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Retention of potentially mobile radiocesium in forest surface soils affected by the Fukushima nuclear accident

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The fate of ^{137}Cs derived from the Fukushima nuclear accident fallout and associated radiological hazards are largely dependent on its mobility in the surface soils of forest ecosystems. Thus, we quantified microbial and adsorptive retentions of ^{137}Cs in forest surface (0–3 cm) soils. The K_2SO_4 extraction process liberated 2.1%–12.8% of the total ^{137}Cs from the soils. Two soils with a higher content of clay- and silt-sized particles, organic carbon content, and cation exchange capacity showed higher ^{137}Cs extractability. Microbial biomass was observed in all of the soils. However, the ^{137}Cs extractability did not increase after destruction of the microbial biomass by chloroform fumigation, providing no evidence for microbial retention of the Fukushima-fallout ^{137}Cs. The results indicate that uptake of ^{137}Cs by soil microorganisms is less important for retention of potentially mobile ^{137}Cs in the forest surface soils compared to ion-exchange adsorption on non-specific sites provided by abiotic components.

The Fukushima Daiichi nuclear power plant (NPP) accident, triggered by a catastrophic earthquake (M9.0) and resulting tsunami on March 11, 2011, released a huge amount of radiocesium (^{137}Cs and ^{134}Cs; physical half-lives of 30.1 and 2.1 years, respectively) into the atmosphere[1], and consequently caused serious radioactive contamination of the soils in a range of terrestrial ecosystems over a wide area[2].

Among the terrestrial ecosystems, forest ecosystems have received the most attention, mainly for two reasons. First, there have been observations[3,4] that, in forest ecosystems, ^{137}Cs deposited by the Chernobyl NPP accident has still been observed in topsoils (including the litter layer or O horizon) and has become a major potential source for soil-to-plant transfer for the long term. There are very few countermeasures to effectively reduce the cycling of ^{137}Cs in the forest ecosystems[5]. Therefore, long-term external and internal (through consumption of food products from ^{137}Cs-contaminated forests) radiation exposure in the affected areas such as the Fukushima NPP area is possible. Second, forest ecosystems occupy over 70% of the entire land area in the Fukushima prefecture[6] and 69% of the total land area of Japan is covered by forest[7].

The fate of ^{137}Cs deposited in forest ecosystems, and the associated radiation risks, are largely dependent on the mobility of the ^{137}Cs in the topsoils. It is generally accepted that ^{137}Cs can be strongly, almost irreversibly, fixed by clay minerals in the mineral soil layers[8,9], resulting in a low migration rate and reduced availability for uptake by plants. However, there is evidence that a considerable percentage of ^{137}Cs remains in mineral soil layers as an easily exchangeable (loosely bound) form over a prolonged period of time[10–12]. It has been reported that even one decade after the Chernobyl fallout, high ^{137}Cs concentrations were still observed in the bottom layer of the O horizon and in the upper (transient) layer of mineral soils in Russian forests[3]. It was also found that the amount of exchangeable ^{137}Cs in the soils is one of the key factors governing the availability of ^{137}Cs for transfer to other forest components[3].

Microorganisms in soils are believed to influence the cycling of ^{137}Cs in forest ecosystems[13,14], because they are able to mobilize and subsequently accumulate ^{137}Cs (ref. 14–18). Therefore, it is conceivable that soil microorganisms are responsible for not only the transfer of ^{137}Cs to plants and deeper soil layers but also the retention of

^{137}Cs in the upper soil layers[19]. Nevertheless, the microbial involvement in the retention process of the mobile ^{137}Cs in the natural soil environment is still poorly understood[18–21].

This study presents the first investigation into the microbial retention of the Fukushima-fallout radiocesium in soils. Surface soil samples were collected from five forest sites (FR-1 to FR-5) in the city of Fukushima with different vegetation and soil properties approximately one year after the accident. FR-1 and FR-2 were dominated by broad-leaved trees and FR-3, FR-4, and FR-5 were dominated by coniferous trees. The soils have been classified as Fluvisols (FR-1, FR-2, and FR-3) and Andosols (FR-4 and FR-5). The amount of radiocesium bound to microorganisms and abiotic components (minerals and organic materials) in the soils was estimated using a chloroform fumigation-extraction method[22].

Results

Organic carbon (C) and ^{137}Cs in the soil samples. The soil organic C (SOC) content in the top 0–3 cm of soil ranged from 98 to 253 gC kg^{-1}-dry soil, with higher content in coniferous-dominated forests (FR-3, FR-4, and FR-5) than that in broadleaf-dominated forests (FR-1 and FR-2). The activity concentration of ^{137}Cs in the top soil was relatively similar between the sites, and ranged from 3380 to 4390 Bq kg^{-1}-dry soil (see Table S1).

K_2SO_4-extractable C and microbial biomass C (MBC). The amount of C extracted with K_2SO_4 from non-fumigated soils was 0.27–0.64 gC kg^{-1}-dry soil, which represented only 0.1%–0.3% of the total SOC. The amount of K_2SO_4-extractable C increased after the chloroform fumigation treatment for all five of the soils (Fig. 1a). From the amount of extractable C before and after chloroform fumigation, the amount of MBC was estimated to be ~1.29–2.58 gC kg^{-1}-dry soil, which corresponded to ~0.5%–1.8% of the total SOC, and was larger than that of the K_2SO_4-extractable C in the non-fumigated soils.

K_2SO_4-extractable ^{137}Cs. At all the sites, a considerable amount of ^{137}Cs was extracted with K_2SO_4 from the non-fumigated soil samples (Fig. 2). Although the amount of ^{137}Cs inventory in the soils was rather similar, the amount of K_2SO_4-extractable ^{137}Cs differed greatly from 72 to 538 Bq kg^{-1}-dry soil between the soil samples. The larger amount was found in the Andosol soils collected at two dense coniferous forests (FR-4 and FR-5) (see Fig. 2 and Table S1). The amount of K_2SO_4-extractable ^{137}Cs represented from 2.1% to 12.8% of the total ^{137}Cs inventory in the non-fumigated soil samples (see vertical axes in Fig. 3).

For all of the soils, no increase in the amount of K_2SO_4-extractable ^{137}Cs was observed after the chloroform fumigation treatment (Fig. 2). On the contrary, a significant (by a t-test at a 5% significance level) decrease in the amount of K_2SO_4-extractable ^{137}Cs was found in two of the soils (FR-3 and FR-4).

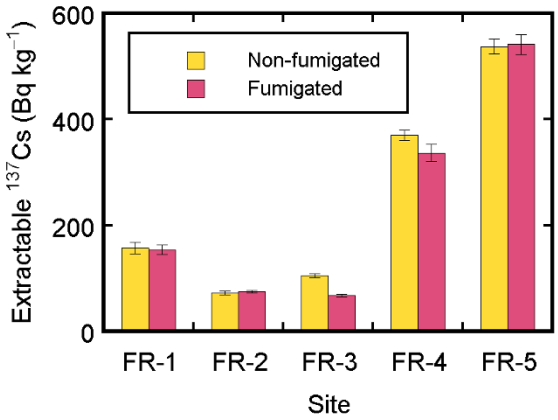

Figure 2 | Amount of ^{137}Cs extracted with K_2SO_4 from five forest surface (0–3 cm) soils before (non-fumigated) and after (fumigated) chloroform fumigation. Error bars indicate the standard deviation of the mean (N = 3).

Water-extractable C and ^{137}Cs. The water extraction liberated similar, but smaller, amounts of C from the non-fumigated soils as compared to that obtained via K_2SO_4 extraction (Fig. 1b). The amounts of MBC estimated from the water extractions were very similar to those estimated from the K_2SO_4 extractions. However, ^{137}Cs showed a contrasting extractability between the two extraction methods. The concentrations of ^{137}Cs in the water extracts from the FR-3 and FR-4 soils were all less than the detection limit. The concentration in the water extract from the FR-5 soil was close to the detection limit (0.0023 Bq mL^{-1}). As a result, the amount of water-extractable ^{137}Cs was estimated to be <26.3 Bq kg^{-1}-dry soil for the FR-3 and FR-4 soils and 22.8 Bq kg^{-1}-dry soil for the FR-5 soil.

Relation between K_2SO_4-extractable ^{137}Cs and the soil properties. The K_2SO_4-extractable ^{137}Cs (as a % of the total ^{137}Cs in the soil) showed no statistically significant (at a 5% significance level) correlation with the soil properties (Fig. 3), including the SOC (gC kg^{-1}) (correlation coefficient r^2 = 0.55); K_2SO_4-extractable C (gC kg^{-1}) (r^2 = 0.23); MBC (gC kg^{-1}) (r^2 = 0.18); clay-sized particle (<0.002 mm) content (g kg^{-1}) (r^2 = 0.34); silt-sized particle (0.002–0.02 mm) content (g kg^{-1}) (r^2 = 0.46); soil pH (r^2 = 0.05); cation exchange capacity (CEC; cmol kg^{-1}) (r^2 = 0.57); and base saturation (%) (r^2 = 0.05). However, it was clear that the ^{137}Cs extractability was high in two dense coniferous forest soils (FR-4

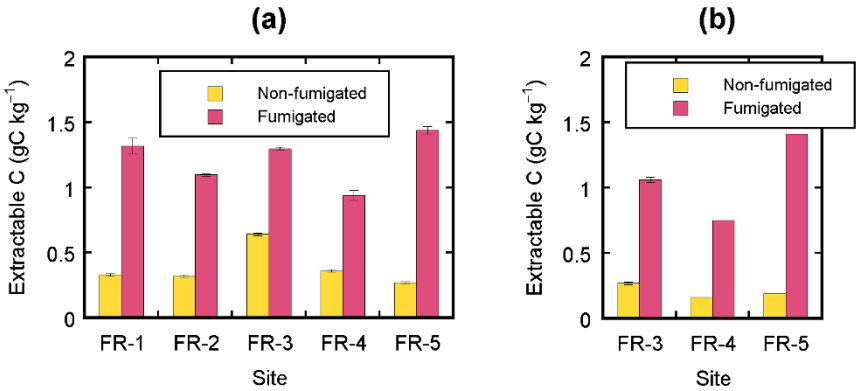

Figure 1 | Amount of C extracted with (a) K_2SO_4 and (b) water from five forest surface (0–3 cm) soils before (non-fumigated) and after (fumigated) chloroform fumigation. Error bars indicate the standard deviation of the mean (N = 3). The water extraction for the FR-4 and FR-5 soils was not replicated (N = 1).

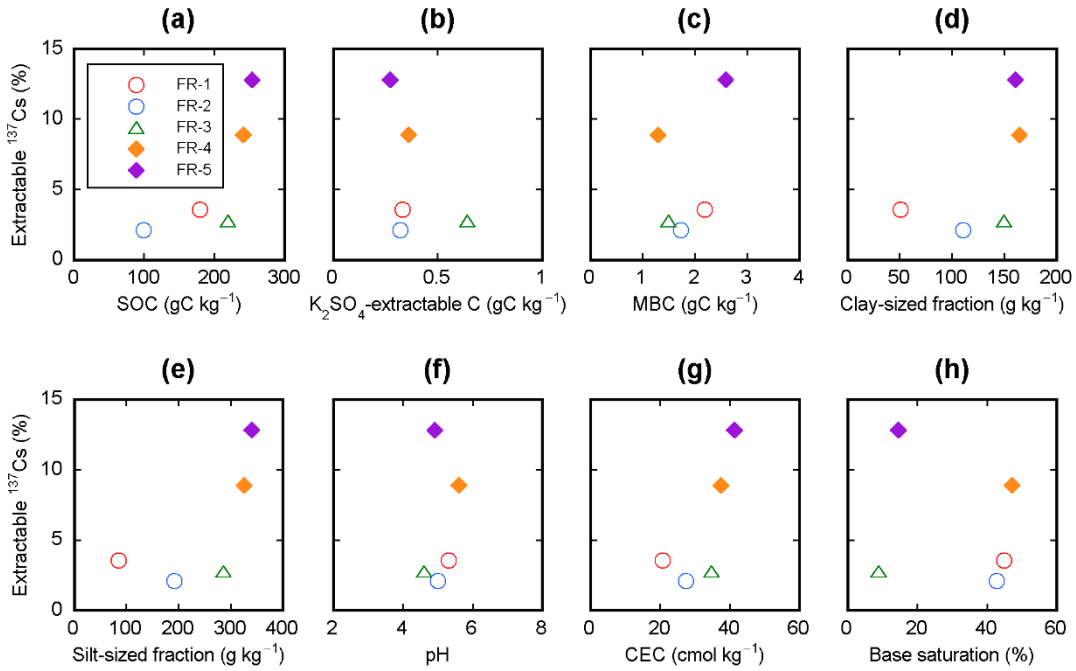

Figure 3 | Correlation between the K_2SO_4-extractable ^{137}Cs (as the % of total ^{137}Cs in the soil) and soil properties: (a) SOC; (b) K_2SO_4-extractable C; (c) MBC; (d) clay content; (e) silt content; (f) pH; (g) CEC; and (h) base saturation.

and FR-5) that had a higher SOC, clay, silt content, and CEC. A strong positive correlation ($r^2 = 0.93$, $p < 0.01$, Fig. 4) was found between the ^{137}Cs extractability and the relaxation length parameter (α in cm, an index characterizing the ^{137}Cs penetration through 0–20 cm of mineral soil) determined on the basis of the cumulative ^{137}Cs inventory data obtained in June 2011 (ref. 23).

Discussion

The chloroform fumigation resulted in a significant increase in the amount of K_2SO_4-extractable C of all five soils (Fig. 1a), indicating that microbial biomass was destroyed by the chloroform fumigation and then extracted[22]. The amount of MBC estimated for the studied soils was within the range of that typically measured for forest surface soils in Japan[24].

Although microbial biomass was found in all of the soils, no increase in the amount of K_2SO_4-extractable ^{137}Cs was observed after the destruction of the microbial biomass by chloroform fumigation (Fig. 2), suggesting that a negligible amount of ^{137}Cs was retained in the microbial biomass in the soils. This result is consistent with a previous study showing that the amount of microbial biomass-bound ^{137}Cs was <2% of the total ^{137}Cs inventory in the top 3 cm of soil collected from undisturbed alpine meadows in Austria[20]. A similar result was also obtained for subtropical high mountain forest soils (0–10 cm depth) in Taiwan[21]. In addition, it has been reported that in O (O_f and O_h) horizons with substantial quantities of soil minerals, microbially retained ^{137}Cs contributes to only a minor portion of the total ^{137}Cs in spruce and beech stands in Germany[19].

However, laboratory studies have demonstrated the ability of microorganisms to accumulate ^{137}Cs from their external environment via active K^+ transport systems[13,16]. For highly organic soils, it was found that ^{137}Cs can be retained by and be strongly bound to soil constituents in the presence of microorganisms[25,26]. There have also been studies showing that, on an average, 13% (with a large variability between 1% and 56%) of the ^{137}Cs inventory can be stored within the microbial biomass in O horizons of forest ecosystems[18]. These studies suggest microbial involvement in the radiocesium retention process by "organic" soils.

Then why was the microbial retention of ^{137}Cs not observed in the soils evaluated in this study? The microbial population in soil is very diverse and varies significantly in different soils[27]. Given the fact that not all soil microorganisms are capable of ^{137}Cs uptake[16,17], it is possible that ^{137}Cs-accumulating microorganisms do not exist or are present in a much smaller quantity in the soils used for this study. Even though such microorganisms are dominant, it has been estimated that majority of soil microorganisms are in a dormant or even completely non-vital state under normal conditions[28]. In the present study, the soil sampling was conducted in March 2012, with a forest floor covered with >20 cm of snow. Soil temperature and moisture conditions can affect the amount and activity of soil microbial biomass[28]. Therefore, it is likely that the microbial activity under this condition was low by comparison with other periods of the year. Clearly, further investigations under different meteorological conditions (e.g., in a warmer season without snow cover and in a highly humid rainy season) are needed.

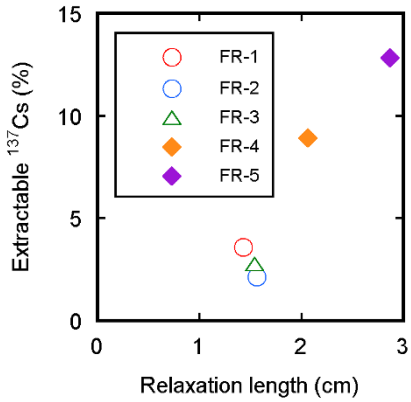

Figure 4 | Correlation between the K_2SO_4-extractable ^{137}Cs (as a % of the total ^{137}Cs in the soil) and relaxation length parameter. The relaxation length parameter is an index characterizing the ^{137}Cs penetration through the soil profile, and was determined by fitting an exponential function to the cumulative ^{137}Cs inventory down to a depth of 20 cm in June 2011 (ref. 23).

Furthermore, the possibility that the chloroform fumigation-extraction method causes an underestimation of the amount of microbially-bound ^{137}Cs in "mineral" soil samples cannot be completely ruled out. Chloroform fumigation does not render all of the microbial components extractable, and thus requires a proportion factor (k_c = 0.45, see Eq. (1)) to compensate for the unextracted fraction of microbial origin[22,29,30]. In addition, the ^{137}Cs liberated from the microbial biomass following chloroform fumigation might be rapidly re-adsorbed to some extent by other soil constituents[19], although the current results indicate that even if this process is possible, it must be highly selective for the liberated ^{137}Cs. The redistribution of the solubilized ^{137}Cs between the soil and solution likely depends on the clay content and mineralogy and the dominance of other cations in the soil exchange complex, and might differ from the redistribution of the solubilized C between the two. In addition, chloroform dissolved in the soil water itself might act as a mild flocculant for binding solubilized ^{137}Cs (as well as C) to the clay minerals[30]. In the present study, no increase in the amount of K_2SO_4-extractable ^{137}Cs was observed after the chloroform fumigation (Fig. 2). However, a significant decrease in the amount for two soils (FR-3 and FR-4) was observed, indicating that there existed ^{137}Cs isotopes that are easily exchangeable but become non-exchangeable as a result of chloroform fumigation; the underlying mechanisms for this behavior remain unclear.

The results of this study show that adsorption by abiotic components, rather than uptake by microorganisms, is important for the retention of potentially mobile ^{137}Cs in mineral soils. The K_2SO_4 extraction process released ~2.1%–12.8% of the total ^{137}Cs as easily exchangeable ions[9,18,19] from the non-fumigated soils (Fig. 3). In contrast, water extraction released a certain amount of C but almost a negligible amount of ^{137}Cs from the non-fumigated soil samples. The different ^{137}Cs extractability between the two extraction methods indicates that adsorption of ^{137}Cs on non-specific cation exchange sites provided by soil organic matter and the planar surfaces of clay minerals (and perhaps, to some extent, adsorption on specific sites at the edges of clay interlayers[31]) plays a role in the retention of mobile ^{137}Cs in mineral soils. The adsorption can decrease the ^{137}Cs content (including ^{137}Cs bound to water-soluble organic substances) in the solution phase, resulting in little transfer of ^{137}Cs along the mass flow of the soil water in mineral soils. It should be noted that, in agreement with well-established theory[8,9], a large fraction (>87%) of the ^{137}Cs appeared to be strongly (irreversibly) adsorbed onto specific sites of the clay minerals in the soils evaluated in this study.

The ^{137}Cs extractability differed largely between the soils. In particular, the two Andosol soils (FR-4 and FR-5), for which the clay and silt content, SOC content, and CEC were relatively high compared to the other soils, showed higher extractability (Fig. 3). The Fukushima-fallout ^{137}Cs penetrated deeper into the soil profile at these two forest sites than at the other three sites as of June 2011 (ref. 23), which was approximately three months after the accident (Fig. 4). The role of non-specific exchange sites can be compared with that of specific sites in the adsorption of ^{137}Cs in highly organic soils[32]. A positive relation between the soil-to-plant aggregated transfer factor (expressed in Bq kg^{-1} in the plant per Bq m^{-2} in the soil) and exchangeable fraction of ^{137}Cs in the soil has been reported[3]. These results support the suggestion that the adsorption process via ion exchange at non-specific sites exerts a strong influence on the mobility and bioavailability of ^{137}Cs in forest surface mineral soils.

Although the uptake and retention of ^{137}Cs by microorganisms in the forest surface soils was found to be very minor, microorganisms might contribute to the mobilization of the adsorbed ^{137}Cs (and consequently the transfer to plants) by decomposition of the soil organic matter into dissolved organic matter through production of extracellular enzymes[33]. A larger downward flux of ^{137}Cs accompanied by a larger flux of dissolved organic carbon has been observed in a forest soil[15]. Therefore, more studies, including methodological advances (e.g., monitoring of changes in ^{137}Cs behavior during microbial growth and metabolism under controlled laboratory conditions), are needed to fully understand the role of soil microorganisms in controlling the mobility and bioavailability of ^{137}Cs in natural biotic systems with interactions among the minerals, organic materials, and microorganisms.

Methods

Study site. The study was conducted at five forest sites within a 2 km × 2 km mesh area located in the southwestern part of the city of Fukushima (37.71°N, 140.36°E), ~70 km northwest of the Fukushima Daiichi NPP. The sites have been studied for inventory and vertical distribution of the radiocesium in soils since the accident in March of 2011 (ref. 23). Two of the five forest sites, FR-1 and FR-2, were dominated by broad-leaved trees (Japanese oak, hornbeam, etc.), with a promenade connected to a public park. FR-3 was a relatively sparse coniferous forest dominated by Japanese red pine. FR-4 and FR-5 were dense coniferous forests dominated by Japanese cedar, located at different elevations along a trail over a small mountain. The mean annual temperature and precipitation for the last ten years at the Fukushima meteorological station are 13.3°C and 1234 mm, respectively (Japan Meteorological Agency).

The city of Fukushima is located on an extended plain to the north and south between the Abukuma granite plateau and Ou Mountains. Rocks in this region consist mainly of gravel-rich unconsolidated and consolidated sediments, granite, andesite, and tuff[34]. The soils studied have been classified as Fluvisols (FR-1, FR-2, and FR-3) and Andosols (FR-4 and FR-5) using the classification of the Food and Agriculture Organization[34]. Depth profiles for the physicochemical properties (bulk density, particle size distribution, pH, cation exchange capacity (CEC), exchangeable base cations, and base saturation) of the soils have also been investigated[23], and values for the surface 0–3 cm soil layers were estimated by the averaged value of the 0–1 cm and 1–3 cm layers (see horizontal axes in Fig. 3d–h). The soils generally had a low bulk density (0.40–0.84 g cm^{-3}) and acidic pH (4.6–5.6). The clay-sized (<0.002 mm) particle content was <16.4%, and lower at the FR-1 site than at the others. The CEC ranged from 20 to 41 cmol kg^{-1}, and the exchangeable Ca and K content was 1.4–12.4 and 0.3–0.6 cmol kg^{-1}, respectively. The base saturation was higher (>40%) at three sites FR-1, FR-2, and FR-4 than at the other two sites FR-3 and FR-5 (<15%). Note that the base saturation was calculated by dividing the sum of the exchangeable base cations (Ca^{2+}, Mg^{2+}, K^+, and Na^+) by the CEC and multiplying by 100.

Soil sampling. Soil sampling was conducted on March 7 and 8, 2012, about one year after the Fukushima Daiichi NPP accident. At the time of sampling, the forest floor was covered with >20 cm of snow at all of the sites. Therefore, the top soil (0–3 cm) samples were collected by grab sampling after removal of the snow cover and litter layer (O horizon) from the forest floor by hand. The soil samples were immediately placed in containers, cooled with dry ice, brought back to the laboratory, and stored at 6°C for 12–18 days until initiation of the fumigation treatment. The components of the litter layer generally reflect the standing vegetation. The density of litter layer was higher (1.3–1.7 kg-dry mass m^{-2}) in coniferous-dominated forests (FR-3 to FR-5) than in broadleaf-dominated forests (0.6–0.7 kg-dry mass m^{-2}; FR-1 and FR-2).

Chloroform fumigation and extraction. The soil samples were sieved through a 2-mm mesh while still moist, and aliquots of the <2-mm samples were dried at 50°C for determination of the dry weight (or, water content of the samples). A portion of each of the <2-mm moist soils corresponding to 10 g of dry weight was then fumigated with ethanol-free chloroform for 24 h at 25°C in a sealed desiccator[18,19,22]. After fumigant removal, the fumigated soils (number of replicates N = 3) were extracted with 100 mL of 0.5M K_2SO_4 at room temperature with 30-min of shaking, and then filtered using pre-combusted glass microfiber filters (Whatman GF/F). The non-fumigated soils (N = 3) were similarly extracted at the time fumigation commenced.

For the non-fumigated soil samples from the FR-3, FR-4, and FR-5 sites, we also employed an extraction with 100 mL of deionized water for comparison.

Organic C analysis and MBC estimation. The organic C content (gC kg^{-1}-dry soil) of ground <2-mm soil samples was measured with a total organic carbon analyzer (TOC-L CPH with a solid sample module: SSM-5000A, Shimadzu, Japan). Aliquots of the K_2SO_4 and water extracts from the fumigated and non-fumigated soil samples were also analyzed for organic C content (gC L^{-1}) using the analyzer (TOC-L CPH), and then the C content was expressed as the amount of C extracted per unit dry weight of soil (i.e., gC kg^{-1}-dry soil).

The soil microbial biomass C (MBC; gC kg^{-1}-dry soil) was calculated as follows:

$$\mathrm{MBC} = [(\text{C extracted from fumigated soil}) - (\text{C extracted from non-fumigated soil})]/k_c, \quad (1)$$

where k_c = 0.45 (ref. 29), the ratio of the C extracted from the cells of microbial biomass killed by fumigation to the total microbial biomass C.

Radiocesium analysis. The activity concentrations of ^{137}Cs and ^{134}Cs in the soil (Bq kg^{-1}-dry soil) and liquid (Bq L^{-1}) samples were determined using gamma-ray spectrometry, according to an officially recommended method in Japan[35]. Samples

(~50–200 g and ~70 mL for the soil and liquid samples, respectively) were sealed in plastic tubes and analyzed for ^{137}Cs and ^{134}Cs using high-purity coaxial germanium detectors at the Nuclear Fuel Cycle Engineering Labs of the Japan Atomic Energy Agency. The detectors were calibrated with standard gamma sources (each with a relative error of ~5%) prepared in the same shape as the measurement samples. The measurement times were 3,600 s and 80,000 s for the soil and liquid samples, respectively. The activities obtained were corrected for radioactive decay to the sampling date (March 7 or 8, 2012). The ^{137}Cs detection limits for the soil and liquid samples were around 0.059 Bq g^{-1} and 0.0026 Bq mL^{-1}, respectively.

The measured ^{137}Cs and ^{134}Cs showed a similar pattern of extractability for all of the investigated soils, and therefore, only the ^{137}Cs results were discussed in this study for simplicity. The ratio of ^{137}Cs to ^{134}Cs concentrations was generally 1.24–1.38 for all of the soil and liquid samples (see Table S1 in Supplementary Information).

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Author contributions

J.K., K.M. and M.A.A. designed the study; J.K., M.A.A. and T.M. participated in the field work; J.K. and K.M. performed the laboratory experiments; H.F. and M.N. did the radioactivity measurements; J.K. wrote the manuscript. All of the authors contributed extensively to discussions about this study and reviewed the manuscript.

Additional information

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