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Distribution of radiocesium fallout on forest area throughout Japan after decades from former atmospheric nuclear tests

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To predict the movement of radioactive contamination caused by Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident is a strong concern, especially for the forest and forestry sector. To learn from the precedent, we investigated soil samples collected systematically from 316 forest sites in Japan just before the accident, which retain the global fallout ^{137}Cs (^{137}Cs-GFO) from the nuclear test bomb during the 1950s and 60s. We measured the radioactivity of ^{137}Cs-GFO in three layers of soil samples (0-5, 5-15 and 15-30 cm in depth) at each site. We divided 316 sampling sites into 10 groups separated by one longitudinal line and four transversal lines on the islands of Japan, then analyzed rainfall and geomorphological effects on ^{137}Cs-GFO inventories. In addition to the analysis of ^{137}Cs-GFO above, we examined the behavior of ^{137}Cs discharged from FDNPP (^{137}Cs-Fk) within the whole trees to study a possibility of biological effect on ^{137}Cs transport to soils from trees. We measured the radioactivity of ^{137}Cs-Fk of above- and belowground tree parts of three 26 year-old *Quercus serrata* and associated soils at a contaminated area in Fukushima in April, 2014.

We estimated an average of ^{137}Cs-GFO inventories of forest soils in Japan to be 1.7 $\pm$ 1.4 kBq/m^2 as of 2008. ^{137}Cs-GFO inventories varied largely from 0-7.9 kBq/m^2 around the country. We found high accumulation of ^{137}Cs-GFO in the north-western part facing to the Sea of Japan. We detected significant rainfall effects on the high accumulation due to winter rainfall. The vertical distribution of ^{137}Cs-GFO showed that 44% of ^{137}Cs-GFO remained within the 5 cm of soil from the surface whereas the rest of 56% was found in the layer of 5-30 cm in depth, indicating that considerable downward migration of ^{137}Cs-GFO occurred during these fifty years in forest soils in Japan. However, multiple linear regression analysis by geomorphological factors related to soil erosion, such as inclination angle or catchment area calculated from Digital Elevation Model, showed almost no significant effects on the distribution of ^{137}Cs-GFO.

The radioactivity of ^{137}Cs-Fk concentrations of fine roots collected from the 0-10 cm layer were 1600-2400 Bq/kg, which were comparable to those of one-year old branches (1400-2200 Bq/kg). The radioactivity of the fine roots was 7 times higher than that found in the soil of 50-100 cm layer (220-350 Bq/kg). This difference the radioactivity of the fine roots among the soil layers was remarkably small when compared with the 1000 times or more difference of radioactivity of soils in the same layers (one outlier sample in the 40-60 cm layer was excluded). The findings indicated that ^{137}Cs-Fk circulated through the whole tree within three years after the accident. Considering root litter fall inside the soils we estimated that contaminated ^{137}Cs on trees at the above ground part could be transported to soils through roots.

We clarified that ^{137}Cs-GFO has been held at deposited site and migrated downward gradually in soil. There are two possible major driving forces to be considered to explain the downward migration of ^{137}Cs-GFO. One is the migration of ^{137}Cs associated with vertical water movement and the other one is the transport of ^{137}Cs by root litter fall or root exudate. Further research is needed to analyze these processes to obtain reliable prediction of future distribution of ^{137}Cs-Fk.

Keywords: radioactive contamination, Fukushima Dai-ichi Nuclear Power Plant, 137Cs, forest soil, trees, secondary migration