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Continuous nitrogen fertilization retards the vertical migration of Fukushima nuclear accident-derived cesium-137 in apple orchard soil

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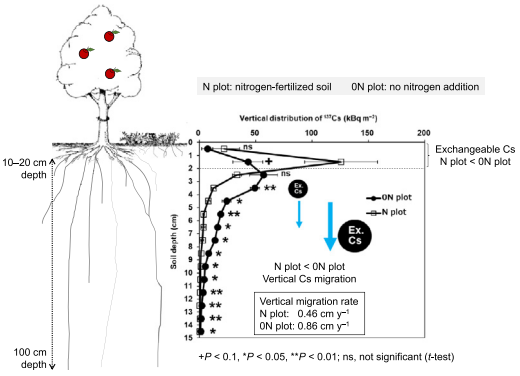
HIGHLIGHTS

- Orchard topsoil retained 42–83% of ^{137}Cs from the Fukushima accident after 5 years.
- Long-term N fertilization reduced the soil ^{137}Cs migration rate by about one-half.
- N fertilization resulted in less Cs with mobile exchangeable form at the surface.
- Reduced ^{137}Cs migration may be due to higher fixation at the surface.
- N fertilization may reduce radiation risk via tree roots by slowing ^{137}Cs migration.

GRAPHICAL ABSTRACT



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ABSTRACT

We monitored the levels of cesium-137 (^{137}Cs) in the soils of five orchards for six years following the Fukushima Daiichi Nuclear Power Plant accident on 11 March 2011 and found that the vertical distribution of accident-derived ^{137}Cs varied significantly among the orchards with varying land-use and fertilizer management. Based on these results, this study evaluated how nitrogen (N) fertilizer management may have affected the vertical migration of ^{137}Cs in the orchard soils. We selected an experimental orchard producing 'Jonathan' apples, where a long-term N-fertilizer trial has continued since 1973, with an N-fertilized plot (N plot; N added at 20 g m^{-2} y^{-1}) and a non-fertilized plot (0 N plot). Five years after the accident, the vertical migration of accident-derived ^{137}Cs was significantly lower in the N plot (2.3 cm) than in the 0 N plot (4.3 cm), suggesting greater ^{137}Cs retention in the surface of the N plot. Application of a cesium bromide (^{133}CsBr) tracer suggested that the retarded vertical migration of ^{137}Cs in the N plot may be related to significantly lower amounts of exchangeable ^{133}Cs and significantly higher proportions of non-exchangeable ^{133}Cs in the upper 2 cm. We did not find any evidence of the aboveground plants contributing to more ^{133}Cs retention in the N plot. However, greater ^{137}Cs retention in the surface (0–2 cm) of the N plot may be due to more dissolved organic carbon (DOC), and high DOC may have

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Soil physicochemical properties

contributed to deeper ^{137}Cs migration in both the plots. Our results suggest that continuous N fertilizer application significantly retarded the migration of ^{137}Cs by approximately one-half and resulted in less ^{137}Cs reaching the mobile exchangeable form in the deeper root-zone layers.

1. Introduction

The accident at the Fukushima Daiichi Nuclear Power Plant (NPP) on 11 March 2011 caused substantial radioactive contamination through radionuclide deposition across agricultural areas, including the orchards in Fukushima Prefecture, Japan (Nuclear Regulation Authority, 2018), which are major areas of deciduous fruit production (Ministry of Agriculture, Forestry and Fisheries, 2018). Cesium-137 (^{137}Cs) is a radionuclide with a long half-life (30.1 y), and it is a serious concern in orchards because of the decades-long radiation risk in the fruits of perennial trees. After the 1986 Chernobyl NPP accident, blueberries in the Czech Republic contained relatively high ^{137}Cs levels, and the concentrations within the fruits correlated with soil ^{137}Cs concentrations ($r = 0.93$) long after the accident (Cervinkova and Poschl, 2014). Therefore, the risks of ^{137}Cs transfer to fruit-bearing plants through the roots in contaminated soils require prolonged monitoring. This is particularly relevant in orchards, where the surface soil has not been plowed or removed, thereby allowing ^{137}Cs migration into the deeper soil layers (Sato et al., 2019a), and where there has been no advanced potassium (K) fertilization in low-K soils to reduce radiocesium accumulation in the fruits, such as blueberry (Iwabuchi, 2014), lingonberry, bilberry (Rosén et al., 2011), and coconut (Robison et al., 2009).

One method to assess the risk of soil-to-plant ^{137}Cs transfer in orchards is to predict how the deposited ^{137}Cs will migrate through the soil profile to the tree root-zone over time. Prior studies have reported the vertical distribution of radiocesium from the Fukushima nuclear accident in orchards (Koarashi et al., 2012; Ohno et al., 2012; Sato et al., 2019b); >90% of the radiocesium was within 4 cm of the soil surface on 23–24 April 2011 (Ohno et al., 2012) and within 5 cm of the soil surface on 18–20 June 2011 (Koarashi et al., 2012). Sato et al. (2019b) monitored the vertical distribution of ^{137}Cs in five orchards for six years after the accident and found that most of the deposited ^{137}Cs (84–94%) was retained in 3 cm of topsoil in 2011. By 2017, the ^{137}Cs had migrated to the deeper layers (41–75% in the upper 3 cm). There were also significant differences among the orchards with different land-use and fertilizer management four years after the Fukushima Daiichi NPP accident (Sato et al., 2019b). However, the factors underlying these orchard-specific differences have not been investigated to date.

The application of fertilizers (nitrogen (N), phosphorus (P), and K) to orchards is a customary agricultural practice to improve fruit production and tree nutrition in Fukushima Prefecture, Japan (Fukushima Prefecture, 2006), and elsewhere worldwide (e.g., Stiles and Reid, 1991). Nitrogen fertilizer is particularly important, because, after carbon (C), nitrogen is the most- required element by plants (Hawkesford et al., 2012). Pro-longed surface application of ammonium-based fertilizers alters fruit-tree nutrition and the properties of orchard surface soils, including increased total C and N contents, decreased pH (Matsuoka et al., 2018; Raese et al., 2007), and decreased exchangeable base cations (magnesium (Mg), K, and calcium (Ca)) (Matsuoka et al., 2018). The marked increases in orchardgrass (*Dactylis glomerata*) growth (blade height and width) and the N content of the aboveground parts of the undergrowth plants due to ammoniacal N fertilization in apple orchards (Agriculture, Forestry and Fisheries Research Council and Fruit Tree Research Center, Fukushima Agricultural Technology Center, 1997; Raese et al., 2007) also increase the supply of plant residues to the soils fertilized with N. Consequently, these fertilizer-induced changes may alter the retention and vertical migration of accident-derived ^{137}Cs in orchard soils.

The retention, mobility, and vertical migration of ^{137}Cs in soils are affected by the clay-mineral content, such as the soil type and texture (Rosén et al., 1999), fine-sized particle content (Koarashi et al., 2012; Matsunaga et al., 2013), and physicochemical characteristics, such as the organic matter content (Matsunaga et al., 2013; Sato et al., 2019b; Takenaka et al., 1998), pH (Matsunaga et al., 2013), and cation exchange capacity (CEC) (Matsunaga et al., 2013). The soil physicochemical characteristics may mask the role of clay minerals, as prior studies reported lower effects on radiocesium mobility (Matsunaga et al., 2013; Sato et al., 2019b) and no influential role of the clay-mineral content on ^{137}Cs migration through the soil profile (Shand et al., 2013). Contrastingly, Rosén et al. (1999) suggested that the soil type and texture are related to the rate of vertical ^{137}Cs migration; the mean migration rate was the lowest in clayey soils and highest in organic soils and podzol.

To date, no studies have investigated the effects of fertilization on the vertical migration of accident-derived ^{137}Cs in orchard soils, but a few studies have reported the effects in watersheds and pasture soils. McHenry and Ritchie (1977) found that variability in the ^{137}Cs content of surface soils from 12 arid watersheds could be explained by the soil N content, but Squire and Middleton (1966) showed that ^{137}Cs migration in pasture soils fertilized annually with N, P, K, and Ca for six years was not affected by soil N. A better understanding of how N fertilizer influences the vertical migration of ^{137}Cs in contaminated orchard soils and how long it takes deposited ^{137}Cs to migrate to the main root-zone of fruit trees is essential for well-informed risk management strategies in agriculture. Contaminated surface soils have been removed, and radiocesium-free fruit trees have been replanted in contaminated soils to examine various decontamination strategies in Fukushima Prefecture after the Fukushima Daiichi NPP accident (e.g., Horii et al., 2020; Sato et al., 2019a).

The first goal of this study was to investigate the effects of N fertilization on the vertical migration of deposited ^{137}Cs in orchard soils. To this end, we chose an experimental orchard that was affected by the Fukushima nuclear accident, where ammoniacal fertilizer has been applied annually to the soil surface since 1973. The second goal of this study was to investigate the underlying factors that influence the vertical migration of ^{137}Cs in response to long-term N fertilization. For this purpose, we applied a cesium bromide (^{133}CsBr) tracer solution six years after the accident; the properties of stable ^{133}Cs in nature aid predictions of radioactive ^{137}Cs (Yoshida et al., 2004).

2. Materials and methods

2.1. Long-term N fertilization in an experimental orchard

Our study took place at an experimental orchard (37°48′47.7″N, 140°26′40.5″E) located approximately 60 to 65 km northwest of the Fukushima Daiichi NPP. The experimental orchard belongs to the Fruit Tree Research Center of the Agricultural Technology Center in Fukushima Prefecture, Japan. The soil in the orchard is classified as Humic Cambisol and fine-textured brown forest soil, Kasayama series (Agriculture, Forestry and Fisheries Research Council and Fruit Tree Research Center, Fukushima Agricultural Technology Center, 1997).

The long-term N fertilizer experiment started in 1973 and has continued through 2017. The orchard was planted with six trees bearing 'Jonathan' apples (*Malus pumila* Mill.) on *Malus prunifolia* Borkhausen rootstock; the trees were 54 years old in 2017. The experiment included two plots—one had never been fertilized with N ("0 N plot"; N added at

0 g m^{-2}), while the other had been fertilized with N ("N plot"; N added at up to 20 g m^{-2} annually as ammonium nitrate) (see Matsuoka et al., 2018, 2019 for details). Each plot was 17.5 m × 27.0 m and had three replicate sampling areas (Fig. 1). Phosphorus, K, Ca, and Mg were applied as magnesium multi-phosphate and potassium sulfate to the soil surface of both plots in mid-to-late March every year.

After the Fukushima Daiichi NPP accident on 11 March 2011, the experimental orchard suffered radioactive contamination—radionuclides cesium-134 (^{134}Cs) and ^{137}Cs were deposited at 120 to 160 kBq m^{-2} and 200 to 275 kBq m^{-2}, respectively (measured on 3 November 2012) (Sanderson et al., 2013). None of the topsoil in the orchard has been cultivated since the accident; therefore, any remaining ^{137}Cs is derived from the Fukushima accident. The undergrowth plants on the soil surface in the orchard were cut five to seven times a year with lawnmowers, and the trimmings were left on the ground.

Soils collected from the experimental orchard in 2014 had the following properties at 0–15 cm depth in the non-fertilized (0 N plot) and N-fertilized plots (N plot), respectively (see Matsuoka et al., 2018, 2019 for details): total N, 1.43–3.09 and 1.29–4.34 g kg^{-1}; organic N, 1.34–2.98 and 1.21–4.22 g kg^{-1}; total C, 18.5–36.2 and 16.0–48.9 g kg^{-1}; CEC, 16.3–19.8 and 15.5–20.8 $cmol_c$ kg^{-1}; exchangeable Ca, 2.40–2.86 and 0.90–1.43 $cmol_c$ kg^{-1}; exchangeable Mg, 1.50–2.52 and 0.52–1.44 $cmol_c$ kg^{-1}; exchangeable K, 1.50–2.18 and 0.68–1.25 $cmol_c$ kg^{-1}; exchangeable Al, 1.12–1.21 and 4.00–-6.51 $cmol_c$ kg^{-1}; and sand content, 57.4–60.0% and 57.2–61.7%. Long-term N fertilization significantly increased total N, organic N, and total C contents at the surface (0–5 cm), significantly decreased the pH (H_2O and potassium chloride, KCl) at the surface (0–15 cm), significantly decreased CEC at the surface (10–15 cm), and significantly decreased the content of exchangeable Ca, Mg, and K and increased that of exchangeable Al at the surface (0–15 cm). The absence of a significant difference in the sand contents of the 0 N and N plots suggests that the textural properties were similar. The dominant phyllosilicates in the clay fraction of the soils collected in 2013 were kaolin minerals, mica, vermiculite, and chlorite-vermiculite intergrade (data not shown).

2.2. Soil sampling and analyses of the vertical ^{137}Cs distribution and soil properties affecting ^{137}Cs migration

On 25–26 May 2016, five years after the Fukushima Daiichi NPP accident, we examined the vertical distribution of ^{137}Cs in the experimental orchard soil. We used a scraper plate to collect undisturbed soil at one sampling point per tree (three sampling points per plot) from an area one-third of the distance inward from the edge of the canopy

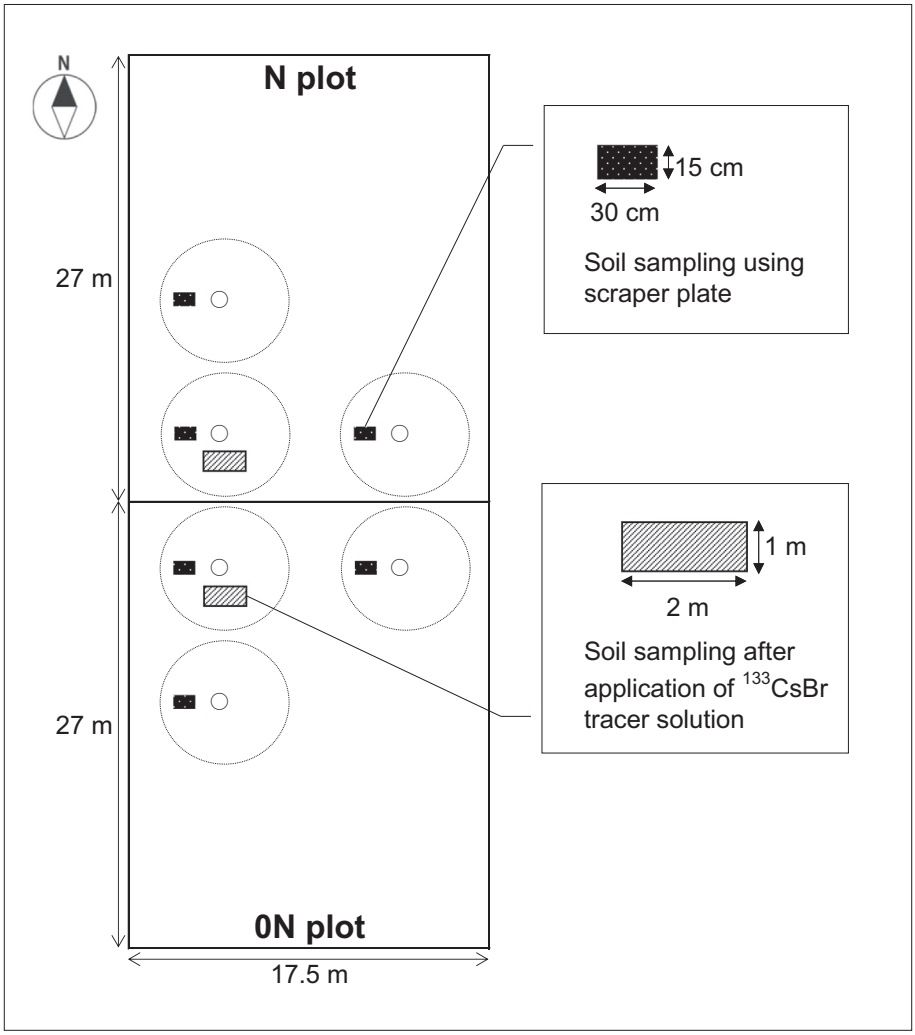

Fig. 1. Schematic of the field experiment showing the layout of the 0 N and N plots. Dark rectangles indicate the soil sampling locations with a scraper plate in 2016 at 0–15 cm soil depth. Light shaded rectangles indicate sampling areas (1 m × 2 m per plot) for the undergrowth plants and soils at 0–50 cm depth after application of a ^{133}CsBr tracer solution in 2017. The large and small circles in each plot show the apple tree canopy and the tree trunk, respectively.

(Fig. 1). The scraper plate has a metal frame with a 450 cm^2 sampling area (15 cm × 30 cm) that is placed on the ground, and an adjustable metal plate that can scrape or remove soil at fixed depth increments within the frame (TSUKAHARA-SS Corporation, Gifu). The undergrowth plants on the soil surface were cut away with scissors, and the topsoil was scraped and collected at 1 cm depth intervals. Deeper soil samples were similarly collected, yielding vertically-sectioned soil samples every cm from 0 to 15 cm depth. To avoid soil contamination, surface soil that fell from the wall of the sampling hole was not included in deeper samples. The soil samples were air-dried and passed through a 2 mm sieve.

The ^{137}Cs concentration of the soil samples was measured using germanium detectors (Ortec GEM 20P4-70 and Ortec GEM 204-70, Seiko EG&G, Tokyo, Japan; and GC4020-7500SL, Canberra, USA). The gamma-ray peak for ^{137}Cs is 661.7 keV, and the counting times ranged from approximately 2000 s to 200,000 s, depending on the sample concentration. A standard source (MX033U8PP; Japan Radioisotope Association, Tokyo, Japan) was used for the efficiency calibration. The counting errors for the ^{137}Cs measurements were 0.2% to 4.9% (dry wt. basis). The activities were corrected for radioactive decay to the sampling date, and the values were expressed in units of activity per unit dry weight (Bq kg^{-1} dry wt.).

To measure the chemical properties of the soil, 6.00 g of the soil sample was extracted for 1 h with distilled water at a soil-to-solution ratio of 1:5, and the pH (H_2O) and electrical conductivity (EC; mS m^{-1}) were measured with a glass electrode. After the measurements, the suspension was centrifuged, the supernatant was filtered with a 0.22 μm membrane (Millipore Express Plus Membrane, Merck KGaA, Darmstadt, Germany), and the dissolved organic carbon (DOC) concentration was measured by a TOC instrument (TOC-L CPH J100, Shimadzu Corporation, Kyoto, Japan).

Soil bulk density (g cm^{-3}) was determined by dividing the soil dry weight in each layer by its volume, calculated from the diameters of the scraper plate frame and the thickness of each layer. We measured the hardness of the top 15 cm of soil in both plots on 23 March 2018 at a point one-third of the distance inward from the edge of the canopy with a Hasegawa-type penetrometer (portable "H-100" type cone penetrometer, Daitou Techno Green, Inc., Tokyo, Japan). Soil hardness (cm/drop) was measured at two sampling points per tree (six sampling points per plot), calculated as the number of drops required for a 2 kg cone weight to pass through the top 15 cm of soil.

2.3. Application of $^{133}CsBr$ tracer solution to the experimental orchard

To investigate the effects of long-term N fertilization on the vertical distribution of the Fukushima-derived ^{137}Cs, we applied a $^{133}CsBr$ tracer solution as a proxy for radioactive ^{137}Cs in the soil of the experimental orchard. The bromide (Br^-) ion also serves as a tracer because it is not strongly adsorbed or fixed in the soil solid phase and easily leaches to the deeper layers with rainwater. Br^- has been used under laboratory and field conditions as an alternative to conventional estimations of nitrate (NO_3^-) leaching loss and NO_3^- movement in soils (Kessavalou et al., 1996; Smith and Davis, 1974).

On 24 March 2017, six years after the Fukushima Daiichi NPP accident, we dissolved 50 g of $^{133}CsBr$ (Fujifilm Wako Pure Chemical Corporation) in distilled water and sprayed the mixture uniformly over the soil surface of a 1 m × 2 m test area under the canopy on the south side of a tree (Fig. 1). The $^{133}CsBr$ solution was applied to one test area per plot at a rate of approximately 500 g m^{-2}, yielding ^{133}Cs at 15.6 g m^{-2} and Br at 9.39 g m^{-2}. The undergrowth plants on the soil surface in the test areas were cut away with scissors before applying the spray. To ensure complete usage of the $^{133}CsBr$ solution, the spray bottle was washed two times with distilled water, and the wash solution was sprayed uniformly over the soil surface.

2.4. Sampling and analyses of the vertical ^{133}Cs and Br distributions in the undergrowth plants and soil

Three core samples including the undergrowth plants with the aboveground parts and roots and soil were collected from each test area (1 m × 2 m) in both plots on 24 March 2017 (day 0, after the application of the $^{133}CsBr$ tracer solution), 6 April (day 13), 21 April (day 28), 13 June (day 81), 8 September (day 168), and 24 November (day 245). The core samples were collected at 0–50 cm depth using a stainless-steel core sampler (40 mm inside diameter). To prevent soil compaction during sampling, the soil was sampled separately at 0–10 cm and 10–50 cm. The inner surface of the core sampler was wiped clean between samples to avoid cross-contamination. The core sample was divided into six sections for ^{133}Cs and nine sections for Br using a spatula, such that for each core there were samples from depths 0–2, 2–4, 4–6, 6–8, 8–10, and 10–15 cm for ^{133}Cs and Br, and 15–20, 20–30, and 30–50 cm for only Br. All of the samples were air-dried, and the aboveground parts of the undergrowth plants were collected by separating the soil from the top of the core (0–2 cm). The soil was passed through a 2 mm sieve.

To investigate the time-dependent changes in ^{133}Cs retained in the undergrowth plants, the aboveground parts of the plants from each core sample were digested with 40 or 80 mL of hydrochloric acid (HCl; 1 mol L^{-1}) at 50 °C for 20 h in an incubator according to the modified method of Schachtschabel and Heinemann (1974). After centrifugation at 760*g* for 10 min, the extract was passed through filter paper (Advantec no. 2) into a clean 50 mL centrifuge tube, and the concentration of ^{133}Cs was measured using an atomic absorption spectrophotometer (AA-7000, Shimadzu Corporation, Kyoto, Japan).

To determine the exchangeable ^{133}Cs concentrations, each soil sample was extracted with ammonium acetate (CH_3COONH_4; 1 mol L^{-1}) at a soil-to-solution ratio of 1:20 (2.00 g:40 mL) in a 50 mL centrifuge tube. The soil-extract mixture was shaken for 1 h and centrifuged at 760*g* for 10 min, and then the supernatant was passed through filter paper (Advantec no. 2) into a clean 50 mL centrifuge tube. The concentration of ^{133}Cs in the extract was measured using an atomic absorption spectrophotometer (AA-7000, Shimadzu Corporation).

To measure the non-exchangeable ^{133}Cs concentrations, 2.00 g of soil was extracted with HCl (1 mol L^{-1}) at a soil-to-solution ratio of 1:20 (2.00 g:40 mL) at 50 °C for 20 h in an incubator according to the modified method of Schachtschabel and Heinemann (1974). After the extraction, the soil-extract mixture was centrifuged at 760*g* for 10 min, and the supernatant was passed through filter paper (Advantec no. 2) into a clean 50 mL centrifuge tube. An additional 40 mL of HCl (1 mol L^{-1}) was added to the residual soil in the tube, and the tube was extracted for 20 h in the incubator and centrifuged again. The supernatant was passed through filter paper and each filtrate was brought to a final volume of 50 mL with 1 mol L^{-1} HCl. The concentrations of ^{133}Cs in the first and second extracts were separately measured using an atomic absorption spectrophotometer (AA-7000, Shimadzu Corporation).

Bromide in the soil was extracted with distilled water at a soil-to-solution ratio of 1:5 (2.00 g:10 mL) in a 50 mL centrifuge tube. The mixture was shaken for 1 h and centrifuged at 760*g* for 10 min, and then the supernatant was filtered through a 0.2 μm syringe filter (Minisart RC, Sartorius, Goettingen, Germany) into a clean 15 mL centrifuge tube. The concentration of Br in the extract was determined using a personal ion analyzer (PIA-1000, Shimadzu Corporation).

2.5. Data analysis

The amount of ^{133}Cs retained in the aboveground parts of the undergrowth plants (P, mg m^{-2}) was calculated as follows:

$$P = c \cdot b, \quad (1)$$

where c is the concentration of ^{133}Cs (mg kg^{-1} air-dried weight) in the plant matter of each core sample, and b is the weight of the plants (kg m^{-2}) in each core sample.

The amount (S) of ^{137}Cs (Bq m^{-2}), ^{133}Cs (mg m^{-2}), Br (mg m^{-2}), or DOC (g m^{-2}) in a given soil layer was calculated as follows:

$$S = c \cdot b \cdot d, \quad (2)$$

where c is the concentration of ^{137}Cs (Bq kg^{-1} dry wt.), ^{133}Cs (mg kg^{-1} dry wt.), Br (mg kg^{-1} dry wt.), or DOC (mg kg^{-1}) in the layer, b is the soil bulk density (kg m^{-3}) of the layer, and d is the thickness (m) of the layer.

The average migration distance of ^{137}Cs (M_d, cm) in the soil from 2011 to 2016 was calculated as the center (centroid) of the vertical distribution of the ^{137}Cs amount (Bq m^{-2}) in the 0–15 cm layers of the soil in 2016, using a modified equation from Shiozawa et al. (2011) and Shiozawa (2013):

$$M_d = \sum x_i c_i \Delta x_i b_i \left(\sum c_i \Delta x_i b_i\right)^{-1}, \quad (3)$$

where the subscript i refers to the soil layer, x_i represents the mean depth of the ith soil layer, c_i is the concentration of ^{137}Cs in the ith soil layer (Bq kg^{-1} dry wt.), Δx_i is the thickness (m) of the ith soil layer, and b_i is the soil bulk density (kg m^{-3}) of the ith layer.

Data from each sample from the 0 N and N plots were compared using t-tests to assess the effects of long-term N fertilizer application. The soil ^{137}Cs data from the 0 N and N plots were analyzed using Pearson's correlation coefficient (r) to evaluate the significance of the soil properties in each plot. Data from field experiments tend to vary, and therefore, we also considered $P < 0.1$ with $P < 0.01$ and $P < 0.05$ for indications of significant differences between the two plots. All statistical analyses were performed in Microsoft Excel 2016 and Excel Toukei (BellCurve for Excel) (Social Survey Research Information, Tokyo, Japan).

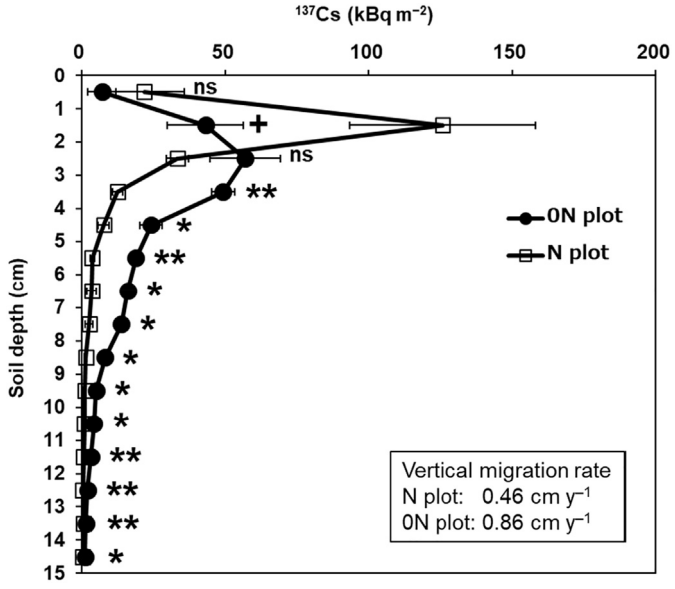

Fig. 2. Vertical distribution of ^{137}Cs (kBq m^{-2}) in the 0–15 cm soil layers in a long-term N fertilization experimental orchard five years after the Fukushima nuclear accident. Differences between the 0 N and N plots in the corresponding soil layer are significant at $+P < 0.1$, $^{*}P < 0.05$, $^{**}P < 0.01$; ns, not significant (t-test). Error bars represent standard errors of the means ($n = 3$).

3. Results

3.1. Effects of long-term N fertilizer application on the vertical distribution of soil ^{137}Cs

We determined the vertical distribution of ^{137}Cs (kBq m^{-2}) in the experimental orchard with and without continual surface application of ammoniacal fertilizer since 1973 (Fig. 2). Five years after the Fukushima Daiichi NPP accident, the highest levels of ^{137}Cs were found at the surface (1–3 cm) and the amount decreased with depth through the soil profile in both plots. Of the total amount of ^{137}Cs, 42% (0 N plot) and 83% (N plot) remained in the upper 3 cm of the soil at the time of sampling.

Significantly higher amounts of ^{137}Cs in the top 1–2 cm and significantly lower amounts at 3–15 cm indicates that the long-term application of N fertilizer retarded the vertical migration of ^{137}Cs.

The average accident-derived ^{137}Cs migration distance significantly differed between the two plots ($P < 0.01$, $n = 3$), moving 2.3 ± 0.17 cm in the N plot and 4.3 ± 0.33 cm in the 0 N plot (at 0–15 cm depth from 2011 to 2016) (data not shown). This corresponds to 0.46 cm y^{-1} in the N plot and 0.86 cm y^{-1} in the 0 N plot (Fig. 2). The continuous application of N fertilizer reduced the migration rate of ^{137}Cs by approximately one-half.

3.2. Time-dependent distribution of the ^{133}CsBr tracer and the potential N fertilizer-related processes limiting vertical ^{137}Cs migration

We used the ^{133}CsBr tracer to investigate the factors underlying greater ^{137}Cs retention in the surface layer of the N plot after the Fukushima Daiichi NPP accident. The distribution of the ^{133}Cs tracer was evaluated as four fractions: ^{133}Cs retained in the aboveground parts of the undergrowth plants in the 0–2 cm layer, exchangeable ^{133}Cs in the 0–2 cm layer, non-exchangeable ^{133}Cs in the 0–2 cm layer, and exchangeable ^{133}Cs in the 2–4 cm layer (Fig. 3). Following the tracer application, the percentages of ^{133}Cs in the aboveground parts of the undergrowth plants gradually decreased with time in both plots, from 23% to 1% in the 0 N plot and from 29% to 0% in the N plot. However, there were no significant differences in the amount of ^{133}Cs in the undergrowth plants between plots, except for day 245 ($P < 0.1$) (Fig. 4). The concomitant increase in exchangeable form of ^{133}Cs at 0–2 cm in both plots implies that ^{133}Cs retained in the aboveground parts of the undergrowth plants leached into the soil over time.

Most ^{133}Cs was distributed in the exchangeable form in the 0–2 cm layer and some exchangeable ^{133}Cs migrated to the 2–4 cm layer in both plots as time progressed (from 0% to 36% in the 0 N plot and from 0% to 33% in the N plot) (Fig. 3). The percentage of non-exchangeable ^{133}Cs was relatively constant in the 0–2 cm layer during the 245-day experimental period in both plots (9–16% in the 0 N plot and 10–32% in the N plot). This suggests that non-exchangeable ^{133}Cs is more strongly bound to the soils than exchangeable ^{133}Cs. The largest difference between the two plots was in the exchangeable ^{133}Cs in the 0–2 cm layer; the percentage was somewhat lower in the N plot (15–67%) than in the 0 N plot (32–82%). Overall, total ^{133}Cs was significantly lower in the N plot on days 28 and 168, but this trend was absent on the other sampling days. In addition, total ^{133}Cs in the N plot was below 100% on most sampling days, indicating a greater unknown ^{133}Cs fraction.

We also investigated temporal changes in the amount of exchangeable ^{133}Cs in the 0–2 cm layer during the tracer experiment (Fig. 4). The N fertilizer resulted in significantly lower amounts of exchangeable ^{133}Cs in the 0–2 cm layer on days 28, 81, 168, and 245. Some of the exchangeable ^{133}Cs gradually migrated to the next layer (2–4 cm), but there were no significant between-plot differences in the 2–4 cm layer. There were also no significant differences in the amount of non-exchangeable ^{133}Cs between the 0 N and N plots in the 0–2 cm layer. To compare non-exchangeable ^{133}Cs in the 0 N and N plots, we

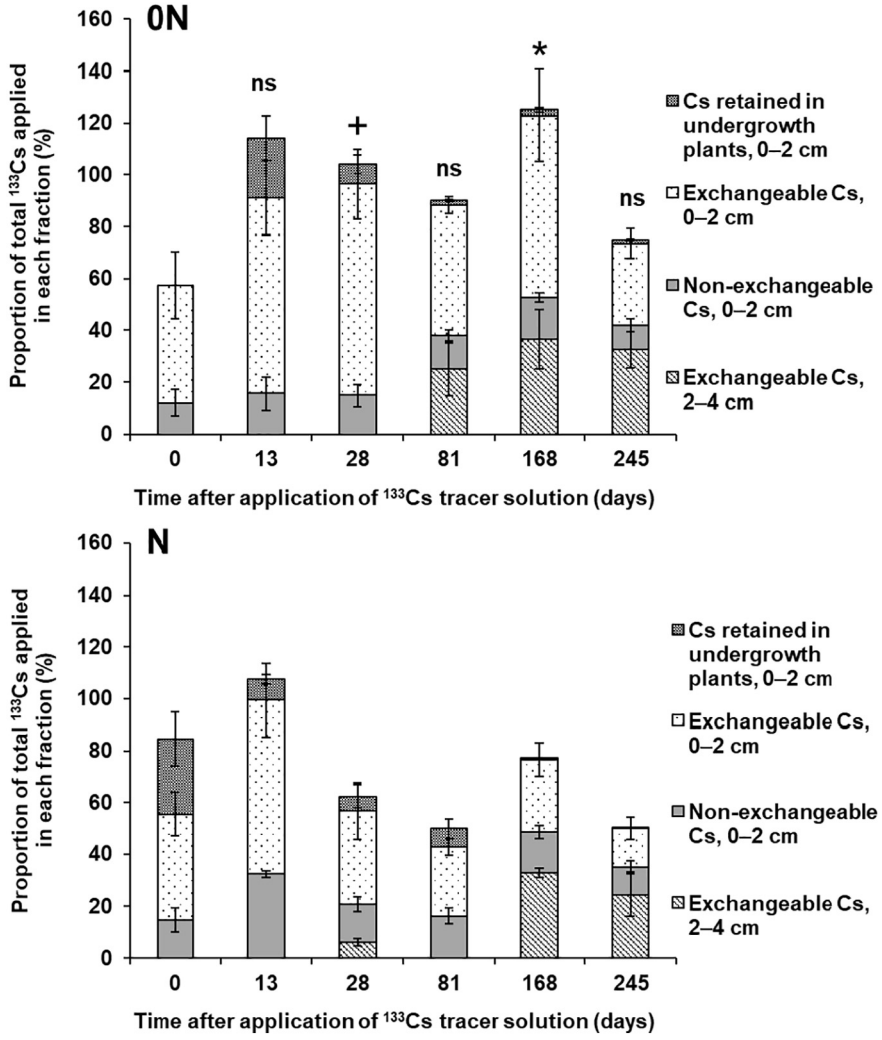

Fig. 3. Time-dependent changes in ^{133}Cs tracer in the four fractions (%) of the 0–2 and 2–4 cm soil layers following the application of a ^{133}CsBr tracer solution. There was no undergrowth plant sample in the 0 N plot on day 0. Significant differences in the total amounts of ^{133}Cs in all four fractions of the 0 N and N plots are indicated in the upper figure: $+P < 0.1$, $^{*}P < 0.05$; ns, not significant (t-test). Error bars represent standard errors of the means ($n = 3$).

calculated the percent ratio of non-exchangeable to exchangeable ^{133}Cs in the 0–2 cm layer (Fig. 5) and found that the ratio was significantly higher in the N plot throughout the experimental period. This result indicates a higher contribution of non-exchangeable form in the 0–2 cm layer of the N plot, which may be due to the higher ^{133}Cs fixation that retards the vertical migration of accident-derived ^{137}Cs (Fig. 2).

Less vertical migration of ^{137}Cs in the soil with long-term N fertilization may also be due to slower rainwater infiltration into the N plot soil profile, compared to the 0 N plot. This is supported by slower Br migration in the tracer solution in the N plot, where significantly more Br remained in the top 2–10 cm on day 81 and in the 30–50 cm on day 168 (Fig. 6). The N plot also had a greater soil hardness value at 0–15 cm (1.1 cm/drop), compared to the 0 N plot (1.7 cm/drop) ($P < 0.05$, $n = 6$) (Fig. 7; the representative data for each plot). To identify the factors affecting the vertical migration of ^{137}Cs, we focused on the interactions between soil ^{137}Cs and the four soil properties in the 0–2 and 2–15 cm layers (Table 1) (see Table A.1 for the raw data). DOC (g m^{-2}) was positively correlated with ^{137}Cs (kBq m^{-2}) at the surface (0–2 cm) of the N plot and in the layers below 2 cm in both plots. Greater ^{137}Cs retention in the surface soil (0–2 cm) of the N plot can be ascribed to higher DOC contents. Deeper ^{137}Cs migration in both plots (below 2 cm) is also partially affected by increasing DOC. In contrast, soil pH (H_2O) was not correlated with soil ^{137}Cs in either plot. EC (mS m^{-1}) was negatively correlated with ^{137}Cs at the soil surface of the 0N plot but positively correlated with ^{137}Cs in the layers below 2 cm. The soil bulk density (g cm^{-3}) was not associated with soil ^{137}Cs.

4. Discussion

4.1. Nitrogen fertilization effects on the vertical migration of ^{137}Cs and the processes involved

Five years after the Fukushima nuclear accident, >42% of the deposited ^{137}Cs remained in the upper 3 cm of the soil in the 0 N and N plots. This is consistent with other orchards in the region affected by the Fukushima nuclear accident (Koarashi et al., 2012; Ohno et al., 2012; Sato et al., 2019b). Importantly, the vertical distribution of ^{137}Cs in the orchard soil was significantly influenced by continuous N fertilizer application—more ^{137}Cs was retained in the surface 1–2 cm layer of the N plot compared to the 0 N plot, and there was slower ^{137}Cs migration to the deeper layers (Fig. 2). These results indicate increased ^{137}Cs residence time in the surface soil that likely resulted from N fertilization. There are no similar reports of N fertilization retarding the vertical migration of ^{137}Cs in orchards, but there are a few reports from watersheds and pasture soils. For example, variation in the ^{137}Cs content of surface soils from 12 arid watersheds could be explained by the soil N-content

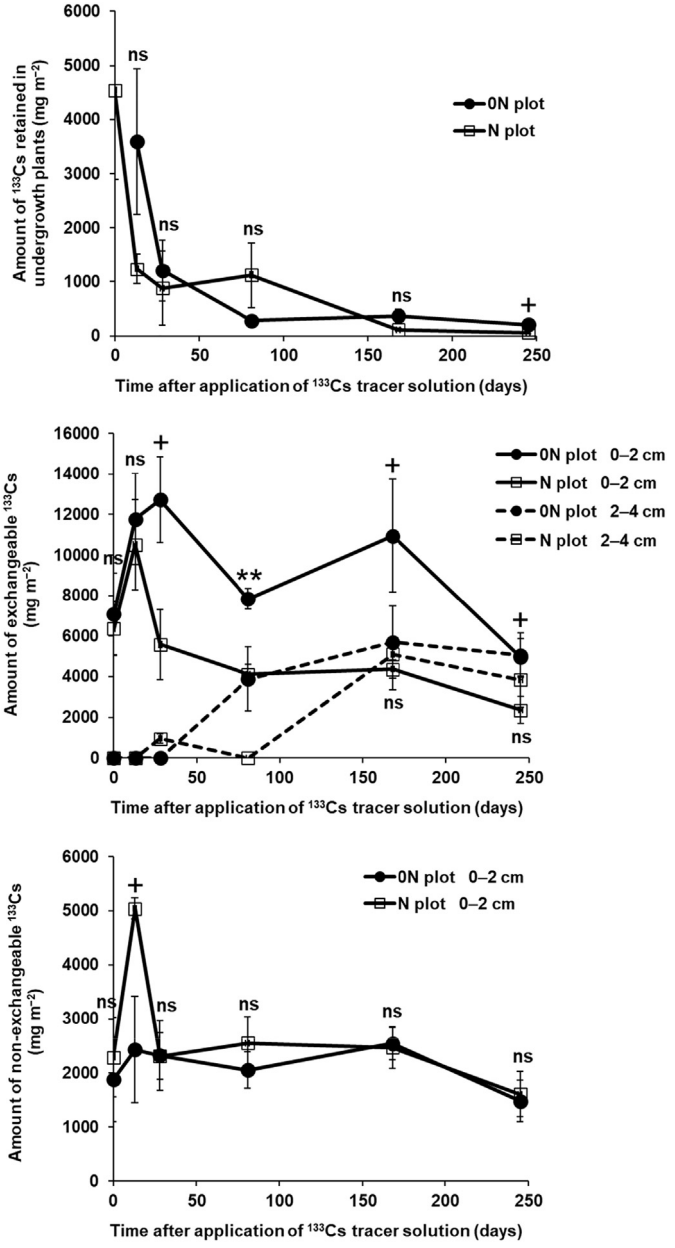

Fig. 4. Temporal changes in ^{133}Cs that was distributed in the aboveground parts of the undergrowth plants (mg m^{-2}) (the upper figure), the exchangeable ^{133}Cs (mg m^{-2}) in the 0–2 and 2–4 cm soil layers (the middle figure), and the non-exchangeable ^{133}Cs (mg m^{-2}) in the 0–2 cm soil layers (the lower figure) following the application of a ^{133}CsBr tracer solution. Differences between the 0 N and N plots in the corresponding soil layer are significant at $+P < 0.1$, $^{**}P < 0.01$; ns, not significant (t-test). Error bars represent standard errors of the means ($n = 3$).

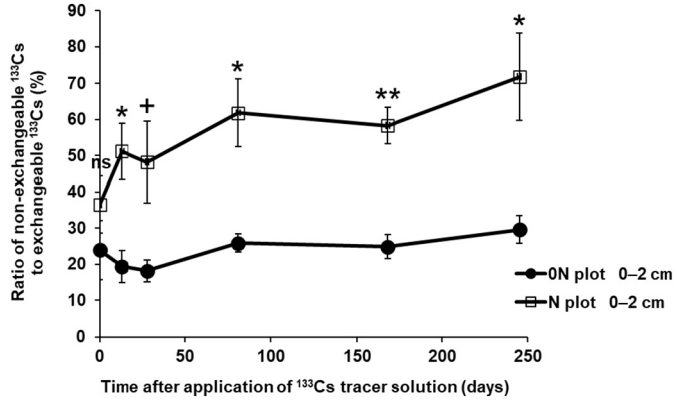

Fig. 5. Ratio of non-exchangeable ^{133}Cs (mg m^{-2}) to exchangeable ^{133}Cs (mg m^{-2}) in the 0–2 and 2–4 cm soil layers of a long-term N fertilization experimental orchard following the application of a ^{133}CsBr tracer solution. Difference between the 0 N and N plots in the corresponding soil layer are significant at $+P < 0.1$, $^{*}P < 0.05$, $^{**}P < 0.01$; ns, not significant (t-test). Error bars represent standard errors of the means ($n = 3$).

(McHenry and Ritchie, 1977), but ^{137}Cs migration in pasture soils were not affected by annual fertilization with N, P, K, and Ca for six years (Squire and Middleton, 1966).

In this study, the slower migration rate of ^{137}Cs in the N plot can be partially explained by significant reductions in exchangeable ^{133}Cs in the surface soil due to N fertilizer application (Fig. 4). This suggests that continuous N fertilization results in less exchangeable form of Cs and reduces the Cs migration rate in the N plot. Sato et al. (2019b) identified the distinct role of exchangeable ^{137}Cs in the vertical migration of ^{137}Cs to deeper soil layers in orchards, and Shiraishi (1973) also reported a strong contribution of soil exchangeable ^{137}Cs to the fruits of the satsuma orange, as observed by the global fallout. These findings highlight the importance of controlling the mobile and bioavailable forms of ^{137}Cs to reduce the risks of contaminant transfer from the soils to the fruits (via the roots).

We observed a higher proportion of the non-exchangeable form in the surface layer of the N plot, as evidenced by the data from the ^{133}CsBr tracer experiment (Fig. 5). Clay minerals affect the retention of ^{137}Cs in soils (e.g., Koarashi et al., 2012), and the Cs-fixation ability of the soils would affect the initial distribution of ^{137}Cs approximately six months after the accident (Takahashi et al., 2015). The significantly higher proportion of the non-exchangeable form in the surface layer of the N plot can be explained (in part) by immediate fixation of the deposited ^{137}Cs by the clay minerals, rather than it being retained in the organic-matter-rich fraction. This partitioning of ^{137}Cs contrasts with previous observations from organic-rich soils, including an orchard and forest (Koarashi et al., 2019) and a Scotland transect (Shand et al., 2013). Most orchards, including the one used here, are covered with undergrowth grasses; thus, there are abundant undergrowth plant roots in the surface layer of the soil (Sato et al., 2019a). The long-term application of N fertilizer reflects the higher biomass of the undergrowth plants in the N plot (Iwabuchi et al., 2012). Other studies have shown that the organic matter content has to be considered for assessments of ^{137}Cs immobilization. Cesium-137 may be involved in a shortcut element cycle of the forest and adjacent grassland, where the organic layer (humus [Oh] and fermented [Of] layers)–plants uptake–litter (Kruse-Irmer and Giani, 2003), and the organic matter also serves as a reservoir, with approximately half of the ^{137}Cs retained in the organic layer (litter [Ol] and Of layers) of the forest in the initial period after the Fukushima accident (Teramage et al., 2014). In this study, the pronounced effects of ^{133}Cs retention by the undergrowth plants in the N plot are unlikely because there were no significant differences in the ^{133}Cs content of the aboveground parts of the undergrowth plants in the tracer experiment (Fig. 4). Contrastingly, the underlying processes of vertical ^{137}Cs migration may have been affected by DOC, as evidenced by greater ^{137}Cs retention in the top 2 cm layers of the N plot and deeper migration (below the 2 cm layers) in both plots (Table 1). Prior studies have also reported the role of soil organic matter (carbon) in ^{137}Cs mobility in agricultural soils (Nakamaru et al., 2007) and ^{137}Cs migration to deeper layer in agricultural soils (Wells and Hancock, 2014). Nakamaru et al. (2007) suggested that soil organic matter could increase ^{137}Cs mobility in agricultural soils by binding the exchangeable and dissolved organic matter-bound forms.

Reduced rainwater infiltration may represent another underlying factor of retarded vertical ^{137}Cs migration in the N plot. This is supported by slower Br migration to the deeper layers of the N plot (Fig. 6), suggesting less Br infiltration with rainwater due to N fertilizer application

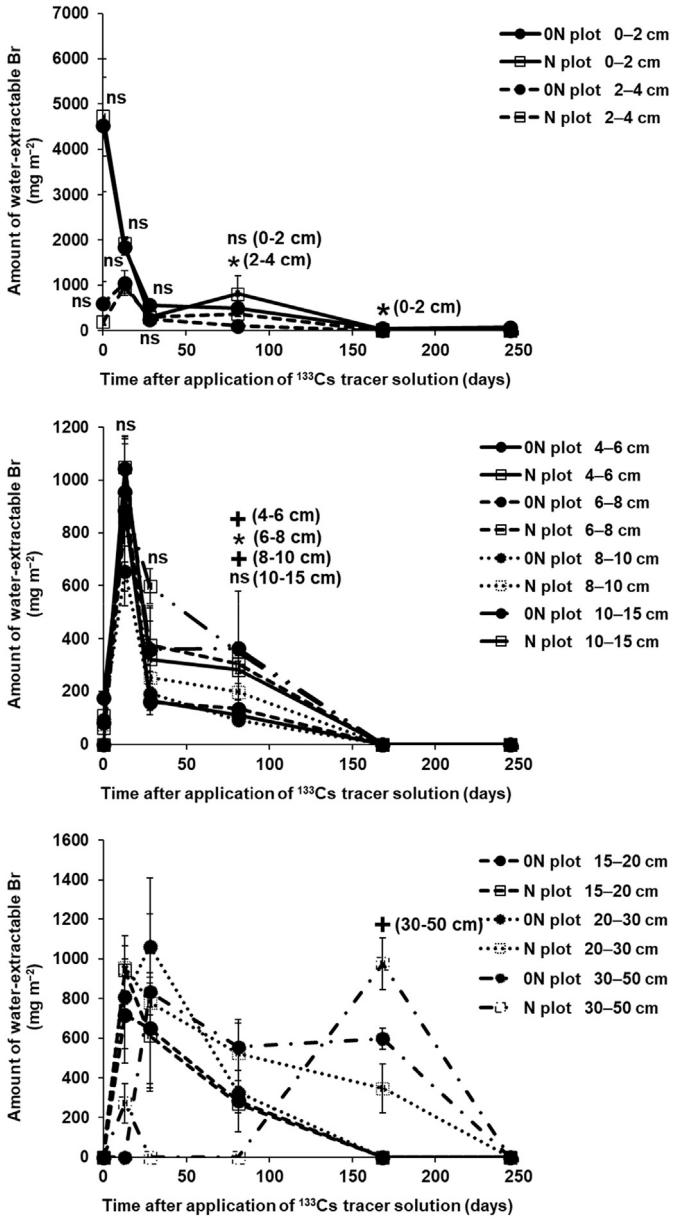

Fig. 6. Temporal changes in the vertical distribution of water-extractable Br (mg m^{-2}) in the 0–50 cm soil layers of a long-term N fertilization experimental orchard after the application of a ^{133}CsBr tracer solution. Differences between the 0 N and N plots in the corresponding soil layer are significant at $+P < 0.1$, $^{*}P < 0.05$; ns, not significant (*t*-test). Error bars represent standard errors of the means ($n = 3$).

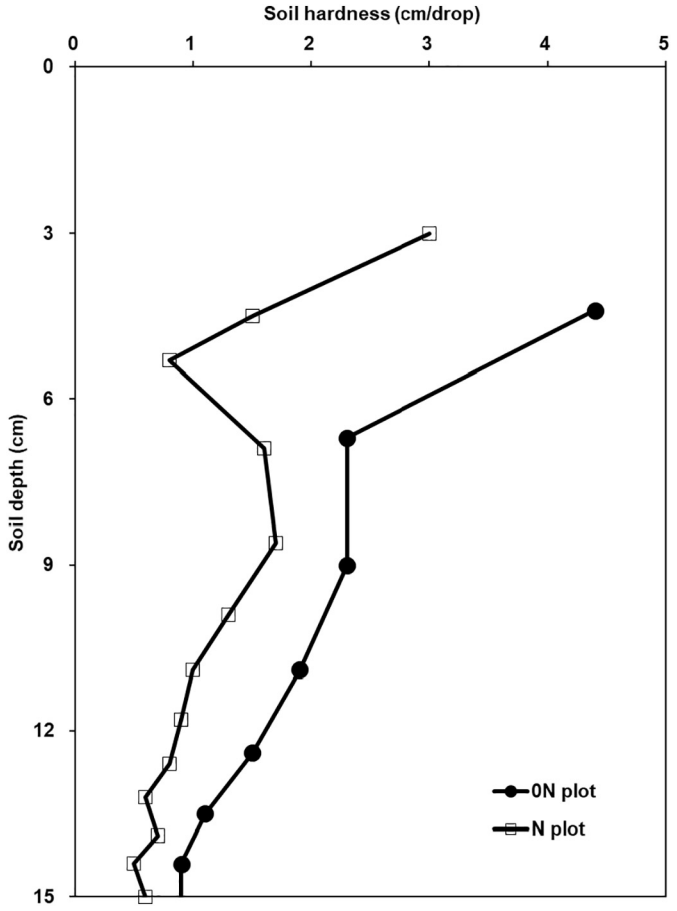

Fig. 7. Soil hardness (cm/drop) in a long-term N fertilization experimental orchard. This figure shows representative data for each plot.

and harder soils (0–15 cm; Fig. 7). Greater soil hardness in the N plot implies a lower soil pore volume (i.e., a larger soil solid phase). Moreover, ammonium (NH_4^+) ion has a disaggregating effect on the soil structure (Bresson and Boiffin, 1990), and the formation of a crust at the soil surface after rainfall events in an acidified soil with long-term ammoniacal (NH_4Cl) fertilization (Bresson and Boiffin, 1990) strongly reduces water infiltration and the downward movement of dissolved ^{137}Cs. This may explain lower ^{137}Cs migration rates in soil with continuous ammoniacal (NH_4NO_3) fertilization (Monna et al., 2009). The periodic surface application of ammonium-based fertilizers also decreases soil pH (Greenham, 1965; Sadowski et al., 1988; Matsuoka et al., 2018) and may be an important factor affecting the vertical migration of ^{137}Cs. In this study, however, there was no relationship between the pH (H_2O) of the 0–2 am and 2–15 cm layers and the vertical migration of ^{137}Cs (Table 1). This contradicts previous work showing less downward migration of ^{137}Cs in acidic soil with high clay content and more downward migration in calcareous soil with over 25 meq $(100\ g)^{-1}$ exchangeable cations (Squire and Middleton, 1966).

4.2. Nitrogen fertilizer retards the vertical migration of ^{137}Cs and may enhance its removal

The average ^{137}Cs migration rate in the experimental orchard in the five years after the accident was 0.46 cm y^{-1} in the N plot and 0.86 cm y^{-1} in the 0 N plot (Fig. 2). These rates are comparable to the range of values (0.44 to 0.97 cm y^{-1}) observed across five orchards in the accident-affected region, including the experimental orchard used

Table 1
Pearson's correlation coefficients (*r*) of soil ^{137}Cs and the soil properties, bulk density, pH (H_2O), EC, and DOC, in the surface 0–2 and 2–15 cm layers of a long-term N fertilization experimental orchard five years after the Fukushima nuclear accident.

	^{137}Cs (kBq m^{-2})							
	0–2 cm layers				2–15 cm layers			
	0N plot		N plot		0N plot		N plot	
Bulk density (g cm^{-3})	0.947	+	0.635	ns	−0.164	ns	−0.194	ns
pH (H_2O)	−0.863	ns	−0.622	ns	0.202	ns	0.265	ns
EC (mS m^{-1})	−0.961	*	−0.448	ns	0.685	**	0.023	ns
DOC (g m^{-2})	0.879	ns	0.998	**	0.920	**	0.818	**

$+P < 0.1$, $^{*}P < 0.05$; $^{**}P < 0.01$; ns, not significant; $n = 4$ (0–2 cm layers), or $n = 39$ (2–15 cm layers).

here (Sato et al., 2019b). The application of N fertilizer led to a significant reduction in the downward migration of ^{137}Cs in the orchard by approximately one-half (Fig. 2) and a significant decrease in the exchangeable form of ^{133}Cs (Fig. 4), suggesting decreased radiation risks through the absorption of exchangeable ^{137}Cs, because soil exchangeable ^{137}Cs is more available to fruits (Shiraishi, 1973). Retarded vertical ^{137}Cs migration in the soil of the N plot might also decrease contamination risks through delayed migration to the tree root-zones at approximately 10–20 cm depth (and up to 100 cm) in both plots (data not shown). This finding may also highlight an effective means of ^{137}Cs removal from a contaminated orchard.

5. Conclusions

Our findings show that the long-term application of N fertilizer in an apple orchard retarded the vertical migration of ^{137}Cs from the Fukushima nuclear accident. Slower migration in the fertilized N plot can be ascribed to lower exchangeable ^{133}Cs in the upper 2 cm and a higher proportion of non-exchangeable ^{133}Cs that was observed from ^{133}CsBr tracer experiment. This leads to increased ^{137}Cs retention at the surface and less migration in N plot, compared to more mobile exchangeable form in 0 N plot that may migrate to the deeper root-zone layers. These results suggest that continuous N fertilization may reduce the risk of radioactive contamination through tree root uptake and facilitate the mechanical removal of contaminated surface soil.

CRediT authorship contribution statement

Kaori Matsuoka: Conceptualization, Methodology, Validation, Investigation, Writing - original draft, Writing - review & editing.**Naoki Moritsuka:**Methodology, Investigation, Writing - review & editing. **Mitsuhiko Nukada:**Data curation.**Mamoru Sato:**Supervision.

Declaration of competing interest

No conflict of interest.

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Appendix A

Table A.1
Vertical distribution of ^{137}Cs and the soil properties, bulk density, pH (H_2O), EC, and DOC, in the 0–15 cm soil layers in a long-term N fertilization experimental orchard five years after the Fukushima nuclear accident.

Plot	Soil depth (cm)	^{137}Cs		Bulk density		pH (H_2O)		EC		DOC	
		kBq m^{-2}	*P*-value	(g cm^{-3})	*P*-value		*P*-value	(mS m^{-1})	*P*-value	(g m^{-2})	*P*-value
0N	0–1	7.16	ns[a]	0.103	ns	6.48	ND[b]	66.0	ND	3.26	ND
0N	1–2	43.1	<0.1	0.612	ns	6.20	<0.01	34.1	ns	4.59	ns
0N	2–3	57.0	ns	0.903	ns	5.98	<0.01	17.1	<0.1	3.89	ns
0N	3–4	49.2	<0.01	1.34	ns	5.87	<0.01	10.9	ns	3.69	ns
0N	4–5	24.3	<0.05	1.15	ns	5.79	<0.01	9.49	ns	2.55	ns
0N	5–6	18.9	<0.01	1.16	ns	5.76	<0.01	8.19	ns	2.26	ns
0N	6–7	16.1	<0.05	1.23	ns	5.71	<0.01	7.78	ns	2.17	ns
0N	7–8	13.9	<0.05	1.32	ns	5.69	<0.01	7.43	ns	2.20	ns
0N	8–9	8.14	<0.05	1.17	ns	5.69	<0.01	6.73	ns	1.76	ns
0N	9–10	5.12	<0.05	1.08	ns	5.67	<0.01	6.56	ns	1.69	ns
0N	10–11	4.31	<0.05	1.18	ns	5.66	<0.01	6.48	ns	1.76	ns
0N	11–12	3.23	<0.01	1.33	ns	5.73	<0.01	5.78	ns	1.70	ns
0N	12–13	2.11	<0.01	1.26	ns	5.80	<0.01	5.48	ns	1.29	ns
0N	13–14	1.48	<0.01	1.21	ns	5.82	<0.01	5.60	ns	1.23	ns
0N	14–15	1.14	<0.05	1.27	ns	5.81	<0.01	5.36	ns	1.32	ns
N	0–1	21.8		0.120		5.17		74.6		3.59	
N	1–2	125.8		0.839		4.77		19.1		5.95	
N	2–3	33.4		0.930		4.46		8.48		3.81	
N	3–4	12.5		1.38		4.36		8.10		3.25	
N	4–5	7.70		1.47		4.31		7.95		3.08	
N	5–6	3.81		1.11		4.26		8.12		2.01	
N	6–7	3.53		1.24		4.25		8.19		2.18	
N	7–8	2.75		1.26		4.25		7.87		2.01	
N	8–9	1.55		1.02		4.26		7.69		1.57	
N	9–10	1.21		1.17		4.28		7.63		1.58	
N	10–11	1.10		1.13		4.31		7.31		1.42	
N	11–12	0.838		1.14		4.36		7.30		1.23	
N	12–13	0.544		1.14		4.40		7.39		1.00	
N	13–14	0.564		1.25		4.46		7.42		1.07	
N	14–15	0.468		1.09		4.52		7.38		0.91	

[a] Differences between the 0 N and N plots in the corresponding soil layers are significant at $P < 0.1$, $P < 0.05$, $P < 0.01$; ns, not significant (*t*-test).
[b] ND, no determination.

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