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Short communication

Radiocaesium contamination and dose rate estimation of terrestrial and freshwater wildlife in the exclusion zone of the Fukushima Dai-ichi Nuclear Power Plant accident

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ABSTRACT

To characterise the radioactive contamination of terrestrial and freshwater wildlife caused by the Fukushima Dai-ichi Nuclear Power Plant accident, biological samples, namely, fungi, mosses, plants, amphibians, reptiles, insects, molluscs, and earthworms, were collected mainly from the forests of the exclusion zone in the Fukushima Prefecture from 2011 to 2012. Caesium-134 and ^{137}Cs were detected by gamma spectrometry in almost all the samples. Fungi, ferns, and mosses accumulated high amounts of radiocaesium, as they did in Chernobyl, with ^{134}Cs + ^{137}Cs activity concentrations of 10^4–10^6 Bq kg^{-1} fresh mass (FM). Earthworms, amphibians, and the soft tissue of the garden snail *Acusta despecta sieboldiana*, also had levels as high as 10^4–10^5 Bq kg^{-1} FM of ^{134}Cs + ^{137}Cs. Most of the estimated total (internal + external) dose rates to herbaceous plants, amphibians, insects, and earthworms were below the corresponding derived consideration reference levels (DCRLs) recommended by the ICRP. This suggests that, in most cases, there was little chance of deleterious effects of ionising radiation on these organisms in the exclusion zone for the first year after the accident, though the dose rates were underestimated mainly due to the lack of consideration of short-lived radionuclides.

1. Introduction

On 11 March 2011, a catastrophic earthquake (M 9.0) occurred in the northwest Pacific, about 130 km off northeastern Japan. This triggered a gigantic tsunami, which seriously damaged the electric system of the Fukushima Dai-ichi Nuclear Power Plant (F1-NPP). As a consequence, the cooling systems of the nuclear reactors failed, and the reactors were damaged as the fuel overheated and melted. This accident at the F1-NPP led to the release of radionuclides into the atmosphere. The highly volatile fission products such as ^{129m}Te, ^{132}Te, ^{131}I, ^{134}Cs, ^{136}Cs, and ^{137}Cs isotopes were carried together with air parcels, and subsequent wet and dry depositions led them to accumulate on the ground and in water bodies.

Large areas in the eastern parts of Japan from the Kanto to Tohoku districts, mainly the Fukushima Prefecture, were contaminated with the radionuclides. Naturally, wild animals and plants in the area were also radioactively contaminated as indicated by environmental monitoring (e.g., MEXT, 2012a; Fukushima Prefectural Government, 2012) and some radioecological studies (e.g., Ayabe et al., 2014; Hasegawa et al., 2013; Takahara et al., 2015).

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Among the contaminated areas, the exclusion zone surrounding F1-NPP was the most severely contaminated due to the accident. Radioecological studies on wildlife can generally be carried out efficiently in exclusion zones, since only small amounts of samples and short counting times are required for radioactivity determination. Contamination levels of various wildlife species in the exclusion zone were important as one of the basic data based on which the potential impact of the F1-NPP accident was evaluated. Accumulators of radionuclides, e.g. mosses and fungi in case of radiocaesium contamination (IAEA, 2006), make particularly effective tools for monitoring environmental contamination levels.

The likelihood of observing the effects of ionising radiation on wildlife is higher in the exclusion zone, therefore, radioactive contamination levels of wildlife in this zone are also important to characterise the risks ionising radiation pose to wildlife. The effects of ionising radiation on non-human biota have been a matter of concern since the accident (Callaway, 2013; Schiermeier, 2011a, b; Strand et al., 2014), and some field studies have reported possible radiation effects in the exclusion zone (e.g., Akimoto, 2014; Hiyama et al., 2015; Kubota et al., 2015a; Møller et al., 2013; Watanabe et al., 2015). However, the absorbed dose rates to wildlife may not have been accurately estimated in these field studies. There have been only a few studies in which internal dose rates were estimated on the basis of radionuclide activity concentrations actually measured in terrestrial and freshwater organisms (e.g., Fuma et al., 2015; Kubota et al., 2015a, b; Urushihara et al., 2016; Yamashiro et al., 2013).

In spite of these conditions, a limited number of studies (e.g., Tazoe et al., 2012) have reported radioactivity data in wildlife collected in the exclusion zone shortly after the F1-NPP accident, i.e. for the first year of the accident, while some other studies (e.g., Dohi et al., 2015; Fuma et al., 2015; Kubota et al., 2015a, b; Matsushima et al., 2015; Okano et al., 2016) have reported data on the wildlife collected in 2012 or later, when radioactive contamination levels were generally expected to be lower than they were in 2011.

The objective of this study is to characterise radioactive contamination of terrestrial and freshwater wildlife in the exclusion zone in the first year of the F1-NPP accident and to conduct a preliminarily evaluation of the radiation risks to these organisms. To accomplish this, various terrestrial and freshwater biological samples and environmental media (water and sediment) were collected in the exclusion zone as well as some other areas of the Fukushima Prefecture from April 2011 to March 2012, and the activity concentrations of ^{134}Cs and ^{137}Cs were determined. Absorbed dose rates to wildlife were estimated from the activity concentration data and ambient dose rates.

2. Materials and methods

2.1. Sampling

Sampling sites were selected in the Fukushima Prefecture based on radioactive contamination levels deduced from airborne monitoring survey data (e.g., MEXT, 2011). Sampling was mainly carried out in 12 sites of the areas that were heavily contaminated by the F1-NPP accident (ground deposition of ^{134}Cs + ^{137}Cs: >1,000,000 Bq m^{-2}, as of 29 April 2011; MEXT, 2011). As shown in Fig. S1, these sampling sites are located in the northern part of the Abukuma Mountains, and are within the exclusion zone (METI, 2011). For comparison, sampling was also carried out in five sites of the less contaminated areas (ground deposition of ^{134}C + ^{137}Cs: <60,000 Bq m^{-2}, as of 29 April 2011; MEXT, 2011) neighbouring the exclusion zone (Fig. S1). Details of each sampling site are shown in Table 1.

Terrestrial and freshwater biological samples of fungi, mosses, plants, amphibians, reptiles, insects, molluscs, and earthworms were collected at each site. These samples satisfied at least one of the following conditions: (i) widely distributed species; (ii) rare species; (iii) expected to accumulate radiocaesium; (iv) included in the reference animals and plants (RAPs) proposed by the International Commission on Radiation Protection (ICRP, 2008); and (v) expected to be radiosensitive. The samples were collected during the period from 27 April 2011 to 31 March 2012. Dimensions (length, width, and height) and mass of most animal samples were measured, while those of the other samples failed to be measured. The samples were stored at −20 or −80 °C until their radioactivity was measured. Because the sampling time was limited, especially in the exclusion zone, sample sizes were small (n = 1–4), and upland soil samples were not collected.

At each sampling point, ambient dose rates at a height of 1 m above the ground were measured with a NaI(Tl) scintillation survey meter (Hitachi Aloka Medical TCS-161, Tokyo, Japan) to check radiation levels and estimate external dose rates to wildlife. The measured ambient dose rates was H*(10), the dose equivalent at a depth of 10 mm inside the ICRU sphere (tissue equivalent sphere with a diameter of 300 mm to simulate the human body), that is at the place of an assumed inner organ (IAEA, 2000). The measurements of H*(10) are shown in Table 1.

Samples of water and the top layers of bottom sediment were collected from the same ponds or streams from which the freshwater animals were collected. One water sample and one sediment sample were collected from each aquatic system. The sample size was limited because the aquatic habitats were small (several meters wide) and the sampling time was limited. After the fresh weight of the collected sediment samples was measured, the samples were dried at 60 °C and weighed. Percentage dry weight values of the sediment samples were calculated by dividing the dry weight by the fresh weight.

2.2. Determination of activity concentrations

All the collected organisms were not washed before sample preparation for radioactivity measurement.

The specific organ distribution of radionuclides was investigated in selected organisms. Leaves, stalks, vines, berries, or tubers were isolated from some of the plant samples. Snails and bivalves were dissected, and soft tissue, which included gastrointestinal (GI) tracts and their contents, and shells were isolated for separate analysis. A Japanese striped snake *Elaphe quadrivirgata* was dissected, and samples of GI tracts, which included their contents, and muscle were isolated for separate analysis. For the other organism samples, entire bodies, from which the contents of the GI tracts were not cleared, were used for radioactivity measurement.

Shells isolated from the bivalves *Margaritifera laevis* and *Inversiunio jokohamensis* were milled with a grinder, and the milled shells were transferred to polystyrene bottles (U8 bottles; Yamayu Plastic Medical Products, Osaka, Japan; 56 mm in diameter × 68 mm in height) for radioactivity measurement. The other biological samples (entire bodies of the non-dissected organisms or isolated individual organs other than shells) were ashed using a method adopted by MEXT (1982). Because the volumes of the resulting ash samples were small, the ash was evenly fixed in agarose gel to calibrate the gamma-ray counting efficiencies of the Ge semiconductor detectors using a certified volume standard source (see below for details). The ash samples were transferred to U8 bottles containing 1% (w/v) agarose powder in distilled water. The U8 bottles were heated in a microwave oven to dissolve the agarose and the contents were mixed well by swirling. They were placed on ice to set agarose gels in which the ash was evenly fixed.

Dried sediment samples were milled with a grinder. Water