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The ^{137}Cs activity concentration of suspended and dissolved fractions in irrigation waters collected from the 80 km zone around TEPCO's Fukushima Daiichi Nuclear Power Station

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ABSTRACT

Fifty-four samples of irrigation water were collected in 2014 from agricultural ponds, rivers, and dams within the 80 km zone around TEPCO's Fukushima Daiichi Nuclear Power Station (FDNPS). The samples were filtered with a 0.45 μm pore-size membrane filter to produce suspended and dissolved fractions. The ^{137}Cs activity concentration of the suspended fraction varied from 1.5 to 300 Bq g^{-1} dry weight and was significantly higher than that in the soil around each sampling site. The range of ^{137}Cs activity concentrations in the dissolved fraction varied over three orders of magnitude at 0.0075–6.7 Bq l^{-1}, which was a larger range than that of the suspended fraction; the higher values for samples were from within the 20 km zone. In the dissolved fraction 87 ± 9% of the ^{137}Cs (n = 37) was in a monovalent cationic form (Cs^+) and therefore potentially mobile. The distribution coefficient (Kd) ranged from 4100 to 2,100,000; the geometric mean value (110,000) was higher than that reported by the IAEA (2010). The geometric mean Kd of samples collected from the 20 km zone was 61,000 (n = 27), which was significantly lower than that collected from 20 to 80 km zone (200,000, n = 27). The Kd-value was negative correlated with the concentration of stable ^{133}Cs and the electric conductivity in the dissolved fraction. This shows relatively higher mobility of radiocaesium in irrigation waters may occur when there are higher contents of cations present.

1. Introduction

The most powerful recorded earthquake in the history of Japan (magnitude 9.0 on the Richter scale), with an epicenter offshore from the Tohoku region, occurred on March 11, 2011. An associated tsunami in the Northern Pacific coast of Japan brought catastrophic damage. TEPCO's (Tokyo Electric Power Company Holdings, Inc.) Fukushima Daiichi Nuclear Power Station (FDNPS) lost all power due to the tsunami, and the cooling systems completely shut down. Significant quantities of radionuclides were released and deposited onto the surrounding landscape and the Pacific Ocean of TEPCO's FDNPS accident site in March 2011 (Chino et al., 2011). Caesium isotopes ^{134}Cs and ^{137}Cs (radiocaesium) were the major long-lived radionuclides released during the accident and are important for the assessment of radiation exposure to the public. The Standard limits for radionuclides in foods were established by the Ministry of Health, Labour and Welfare (MHLW), Japan on April 1, 2012 and were determined on the basis of 1 mSv y^{-1}. The limit in general foods is 100 Bq kg^{-1} for radiocaesium, including the contribution of ^{90}Sr, Pu and ^{106}Ru (MHLW, 2012). The average concentration of radiocaesium in agricultural plants collected in Fukushima-shi and Date-shi, Fukushima Prefecture in 2012 was 7.6 (<0.2–40) Bq kg^{-1} fresh weight (fw), decreasing to 2.0 (<0.1–14) Bq kg^{-1} fw in 2013. The estimated internal radiation doses through food ingestion for males and females (over the age of 19) were 0.066 and 0.052 mSv y^{-1} in 2012, and those in 2013 were 0.016 and 0.012, respectively, reflecting the decreases in the radiocaesium activity concentration of foods with time elapsed (Tsukada et al., 2016). The results show the internal radiation doses from food ingestion are low even if continuously consuming food products produced in Fukushima Prefecture. However, radiocaesium activity concentrations of rice collected at 2013 (28 bags) and 2014 (2 bags) were over the limit

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(Fukushima Prefecture, 2017), and the health risk of radiation exposure needs to be estimated for the pathways of food ingestion from agricultural production. Rice cultivation in the evacuation zone (difficult-to-return area) may be started in the near future.

There were approximately 3700 reservoirs that may have received radiocaesium deposited onto their catchment areas. These reservoirs supply irrigation water for rice paddy fields in Fukushima Prefecture. Suzuki et al. (2015) reported that the presence of radiocaesium dissolved in water increases the concentration of radiocaesium in rice. Therefore, the Ministry of Agriculture, Forestry and Fisheries, Japan (Tohoku Regional Agricultural Administration Office, 2016) and Fukushima Prefecture (2016) have carried out radiocaesium monitoring of irrigation waters in Fukushima Prefecture.

To estimate the migration of radiocaesium within the terrestrial environment, and from different contaminated forested catchments to the Pacific Ocean, information is needed on the radiocaesium activity concentration of both the dissolved and solid phases in irrigation water. The data will enable the bioavailability of radiocaesium to be quantified by calculating the distribution coefficient (Kd) in irrigation system for paddy fields for different areas around the FDNPS. In this study, irrigation waters were collected from 54 reservoirs in an 80 km zone around TEPCO's FDNPS. The study determined the ^{137}Cs activity concentration in both suspended and dissolved fractions, and also monovalent ionic cation $^{137}Cs^{+}$ in the dissolved fraction.

2. Materials and methods

Irrigation waters were collected from 27 reservoirs within a 20 km zone around TEPCO's FDNPS and 27 reservoirs from a 20–80 km zone from April to October 2014 (Fig. 1). The sampling sites were relatively highly contaminated areas in the northwest direction from and around the FDNPS, which were in the evacuation zones within the 20 km zone, limited cultivation, and the catchment areas of the reservoirs were complex areas of on and off evacuation zone. Therefore the boundary was drawn 20 km from the FDNPS for comparison purposes. Irrigation waters were collected from 42 ponds, two dams, and 10 rivers. Water samples were collected from the surface with a plastic bucket or at a depth of 0.5 m with a Bandung water sampler, and put into a 20 L plastic container. The water sample was filtered through a 0.45 μm pore-size membrane filter (Millipore HVLP09050) and separated into the suspended and dissolved fractions. The suspended fraction on the filter was dried at 70 °C for one week, cut into small pieces and then compressed into a plastic vessel. Nitric acid was added to the dissolved sample (1 ml l^{-1}) and five to 10 L of the sample were evaporated to 50 ml and then measured with a Ge detector. The activity concentration of ^{137}Cs occurring as the monovalent ionic cation Cs^{+} in the dissolved fraction was determined with an Empore TM Caesium Rad Disk (3 M). Two disks were used to collect $^{137}Cs^{+}$ in the dissolved fraction because the yield of Cs^{+} was over 95% (Investigative Committee of Technical Report on Monitoring Radio-Caesium in Water, 2015). Five liters of the filtered sample were put into a stainless steel container and Cs^{+} was adsorbed onto hexacyanoferrate in the disk under 0.4 MPa with a compressor.

All samples were measured with a Ge detector connected to a multichannel analyzer system (Canberra, GC3018, GC4018 and GC4020). The detection efficiency of the Ge detector was dependent on the sample thickness and was obtained using the mixed standard radionuclides material made by the Japan Radioisotope Association. Counting statistics standard deviations for ^{137}Cs activity concentration in the sample were less than 10% of the value.

The concentrations of NH_4, K and Ca in the dissolved fraction were measured with ion chromatography (Thermo Scientific Dionex, ICS-1100) and that of ^{133}Cs in the samples was measured with ICP-MS (Perkinelmer, ELAN DRC2). Electric conductivity in the water samples was measured with an Eh meter (Horiba, B-771). Total organic carbon in the dissolved sample was determined with a total organic carbon analyzer (Shimadzu, TOC-L).

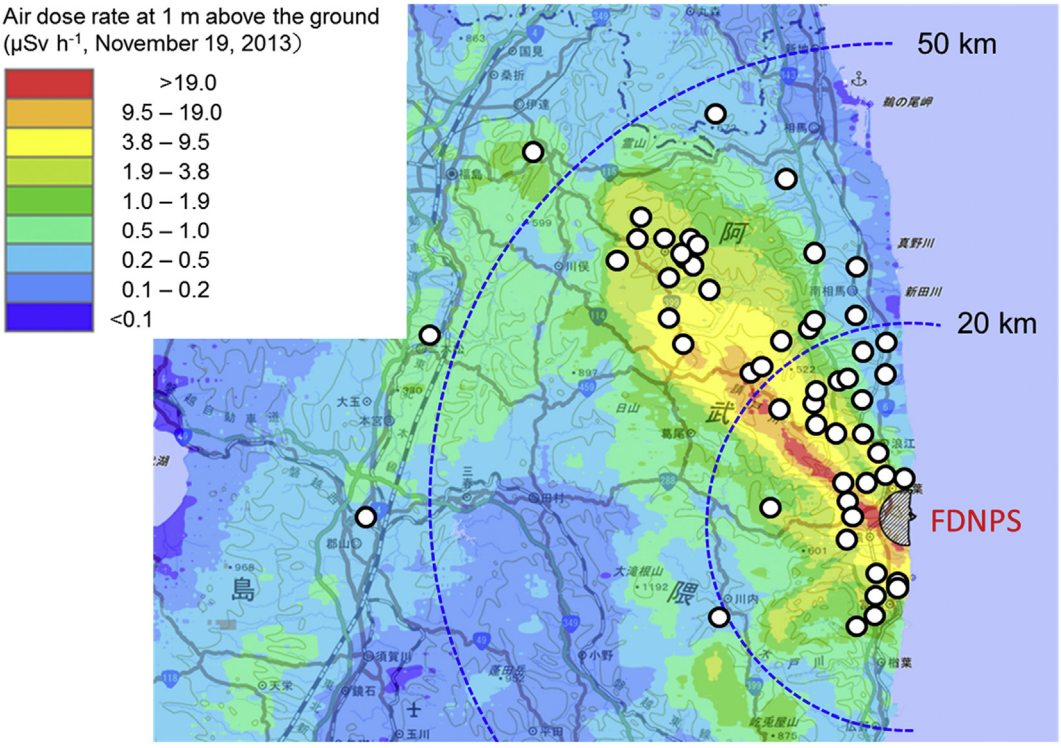

Fig. 1. Sampling location in Fukushima Prefecture. Map of air dose rate according to the fifth airborne monitoring survey (NRA, 2014) on November 19, 2013.