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Effective half-lives of ^{137}Cs from persimmon tree tissue parts in Japan after Fukushima Dai-ichi Nuclear Power Plant accident

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ABSTRACT

To estimate the radiocesium decreasing rates from persimmon trees during a period of about 3 y following the accident at Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP), we conducted measurements of tree tissue parts collected in 2011–2013. The sampling was carried out in Chiba, 220 km south of FDNPP; radioactive fallouts discharged from FDNPP had mainly been observed in March–April 2011 on the sampling site. We measured ^{137}Cs concentrations in the tree tissue parts, i.e., fruits (flesh, skin and seeds), leaves and newly emerged branches, and then the effective half-lives (T_{eff}) of ^{137}Cs were calculated. Leaf samples were classified into two types by sampling months according to the growing stages, that is, immature (April–May) and mature (June–November) leaves. All these parts showed exponential declines in ^{137}Cs concentration with good adjusted contribution ratios of higher than ca. 0.7. The calculated T_{eff} values from all tissue parts were similar with the average of 229 d (range: 216–243 d). From these results, we concluded that each tree tissue was representative for the calculation of T_{eff}. For comparison to these observation results, open source food monitoring data from 2011 to 2013 including ^{137}Cs data for persimmon fruits collected in Fukushima Prefecture were used to calculate T_{eff} for persimmon trees. Values of T_{eff} were obtained for persimmon fruits grown in each local government area in Fukushima Prefecture and they ranged from 303 to 475 d.

1. Introduction

Large amounts of radionuclides were released to the environment mainly in March and April 2011 in the accident at Tokyo Electric Power Company's Fukushima Dai-ichi Nuclear Power Plant (FDNPP). We previously measured radiocesium (^{134}Cs and ^{137}Cs) concentrations in newly emerged leaves of 14 plant species, that is, herbaceous plants and deciduous and evergreen trees (Tagami et al., 2012), collected from the campus of the National Institute of Radiological Sciences (NIRS) in Chiba, Japan located about 220 km south of the FDNPP. The NIRS had received radioactive fallout deposited mainly as wet deposition and the inventory of ^{137}Cs in this area was ca. 15 kBq m^{-2} according to our previous study (Ishii et al., 2013). There were no emerged leaves of deciduous trees in March when heavy deposition was observed for the NIRS campus because it was before the growing season start. Radiocesium originated from the FDNPP was located in a surface thin soil layer at that time. If the root uptake was the major pathway of radiocesium transfer to newly emerged parts of plants, then herbaceous plants should have higher radiocesium concentrations than those in trees because root zones of trees are usually deeper than those of herbaceous plants. However, what we observed was a different order, i.e. concentrations were decreased as: evergreen trees > deciduous trees > herbaceous plants. Similarly, Yoshihara et al. (2013) also found higher radiocesium concentrations in evergreen trees than in deciduous trees. Thus we assumed that major radiocesium uptake pathways to plants were the above ground uptakes of radiocesium through leaves and tree barks in the first year after the accident, and the root uptake process was much smaller than that of the above ground uptake.

To minimize the foliar uptake of radiocesium to orchard fruits in contaminated areas of Fukushima Prefecture, countermeasures such as washing tree trunk bodies, removal of bark, and removal of the surface soil contaminated with radiocesium to prevent leaching to a deeper layer were carried out in many orchards. Even with such countermeasures, people still had concerns because radiocesium taken up in trees could be retained in the tree bodies for a while as was observed after the Chernobyl nuclear power plant accident (Antonopoulos-Domis et al., 1991; Goor and Thiry, 2004; Ünlü et al., 1995). According to the food monitoring data reported monthly by

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the Japanese Ministry of Health, Labour and Welfare (MHLW, 2014), radiocesium concentrations in fruits, such as peaches, apples and Japanese pears, had decreased significantly by more than 50% in 2012 compared to the first year, and their concentration levels were much less than the Japanese food standard limit of 100 Bq kg^{-1}-fresh in 2013. From this result it was assumed that radiocesium was removed rapidly from fruits trees; however, the decreasing rates were not reported yet for fruits in Japan. Pröhl et al. (2006) reported ecological half-lives of ^{137}Cs in various types of animal and plants observed in European countries using monitoring data and found the half-lives were about 5–6 y for pipfruit before and after the Chernobyl accident. Since the climate conditions in Japan differ from those of European countries, it is necessary to obtain effective half-life data in order to assess future use of fruit trees.

Among deciduous type orchard trees, persimmon fruits showed higher radiocesium concentrations than other fruits and the mechanism for the retention was not known. Persimmon fruits are eaten fresh, dried or cooked; one of the local specialties in northern Fukushima is semi-dried persimmon fruit. The water content of semi-dried persimmon is about 50% while that in fresh fruit is about 85%. During the drying process, radiocesium would not be removed, and thus, the concentration of radiocesium would be increased even more in the final products. Due to the water content decrease, fresh persimmon fruits used to make semi-dried fruits should not have a radiocesium concentration higher than 30 Bq/kg-fresh to meet the food standard limit. In order to estimate the concentration in fruits, we found leaves could be a good indicator (Tagami and Uchida, 2014a); however, how fast the radiocesium would be removed from persimmon trees has not been studied yet. Thus in this paper, we have reported on effective half-lives of ^{137}Cs from persimmon trees affected by the FDNPP fallout; we used both our observation results for persimmon trees grown on the NIRS campus as well as using open source food monitoring data for Fukushima Prefecture. The term "effective half-life" is defined as the time requited for a 50% decline of ^{137}Cs in a tree in a natural condition.

2. Materials and methods

2.1. Persimmon sample collection in NIRS campus

We regularly collected fruits (flesh, skin and seeds), leave and newly emerged branches of persimmon trees grown on the NIRS campus (Fig. 1) from April 26, 2011 to November 20, 2013. All these parts were newly grown every year starting from mid-April. Since some previous works had used fruits and leaves to calculate half-lives (Antonopoulos-Domis et al., 1991; Pröhl et al., 2006; Ünlü et al., 1995), we also selected these parts in this study. Two persimmon trees were used as one sample; these trees stood within 5 m of each other and similar heights of about 5–6 m; their ages were unknown. Cesium-137 deposition density in this field has been reported elsewhere (Tagami and Uchida, 2012) and the deposition density was 12.1 kBq m^{-2} at March 26, 2012.

Immediately after the collection, samples were transferred to a laboratory and weighed to obtain the fresh weight. Leaf samples were usually separated into two portions; one portion was washed with dish-washing detergent including surfactant to remove dust from the surface and then washed with tap water five times to remove detergent foam and finally rinsed with reverse osmosis (RO) water, and the other portion was unwashed. For fruits, a whole fruit was washed with running tap water and rinsed with RO water. Then the water was removed with paper towels from the fruits, and each tissue part (flesh, skin and seeds) was separated and weighed.

All tissue part samples were dried to a constant weight at 80 °C in an electric oven for at least 2 d. Each dried sample was pulverized, and then, the powder sample was transferred to a 100 mL polystyrene container. The weight of sample transferred and height (the level to which the sample was filled in the container) were measured for correction of gamma-ray counting. The ^{137}Cs radioactivity was then determined with a germanium detecting system (GEM Model 137CN2-2, Seiko EG&G). A mixed gamma standard solution (Amersham, QCY-46) was used for an efficiency correction and three reference standard materials IAEA-156, 373 and 375 were used for an accuracy check. Data were decay-corrected to the collection date of the sample.

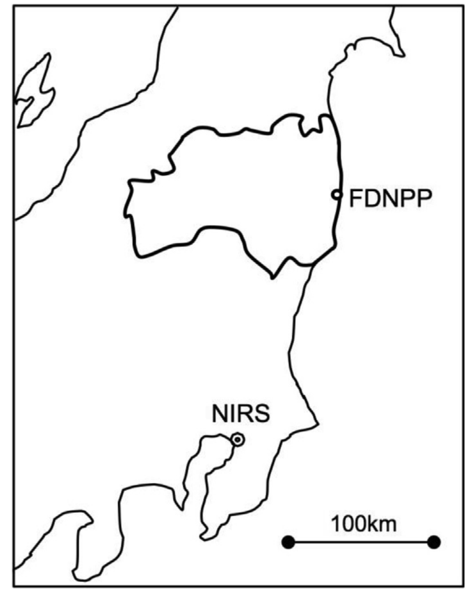

Fig. 1. Sampling location map.

Data were used to calculate effective half-life (T_{eff}), which is defined as.

$$T_{eff} = \ln 2 / \lambda_{eff} \quad (1)$$

where λ_{eff} is the ^{137}Cs loss rate in trees. Value of λ_{eff} is obtained from the slope of the exponential decline in ^{137}Cs concentration in the leaves and fruits over time as follows:

$$A_t = A_0 \exp\left(-\lambda_{eff}\, t\right) \quad (2)$$

where A_t is ^{137}Cs concentration at time t and A_0 is the expected initial ^{137}Cs concentration. Using the concentration data, a best fit exponential trend line for each tissues was computed using JMP™ software (SAS Institute Inc.).

2.2. Persimmon sample data in Fukushima Prefecture

To estimate T_{eff} for persimmon trees in Fukushima Prefecture, open source ^{137}Cs concentration data in edible part of persimmon fruits collected from local government (cities, towns and villages) in Fukushima Prefecture since 2011 were used (MHLW, 2014). Although distribution of radioactivity concentration in a local government area was not uniform, we assumed that the amount of ^{137}Cs deposited to the land surface was the same for this analysis. To select areas, we also set the criterion that more than three data per year should be reported in 2011–2013 by the local government. Finally, data from Date City, Fukushima City, Koori Town, Koriyama City, Kunimi Town, and Nihonmatsu City were selected for analysis.