, these results point to the conclusion that catchments highly affected by human activities showed a rapid reduction of ^{137}Cs activity concentration in suspended sediment in river water[25].

Riverine transport. ^{137}Cs was present in rivers and other water bodies as dissolved ions (which are relatively easily absorbed by plants and aquatic organisms) and sorbed

Suspended particles
Small particles in water; the average diameter is several tens of micrometres in rivers around the Fukushima Daiichi Nuclear Power Plant.

K_d
Solid–liquid distribution coefficient; the ratio of the concentration of radionuclides sorbed on a specified solid phase to the radionuclide concentration in a specified liquid phase.

to suspended particles[128] (including 0–46% of total suspended particles as Cs microparticles)[129–131]. Land use in the river catchment had the greatest influence on the distribution and activity concentration of both dissolved and particulate ^{137}Cs (REFS[132,133]), but whereas catchment-soil type was an important factor impacting dissolved ^{137}Cs, precipitation and slope significantly influenced particulate ^{137}Cs. For example, a large rainfall event caused by Typhoon Roke in September 2011 resulted in substantial flooding, which significantly increased the riverine flux of particulate ^{137}Cs (REFS[134–136]). Additionally, particulate ^{137}Cs activity concentrations are affected by the presence of suspended fine particles such as silt and clay, which adsorb ^{137}Cs more efficiently than coarse particles such as sand[137,138], and by the organic-matter contents of suspended particles, as higher contents adsorbed more ^{137}Cs (REF.[139]). In the Abukuma River between June 2011 and August 2015, more than 96.5% of ^{137}Cs was transported in the particulate form, owing to a high activity concentration of sediment run-off caused by the steep mountains and heavy rainfall[25].

Most of the ^{137}Cs associated with riverine suspended particles was eventually transported into the ocean and little was redeposited. However, fine sediment can be deposited on the riverbanks after storm events — the percentage of ^{137}Cs deposited on the riverbank was estimated to be 0.77% of the total flux in the lower reaches of the Abukuma River after a typhoon in September 2011 (REF.[140]), and a typhoon in 2013 caused a 15% and 18% increase in the ^{137}Cs deposition and the air-dose rate[91,141], respectively. Similarly, exceptional deposition has been reported along riverbeds during large flood events when water levels drop, as well as in large lakes and reservoirs — over 70% of ^{137}Cs was retained in the case of Ogaki Dam[142,143].

One week after the accident, 70 ± 10 Bq l^{-1} of total ^{137}Cs (the sum of dissolved and suspended forms) was found in the river water in a small tributary of the Abukuma River in Fukushima city[144]. Across the catchment, the activity concentration of dissolved ^{137}Cs continuously decreased relative to the initial deposition[25,71,134,139,145–147] (FIG. 6a). The dissolved ^{137}Cs activity concentration in rivers decreased proportionally with the decrease in particulate ^{137}Cs, indicating that ^{137}Cs fixation[148] to mineral particles is not the primary cause of the decline in the activity concentration of ^{137}Cs in Fukushima rivers. If the amount of ^{137}Cs irreversibly adsorbed to the particles had increased, the ratio of particulate to dissolved ^{137}Cs (defined as K_d, a solid–liquid distribution coefficient[118]) should have increased with time, which was not observed.

A six-year intensive continuous monitoring of the particulate ^{137}Cs activity concentrations in rivers at 30 sites within 80 km of the FDNPP indicated an activity concentration decline in two phases[25,149]: the first year post-accident and years 1–6 post-accident. In the first phase, the particulate ^{137}Cs activity concentration quickly decreased, dropping to approximately 10 % of the initial value, with the rate of decline varying amongst catchments (FIG. 6b). In catchments with more than 30% of land use related to human activities (paddy fields, farmlands and urban areas, referred to here as PFU), which have a higher run-off than forested areas[90], there is a rapid decline of ^{137}Cs activity concentrations (yellow line in FIG. 6b). River catchments with low fractions of PFU (such as those with high forest cover) showed no such decline (purple line in FIG. 6b). The Abukuma River basin, for instance, has a high land-use coverage related to PFU (30%) and a high initial rate of decline in ^{137}Cs activity concentrations (2.9 year^{-1})[25]. The rate of particulate ^{137}Cs decline in the second phase was between 0.06 and 0.66 year^{-1} (an average of 0.34 year^{-1}), similar to those measured in reservoir lake sediments (0.33 and 0.44 year^{-1})[74,147], and the activity concentration was about 2% of the initial value by 6 years after the accident (FIG. 6b). The rate of decline was not correlated with catchment land use[25].

Box 1 | **Decontamination**

Decontamination of areas impacted by radionuclide fallout was undertaken after the Fukushima Daiichi Nuclear Power Plant accident to reduce public exposure. In the Fukushima Decontamination Pilot Project, conducted from 2011 to 2012, decontamination measures were tested to explore technical feasibility, efficiency and costs[209]. The Japanese Ministry of the Environment established decontamination guidelines based on the results of the project at the end of 2011 and provided an update in 2013 (REF.[210]).

Decontamination focused on residential areas such as buildings, roads, agricultural fields, bare ground in urban areas and forests adjacent to houses. In general, decontamination of impermeable surfaces (such as building surfaces and asphalt roads) included the manual removal of surface sediments, wiping, brushing, high-pressure water washing and scraping, with the appropriate methods untaken in stages that depend on decontamination efficiency[211]. Decontamination methods in agricultural fields included scraping off the surface soil and deep ploughing. For trees within 20 m of residential areas, their bark was washed and branches pruned.

The decontamination efficiency depended upon the objects to be decontaminated and the methodology used. The Decontamination Pilot Project reported that decontamination reduced 20–70% of the air-dose rate at 1 m above ground in agricultural fields and 40–95% of the rate on asphalt pavement. Soil scraping also showed relatively high air-dose reduction efficiency among the decontamination methods.

Decontamination outside of the evacuation zone was completed by 19 March, 2018 (REF.[127]). The decontaminated areas were more than 41,000, 12,000 and 15,000 ha for agricultural fields, forests and roads, respectively, over Fukushima and surrounding prefectures[212]. The total volume of soil and waste collected during decontamination was estimated at around 14 million m^3 (REF.[213]).

Objects or areas	Components	Methods
Artificial structures	Buildings and others (such as fences)	Removal of surface sediment; brushing
	Yards	Scraping off surface soil
Roads	Paved surfaces	Sweeping road dust; washing using water jet or brushing; surface scraping or removal
	Unpaved surfaces	Topsoil stripping; reversal tillage; covering using clean soil
Bare ground	Playgrounds and agricultural fields	Topsoil stripping; reversal tillage for fields with <5 kBq kg^{-1} of $^{134+137}$Cs; deep tillage for fields cultivated before decontamination
Vegetation	Lawns	Deep mowing or removal with soil
	Trees	Branch trimming; trunk washing
	Forest and forest floor	Litter and surface soil removal[214,215]

Fig. 6 | **^{137}Cs activity concentrations in rivers impacted by the FDNPP accident. a** | Activity concentrations of dissolved ^{137}Cs normalized by the initial deposition density, θ, in rivers affected by the Fukushima Daiichi Nuclear Power Plant (FDNPP) accident[25,71,134,139,145–147]. The black dotted line at the top shows the decrease in θ due to the physical decay of ^{137}Cs, the grey dashed lines are the upper and lower limits of the range in European rivers after the 1986 Chernobyl Nuclear Power Plant (CNPP) accident[21] and the blue dotted line shows the decline of θ in the Pripyat River after the CNPP accident ($\theta = 0.41e^{-13t} + 0.041e^{-0.43t} + 0.0023e^{-0.043t}$)[181,207]. The range of θ in Japanese rivers after the FDNPP accident is lower than that of European rivers after the CNPP accident. **b** | Activity concentrations of particulate ^{137}Cs normalized by the initial deposition density $C'(t)$ in rivers affected by the FDNPP accident[25,149]. The black dotted line shows the decrease in $C'(t)$ due to the physical decay of ^{137}Cs only. The grey curve indicates a double exponential equation fitted by all observed data at 29 monitoring sites $C'(t) = 0.40e^{-3.6t} + 0.062e^{-0.29t}$ ($P < 0.001$), including rivers in both forested catchments (19 rivers, plotted in purple) and catchments with more than 30% of land used for paddy fields, farmland and urban areas (referred to as PFU; 10 rivers, plotted in yellow). The PFU river line is plotted as $C'(t) = 1.5e^{-4.5t} + 0.090e^{-0.36t}$ ($P < 0.001$) and that of the forested area as $C'(t) = 0.059e^{-0.26t}$ ($P < 0.001$). The blue dotted curve shows the decline of activity concentrations of suspended ^{137}Cs in the Pripyat River after the CNPP accident[208] $C'(t) = 1.1e^{-2.3t} + 0.26e^{-0.36t}$ ($P < 0.01$). The initial decline rate averaged over the 29 river sites in Fukushima was faster than that in the Pripyat River. The shaded areas indicate the 95% confidence intervals of the regression curves. **c** | Quarterly loss of ^{137}Cs (flux of ^{137}Cs normalized by the initial deposition in the watershed) between June 2011 and March 2017 (REFS[25,114]). The yellow and purple lines indicate averaged values at monitoring sites with over 30% coverage of PFU and forested areas, respectively, with shaded areas representing the ranges in the loss of ^{137}Cs from the watersheds. Two sites with over 30% PFU were monitored from June 2011 through September 2012 and eight from October 2012 through March 2017. Four forested sites were monitored from June 2011 through September 2012 and nine from October 2012 through March 2017. Sites where there is a large reservoir lake in the watershed were excluded from the plot.

Based on the loss of particulate ^{137}Cs from the catchment (calculated by dividing the quarterly ^{137}Cs fluxes by the initial deposition in the watershed), only approximately 3% of the fallout ^{137}Cs was transported from the Abukuma River watershed to the Pacific Ocean between June 2011 and March 2017 (REFS[25,114]). Half of this ^{137}Cs (1.5% of the total fallout) was lost within the first phase (June 2011 to Mar 2012)[25], owing to high initial ^{137}Cs activity concentration and a heavy-rainfall event caused by Typhoon Roke in September 2011 (REF.[136]). The flux was also affected by the upstream land use, and the primary sources of ^{137}Cs in the first phase were estimated to be the PFU areas (FIG. 6c). Indeed, 85% of the ^{137}Cs transported from the Abukuma River into the ocean was of PFU origin, despite comprising only 38% of the land coverage. In the second phase, the loss of ^{137}Cs from the upstream PFU decreased (FIG. 6c), as did the overall ^{137}Cs flux from rivers into the ocean.

The high percentage of anthropogenic land use is likely responsible for the quick environmental recovery of the river water in Fukushima. The slowing decline of riverine ^{137}Cs implies that forests store ^{137}Cs over the long term, and the relative importance in ^{137}Cs storage increases with time[150]. Similarly, despite the declining riverine ^{137}Cs activity concentrations after 2012 (REFS[25,149]), the relative importance of riverine ^{137}Cs fluxes into the ocean increases over time, as the direct release from the FDNPP decreases at a faster rate[151].

Other radionuclides

In addition to ^{137}Cs, the FDNPP accident released substantial amounts of other radionuclides, such as activation (^{239}Np and ^{59}Fe) and fission products (^{131}I, ^{134}Cs (^{133}Cs), ^{110m}Ag (^{109}Ag), ^{132}Te, ^{132}I, ^{140}Ba, ^{140}La, ^{91}Sr, ^{91}Y, ^{95}Zr and ^{95}Nb)[152], as well as 6,000–12,000 PBq of ^{133}Xe (REFS[3,4]), 43 TBq of ^{89}Sr, 3 TBq of ^{90}Sr (REF.[5]), 3.9 MBq of total U and 0.023 TBq of $^{239+240}$Pu (REF.[6]). Notably, the rapid decay of short-lived radionuclides such as ^{131}I and ^{132}Te/^{132}I reduced total radiation sources by 15% in the first month[5].

Some fallout radionuclides (besides ^{137}Cs) were detected in soil samples one month after the accident[152]. For instance, high deposition densities of ^{129m}Te and ^{110m}Ag were observed north-west of the FDNPP site[11]. Environmental data show fairly consistent ratios between ^{137}Cs and ^{129m}Te or ^{132}Te, indicating that Cs and Te behave in the same way[153]. The ratio of ^{110m}Ag to ^{137}Cs is several factors higher in the north-west region near the FDNPP[102]. Although radioiodine (^{131}I and ^{129}I) was highly concentrated north-west of the FDNPP[11,154,155], and the deposition density of ^{131}I above 3 MBq m^{-2} was distributed up to approximately 20 km north-west of the FDNPP as of 2 April 2011 (REF.[156]), it was also concentrated south of the FDNPP[156]. The difference between the radioiodine and ^{137}Cs patterns is possibly the result of differences in the ways these materials settle out of the air[11]. Moreover, in the highly contaminated soils from the Mano catchment (north-west Fukushima), the proportion of Pu originating from the FDNPP was very low (<7.0 ± 4.5% of the total Pu present), suggesting a low deposition of FDNPP-derived Pu in this area relative to the global fallout[157].

Compared with ^{137}Cs activity concentrations, ^{90}Sr activity concentrations were generally 3–4 orders of magnitude lower[158] — ^{90}Sr:^{137}Cs ranged from 10^{-2} to 10^{-4} in surface soils within the 50-km-radius area from the FDNPP[159]. Specifically, there was a maximum deposition density of 5.7×10^3 Bq m^{-2} ^{90}Sr (REF.[11]) and activity concentration of ^{90}Sr in soil samples ranging from 0.2 to 23.4 Bq kg^{-1} (REFS[160–163]), which are similar to the background level (0.2 to 17.7 Bq kg^{-1})[162]. Therefore, these radionuclides have a very small impact on the terrestrial environment of Fukushima.

Soil profiles indicate that ^{90}Sr, ^{129}I and ^{131}I migrated downwards more quickly than ^{137}Cs (REFS[92,158,164,165]). ^{90}Sr:^{137}Cs in soil samples increased with depth by up to one order of magnitude by 2013 and 2014 (in other words, the downwards migration of ^{90}Sr was much higher than that of ^{137}Cs)[158]. The downwards migration rate of ^{129}I was approximately four times faster than ^{137}Cs in the same place[165], due to lower K_d values. Although the initial depth profile of ^{131}I was similar to ^{137}Cs (REF.[92]), the subsequent downwards migration rate of ^{131}I was slightly higher[164]. In contrast, ^{110m}Ag was used in the sediment fingerprinting in a different area impacted by fallout, because of the similarities to ^{137}C in terms of its depth profile, downwards immobility and K_d values[102,166,167].

Relatively long-lived radionuclides, including ^{90}Sr, ^{129}I and ^{3}H, were observed in rainwater, river water and groundwater after the FDNPP accident from 2011 to 2013 (REFS[13,63,168–170]). For instance, the Niida River had an estimated particulate ^{129}I flux of 7.6–9.0 kBq month^{-1} during September and October 2013 (REF.[168]), corresponding to around one ^{129}I for every million ^{137}Cs. In 2011, groundwater samples taken in the Yamakiya district slightly exceeded the natural background level of ^{3}H after the FDNPP accident[63], and riverine ^{3}H activity concentrations were still higher than the background level in 2012 (REF.[169]); activity concentration decreased with time in both places.

Comparison with Chernobyl

Natural comparisons have been drawn between the impacts of the accidents at the CNPP and the FDNPP. Overall, the FDNPP accident was less environmentally impactful than the CNPP accident — a total activity of 5,300 PBq (excluding noble gases) was released during the latter[2], compared with 520 PBq by the FDNPP[2]. In particular, the CNPP accident released very highly radioactive fuel particles (mainly uranium dioxide) with refractory and intermediate volatility elements[171]. Furthermore, an estimated 85 PBq (REF.[2]) of ^{137}Cs was deposited on land by the CNPP accident, including over 1,480 kBq m^{-2} onto 2,800 km^2 (an area 11 times larger than the area around Fukushima with over 1,480 kBq m^{-2}) and over 100 kBq m^{-2} onto 25,000 km^2 (REF.[172]). Large-scale decontamination efforts[25] after the FDNPP accident (BOX 1) also influenced the differences in the environmental impacts of the accidents. The comparisons between the CNPP and the FDNPP accidents here focus on ^{137}Cs.

Initial interception of the atmospherically deposited ^{137}Cs by forest canopies was similar between forests affected by the CNPP and the FDNPP accidents[31,33,173–175], and was mainly controlled by the biomass of the trees present[176]. This similarity suggests that the interception in both evergreen coniferous and deciduous forests is controlled by similar processes. The site-specific conditions, such as tree species, were less critical than forest density or the presence or absence of leaves (for deciduous tree species).

In both accidents, after initial interception, fallout ^{137}Cs was removed by rainwash and litterfall. The effective half-lives (T_{eff}) of canopy ^{137}Cs during the first phase post-accident were similar for FDNPP-affected Japanese cedar (T_{eff}: 87 days) and CNPP-affected Norway spruce (T_{eff}: 97 days)[33]. However, half-lives for the second phase were likely slower for Japanese cedar (T_{eff}: 490~780 days)[33,53] than Norway spruce (T_{eff}: 195 days)[33,173]. Experimental quantification of radiocaesium loss from the contaminated canopy reported that ^{134}Cs leaching by rainwash depended on the frequency of rainfall episodes during the sampling period[50]. The observed slower loss rate of canopy ^{137}Cs in Fukushima could be attributed to greater closed canopy coverage owing to the greater needle biomass (4.0 kg m^{-2}) and longer needle longevity (that is, dead needles remained in the canopy) of Japanese cedar than Norway spruce[44,51].

Japanese red pine in Fukushima and CNPP-affected Scots pine forest in Finland have similar needle biomass (0.4–0.5 kg m^{-2} and 0.91 kg m^{-1}, respectively) and had similar canopy ^{137}Cs T_{eff} during the second phase (510 days and 530 days, respectively)[53,177]. In deciduous

Effective half-lives
The time interval required for radioactivity in an environment to decrease to half its original value due to radioactive decay and biogeochemical processes.

broad-leaved forests (such as ones dominated by Konara oak) in Fukushima, the T_{eff} of the canopy ^{137}Cs of the second phase was in the range 390~1,200 days (REFS[33,53]). Despite the absence of direct deposition onto foliage, some data indicated that the deciduous broad-leaved stand showed a longer effective half-life of canopy ^{137}Cs than Norway spruce in the Chernobyl-affected area[173]. Due to larger amount of rainfall, the depuration rate of ^{137}Cs trapped in the canopy of trees during the first phase was faster in Fukushima, but was often slower during the second phase in many tree species with larger biomass, such as Japanese cedar.

The longer ^{137}Cs T_{eff} of the Fukushima forest resulted in prolonged ^{137}Cs deposition flux onto the forest floor over a long time period and maintained the apparent slower downwards migration rate of ^{137}Cs observed in Japanese forest soils[60,76] compared with forest soils impacted by the CNPP accident (FIG. 4d). In both cases, fallout ^{137}Cs flowed to streams, but the fallout-normalized activity concentration of both particulate and dissolved ^{137}Cs in Fukushima streams was much lower than in those impacted by the CNPP. For example, the ^{137}Cs activity concentrations of suspended sediment in forested headwater catchments were, on average, one order of magnitude lower[178] than those of the Pripyat River[178] near Chernobyl (FIG. 6b). Dissolved ^{137}Cs activity concentrations were approximately two orders of magnitude lower[62] in Fukushima versus the Pripyat River, and at least one order of magnitude in larger forested catchments[25,147] (FIG. 6a).

These differences in relative ^{137}Cs discharge from forested areas between the two regions can be explained by different topography and rainfall, as Fukushima has steeper topography and more run-off. Thus, Fukushima rivers transported more ^{137}Cs-bound suspended particles than Chernobyl-affected rivers. The particulate fraction in Fukushima exceeded 95%, typically greater than 98% (REF.[25]), whereas it was around 33% in the Pripyat River[179]. Additionally, the lower activity concentration of the dissolved ^{137}Cs (high K_d values)[25,128,134,135], possibly due to a higher run-off in Fukushima, also contributes to these differences.

Like the rivers in Fukushima, the particulate ^{137}Cs activity concentration in Pripyat River declined relative to the initial deposition (FIG. 6b), although the rate of decline in Fukushima was approximately 1.6 times higher than that of the Pripyat River in the first phase post-accident[180]. The difference in the decline rate was smaller in the second phase and, by 6 years post-accident, the C' (particulate ^{137}Cs activity concentration relative to initial deposition) in rivers in Fukushima was about 40% of the Pripyat River (FIG. 6b). After the CNPP accident, the dissolved ^{137}Cs activity concentration in 25 European rivers declined in three exponential components[21], but the dissolved ^{137}Cs activity concentration normalized by the initial deposition, θ, varied by about two orders of magnitude (shaded area in FIG.6a). The θ values of Japanese rivers tended to be one or two orders of magnitude lower than the lowest European rivers. This difference suggests that Japanese rivers recovered from ^{137}Cs more quickly (FIG. 6a), although the differences were also impacted by the higher K_d values (indicating that particulate ^{137}Cs did not easily dissolve) of the Japanese rivers. In European rivers, the coverage of inland water (ponds, lakes and wetlands) showed the most significant positive correlation with θ (REF.[21]), but the coverage of residential areas and the higher dissolved-ion concentrations, represented as the electrical conductivity of river water, positively correlate with θ in Japanese rivers[132,133], suggesting that lower ion concentration in river waters due to high relief and higher run-off by monsoonal climate can also be responsible for the lower dissolved ^{137}Cs in rivers.

Three main mechanisms have been proposed to contribute to the decline in dissolved ^{137}Cs of the river waters[181]: loss of ^{137}Cs activity from the catchment; vertical ^{137}Cs migration to deeper layers of soil; and slow chemical fixation of ^{137}Cs to soil particles. As the loss of ^{137}Cs in Fukushima catchments was low (4.8% in 6 years), the first mechanism likely played little role in the dissolved ^{137}Cs declines there[25,114]. Instead, downwards ^{137}Cs migration could be responsible for the dissolved ^{137}Cs decline in Fukushima rivers. The differences between ^{137}Cs in cultivated versus abandoned paddy field surface soils (3.4 kBq kg^{-1} in cultivated soils in 2012 versus 23 kBq kg^{-1} in abandoned paddy soils in 2011, FIG. 4b,c) and the correspondence between the declines in ^{137}Cs in suspended sediment (blue dots, FIG. 5) and particulate ^{137}Cs in rivers with high PFU cover (FIG. 6b) both point to this mechanism being the most influential. In contrast, gradual chemical fixation of ^{137}Cs (the third mechanism) is likely responsible for the decline of dissolved ^{137}Cs in the Pripyat River. This fixation is indicated by the observed slightly faster rate of decline in dissolved ^{137}Cs than the rate of particulate ^{137}Cs decline (in other words, K_d was increasing over time)[182] (FIG. 6a,b).

Overall, the terrestrial environment and rivers experienced a rapid reduction of the exposed radioactivity in Fukushima, but forests followed a trajectory similar to that of the CNPP-impacted forests. Anthropogenic activities in Fukushima, such as rice and vegetable cultivation and residential activities in the upstream area, have led to a rapid decline in the activity concentration of ^{137}Cs of surface soils, relative to CNPP-impacted soils. Differences in geographic and meteorological factors, such as rainfall, storm events, run-off and topography, also influenced the differences in environmental recovery from ^{137}Cs between the two major accidents.

Broader impacts

Detailed monitoring and analysis of ^{137}Cs dynamics in Fukushima undertaken following the FDNPP accident revealed the gradual transferral of fallout ^{137}Cs in soil and water to the ecosystem. The International Atomic Energy Agency (IAEA) working group on 'Modelling and Data for Radiological Impact Assessments' (MODARIA) summarized the dataset to simulate radionuclide transfer in the environment and to assess exposure levels of the public and in the environment, and these transfer parameters are summarized in a IAEA Technical Report[118]. After the FDNPP accident, the transfer data of many types of crops and animals were available from the early stages of contamination.

In Fukushima, leafy vegetables were contaminated by ^{137}Cs and ^{131}I immediately after the accident[183], followed by a declining trend in activity concentrations[184], and found to be similar to the reported values[118]. The brown rice was also contaminated by ^{137}Cs, with a transfer rate three to four times higher than the pre-accident level in the first years after the accident. However, these rates rapidly returned to the same level as before the FDNPP accident in the third year post-accident[185]. Japanese tea plants were contaminated by the translocation of ^{137}Cs deposited onto old leaves and twigs into new leaves, but ^{137}Cs levels in new tea leaves did eventually decline, similar to what was observed after the CNPP accident[186]. The transfer of ^{137}Cs to wild animals and subsequent decline were also generally similar to these seen after the CNPP accident, although sika deer in Japan did not decline during the period 2011–2015 (REF.[187]).

In a notable example of the transfer of radionuclides through the ecosystem after the FDNPP accident, the activity concentration of ^{137}Cs in freshwater fish[132,188] is highly correlated with the activity concentration of dissolved ^{137}Cs in the water[118,189]. As dissolved riverine ^{137}Cs declines, which is related to the decrease in the ^{137}Cs activity concentrations of suspended particles[73,147] and leaf litter in streams[68], so should the ^{137}Cs activity concentrations in fish. Indeed, the activity concentration of ^{137}Cs in fish is expected to continue to decline in Fukushima, rather than experiencing the longer-term persistent contamination that occurred in European freshwater fish after the CNPP accident[23,24].

Overall, these examples and the discussion here highlight that ^{137}Cs transfer in terrestrial areas is not a simple chemical process but also involves hydrological processes and material cycles.

Future perspectives

Recently, owing to strong demand to reduce greenhouse-gas emissions and for air-pollution measures, a growing number of nuclear power reactors are in operation and under construction[1] in Asia, especially in China. Against the background of the rapid expansion of nuclear power, Fukushima environmental monitoring data have a critical bearing on future contingency and remediation planning to tackle fallout contamination of populated areas. An enormous amount of data has been obtained from the Fukushima nuclear accident. However, emergency environmental data is often regarded as only useful to inform the public until radiation levels return to allowable levels, and publically available data published on governmental websites post-accident are being lost. Some of the environmental data collected in response to the FDNPP accident are available through the Database for Radioactive Substance Monitoring Data (maintained by the Japan Atomic Energy Agency)[190] and CRiED (maintained by the University of Tsukuba[191]), but with the passage of time since the accident, it is increasingly difficult to obtain the budget for the continued archiving and reporting of these data.

The IAEA has updated countermeasures guidelines[192], including the Manual on Sampling Methods[193], to account for lessons learned from the FDNPP accident. However, each country must formulate a feasible and concrete sampling and measurement protocol, taking the FDNPP lessons into consideration. Immediately after the Fukushima accident, an interdisciplinary group of scientists was formed[194] and members from the fields of nuclear physics, radiochemistry, geochemistry, atmospheric science and hydrology collaborated with the government[11,32,195]. This group was able to sample soil quickly after the accident, providing key data on the initial contamination levels and subsequent environmental recovery. To prepare for possible nuclear emergencies, we hope that national and local governments will take on the challenge of establishing cross-sectoral cooperation between departments responsible for air and water monitoring and agriculture, beyond just the nuclear power sector. In such a case, the information should be thoroughly disclosed and a series of checks should be in place.

In Fukushima, decontamination for environmental restoration was effective in reducing air doses, but it was also costly[196]. Moreover, a greater emphasis was placed on relieving the anxiety of the residents, rather than on scientific evidence for the establishment of decontamination targets and areas. In this respect, international joint research bodies will be needed to implement sufficient and effective measures, referring to Fukushima's lessons.

Long-term environmental monitoring in Fukushima should continue and be supported by the Japanese government, although there is no financial prospect for such activities at present. Examples should be taken from the international joint studies between the United Kingdom, France, Norway, Germany, Japan and others that have continued in the 30 years after the CNPP accident[171]. During this extended monitoring period, various environmental events such as wildfires[197,198] have occurred in areas affected by the CNPP accident, and further migration of radionuclides has occurred as a result of these events. Fukushima has high rainfall, dense forests, paddy fields and other land uses not found in Chernobyl, as well as a high possibility of landslides triggering large sediment outflow. Therefore, it is essential to study the future migration of radionuclides, especially in order to assess the effects of the accident in East Asia.

The comprehensive datasets acquired in response to the FDNPP accident have the potential to advance the understanding of Earth and environmental sciences, both basic and applied, beyond the impacts of this single event. For example, the combined use of natural tracers such as $^{210}Pb_{ex}$ (REF.[199]) and ^{7}Be (REF.[200]), with stable isotopes[201], and these data could lead to advances in tracing the transport and fate of radioactive materials. The dense and continuous monitoring network established following the accident also provides opportunity for validation of natural radionuclides as tracers of sediment sources and for mass budgeting[202], evaluation of the behaviour of substances and pollutants from various sources and organisms at local scales and prediction of the response of material circulation to global environmental changes.

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