Journal of Environmental Radioactivity 146 (2015) 35–43

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Depth profile and mobility of ^{129}I and ^{137}Cs in soil originating from the Fukushima Dai-ichi Nuclear Power Plant accident

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ARTICLE INFO

Article history:
Received 7 December 2014
Received in revised form
21 March 2015
Accepted 23 March 2015
Available online

Keywords:
FDNPP accident
Iodine-129
Cesium-137
Depth profile
Downward migration

ABSTRACT

The ^{129}I derived from the FDNPP accident were clearly identified near the surface and showed a trend of rapid decrease with depth. The FDNPP ^{129}I and ^{137}Cs was 51.6 ± 1.7 mBq cm^{-2} and 88.2 ± 27.1 kBq cm^{-2} (average of four cores inventory) respectively. On average, 91% of the FDNPP ^{129}I existed within the top 5 g cm^{-2} and 98% within the top 10 g cm^{-2} and average of 100% of the FDNPP ^{137}Cs existed within the top 5 g cm^{-2}. From the observation of the temporal variation of depth profiles from the same upland field (Kawauchi village, 20 km away from the FDNPP to the southwest direction), downward migration rates of 0.81 ± 0.32 g cm^{-2} yr^{-1} for the FDNPP ^{129}I and 0.19 ± 0.17 g cm^{-2} yr^{-1} for the FDNPP ^{137}Cs were estimated. A simple diffusion model was introduced to evaluate the downward mobility of the FDNPP-derived ^{129}I and ^{137}Cs. The apparent diffusion coefficients D of 0.0086 ± 0.0034 and 0.0011 ± 0.0010 g^2 cm^{-4} d^{-1} were obtained for ^{129}I and ^{137}Cs, respectively. These values might be representative for Haplic Gray lowland soils in near the steady state under humid temperate climate.

1. Introduction

A serious nuclear accident occurred at the Tokyo Electric Power Company's (TEPCO) Fukushima Dai-ichi Nuclear Power Plant (FDNPP) after the tsunami on March 11, 2011. A large amount of radionuclides such as iodine-131 (^{131}I: $T_{1/2}$ = 8.01 d), cesium-134 (^{134}Cs: $T_{1/2}$ = 2.07 d) and cesium-137 (^{137}Cs: $T_{1/2}$ = 30.01 d) were discharged in the environment (Tagami et al., 2011). There are many studies related to measurement of such radionuclides, spatial distribution and total releases of radioactivity (Saito et al., 2015; Tsuruta et al., 2014). Iodine-129 (^{129}I: $T_{1/2}$ = 1.57 × 10^7 yr) was also produced by the fission of ^{235}U and ^{239}Pu in FDNPP, emitted into the environment for a short time and deposited into a highly localized area (Muramatsu et al., 2015). Investigating of ^{129}I in the soil is important mainly for two aspects. One is concerned with the reconstruction of the distribution of ^{131}I deposition through the measurement of ^{129}I. Iodine-131 is considered to dominate the primary dose in surface soil after the FDNPP accident and might cause thyroid cancer when ingested (Bennett et al., 2006). The other aspect concerns, the importance of mobility of ^{129}I in the soil for the safety assessment of the geological disposal of high-level radioactive waste. On a long-term basis, ^{129}I is classified as one of the first-priority radionuclides for assessment because ^{129}I is expected to be a major source of radiation exposure in humans in view of its high mobility in minerals and soil after the closure of the repository (FEPC and JAEA, 2007). Because soil is the nearest shielding material that separates humans beings from the underground environment, the evaluation of ^{129}I mobility in the soil is essential.

Straume et al. (2006) reported the mobility of ^{129}I and ^{137}Cs to reconstruct thyroid dose following the Chernobyl accident (1986). ^{129}I and ^{137}Cs are retained firmly in the top ~20 cm of the soil, which were treated 7 and 11 years after the Chernobyl accident. They also suggested that the correlation between ^{129}I and ^{137}Cs deposition distribution across the country of Belarus was poor.

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http://dx.doi.org/10.1016/j.jenvrad.2015.03.029
0265-931X/

Hence, ^{129}I is a better proxy for ^{131}I than ^{137}Cs. These results couldn't be applied directly to Japan because soil properties are totally different due to the climatic conditions including the precipitation. Actually in Japan, the average annual precipitation is more than 1000 mm which is twice that of Ukraine. Also the temperature in Japan is higher than Ukraine. Consequently the soil contains more organic substance in Japan. Therefore, the investigation of the FDNPP ^{129}I and ^{137}Cs depth profiles in the soil for Japan is needed.

Before the FDNPP accident, ^{129}I in the soil of Japan has mainly come from the spent nuclear fuel reprocessing plants (RP) such as Sellafield, Marcoule, and La Hague (Reithmeier et al., 2006). This atmospheric ^{129}I overwhelms the naturally occurring ^{129}I in the soil. European reprocessing plants which are currently under operation are considered the predominant source of ^{129}I for the global environment, including distant countries as Japan (Suzuki et al., 2008). There are two reprocessing plants in Japan, Tokai RP (~130 km north to Tokyo) and Rokkasho RP (~700 km north to Tokyo). Though the amount of ^{129}I emitted from Tokai or Rokkasho RP was smaller than other RP in the EU (UNSCEAR, 2000; Aomori prefecture, 2011), ^{129}I concentration around these RPs areas are high (Muramatsu and Ohmomo, 1986). Several radionuclides emitted from the Chernobyl accident in 1986 (IAEA, 2006) were detected in Japan (Aoyama et al., 1986, 1987); however, no positive ^{129}I signal attributable to the Chernobyl accident was observed (Toyama et al., 2013).

Background ^{137}Cs in the soil (mainly as a result of the global fallout from atmospheric bomb testing and nuclear accidents) is as low as 30 Bq kg^{-1} in Fukushima prefecture (Igarashi et al., 2003; NRA, 2012).

Cesium-134 and ^{137}Cs were also investigated to compare with the FDNPP derived ^{129}I. Because the short lived ^{134}Cs can be detected only soon after a nuclear accident, identification of the FDNPP ^{134}Cs is evidence of the presence of the FDNPP radionuclides. Cesium-137 can be compared to the FDNPP ^{129}I much later. The objectives of this study are 1) to estimate the downward mobility of the FDNPP derived ^{129}I and ^{137}Cs from time variation of depth profiles and 2) to estimate diffusion coefficients from the comparison with a simple one dimensional diffusion model. Our results were compared to other reports about the Chernobyl and Fukushima accidents.

As mentioned earlier, ^{129}I originating from spent nuclear fuel reprocessing plants was present in the environment even before the FDNPP. The FDNPP ^{129}I should be clearly distinguished from the background ^{129}I. In this study, background ^{129}I was effectively subtracted by specifying the sampling field for which the details are described in the next section.

2. Materials and methods

2.1. Sampling and materials

In Kawauchi village (37°19′43″, 140°50′09″), 20 km away from the FDNPP to the southwest direction has upland fields based on Haplic Gray lowland soils (NIAES, Soil Information Web Viewer). The gray lowland soil is one of the dominant soil types, which accounts for approximately 30% of paddy fields in northeast Japan. Four cores (30 cm length, φ5 cm) were collected from this area at different times between April 2011 and March 2012 (ABK12A1 on April 29, 2011, ABK12B1 on October 9, 2011, ABK12C1 on December 23, 2011, and ABK12D1 on March 16, 2012) to observe the temporal variation of the depth profile.

To evaluate precisely the depth profile of the FDNPP ^{129}I, we chose the sampling fields according to the following conditions: 1) open field, 2) turned over before the accident, and 3) undisturbed after the accident. Condition 1) assured that radionuclides were deposited directly from the atmosphere. To identify the FDNPP ^{129}I from the preexisting ^{129}I, condition 2) was preferable because a sharp increase in ^{129}I concentration due to the FDNPP accident would be clearly distinguished on the homogenous depth profile. The Haplic Gray lowland soils fulfill these conditions because the soil was tilled and well mixed by farmers until just before the FDNPP accident. After the FDNPP accident, upland fields in the contaminated region tended to be abandoned. Artificial (exclude Haplic Gray lowland soils. Example reformed soils) fields also fulfill these conditions.

2.2. Experimental procedure

The collected soil core was sliced into 1.5 or 3 cm layers. After the soil samples were air dried, then they were sieved through 2 mm mesh sieved. After heating them at 80 °C in an oven for 48 h, they were pulverized to fine powder and homogenized using a ball mill (Matsuzaki et al., 2007). After homogenization, each soil sample was stuffed into a polystyrene test tube up to a height of 2 cm including ^{134}Cs and ^{137}Cs. The gamma-ray energy peak at 605 keV for ^{134}Cs and 662 keV for ^{137}Cs was measured respectively by a Well-type HPGe semiconductor detector (EGPC 250-P15, CANBERRA) at the Nihon University. The spectrometer was calibrated with a standard material which was prepared by the Nihon University using JRIA (Japan Radioisotope Association) activity standard solution. The gamma-ray was also calibrated with homemade standard material. The measured radioactivity was corrected for the decay with respect to the sampling date. The carbon concentration in each dry soil sample was determined by an NC analyzer (Sumigraph, NC-22F; Sumika Chemical Analysis Service, Osaka, Japan) at the National Institute for Agro-Environmental Sciences, Tsukuba, Japan.

Iodine was extracted by the pyrohydrolysis method described by Muramatsu et al. (2008), where an aliquot of dry soil (0.2–0.5 g) was mounted on a ceramic boat covered with V_2O_5, and heated up to 1000 °C in a quartz tube under oxygen and water vapor flow. The extraction yield of iodine reaches more than 90% within 30 min (Schnetger and Muramatsu, 1996). The evaporated iodine was trapped in 2% tetramethyl ammonium hydroxide as an alkaline solution (total volume about 10 mL). An aliquot (1 mL) was separated for determination of the stable iodine concentration by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500) (Muramatsu and Hans Wedepohl, 1998). The detection limit in this method was about 0.2 ppb in sample solutions, which is about 4 ppb iodine in soil, when a 0.5 g soil is diluted in 10 mL. The statistical error (SD) by ICP-MS measurement is up to 1% and other errors related to ICP-MS determination (calibration, matrix effects etc) are in total around 3%.

The standard solution containing 12,690 ppm iodine (Orion Ionplus Application Solution Iodine Standard 0.1 M, Thermo) of 2.0–8.0 mg was added to a trap solution as iodine carrier, from which iodine was purified by sequential solvent and back extraction and finally extracted as AgI precipitation. The AgI was dried and pressed into the cathode of an accelerator mass spectrometry (AMS) system (Matsuzaki et al., 2007). The ratio of ^{129}I/^{127}I was measured by AMS at Micro Analysis Laboratory, Tandem Accelerator (MALT; The University of Tokyo). The ^{129}I concentration in the soil was calculated from the AMS results and normalized by the standard Z94-0596 provided by PRIME Lab at Purdue University (Indiana, USA) (Sharma et al., 1997). The mechanical detection limit of ^{129}I-AMS system at MALT achieved ^{129}I/^{127}I $< 1 \times 10^{-14}$ but actual background is regulated by ^{129}I amount in iodine carrier (Ionplus), which has ^{129}I/^{127}I $= 18.0 \pm 0.9 \times 10^{-14}$ (Matsuzaki et al., 2007).