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Artificial radionuclides in surface air in Finland following the Fukushima Dai-ichi nuclear power plant accident

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ABSTRACT

We present observations of radionuclides released during the Fukushima Dai-ichi nuclear power plant accident in ambient air and in deposition made in Finland during March–May 2011. The first observed fission product was ^{131}I, which arrived in Finland 8–9 days after the accident. Detections of ^{137}Cs and ^{134}Cs were made 2–3 days after the first ^{131}I observations. The highest concentrations of fission products in Finland were observed during March 31st and April 1st. The highest observed concentrations of the following isotopes were: ^{131}I (10.6 ± 0.4 mBq/m^3), ^{134}Cs (0.397 ± 0.020 mBq/m^3), ^{137}Cs (0.405 ± 0.017 mBq/m^3), ^{136}Cs (28 ± 2 μBq/m^3), ^{129}Te (129 ± 9 μBq/m^3), ^{129m}Te (234 ± 20 μBq/m^3), ^{132}Te (51 ± 3 μBq/m^3) and ^{132}I (54 ± 3 μBq/m^3). Generally, higher concentrations of fission product were observed in Southern Finland than in Northern Finland. The variations in the ^{137}Cs and ^{134}Cs activity concentration data suggest that three separate plumes passed over Finland with decreasing concentrations. The first plume, with highest cesium concentrations, passed over Finland during March 31st – April 2nd, the second plume during April 4th – 6th and the third and smallest one during April 10th – April 11th. Both aerosol and gaseous iodine fractions were sampled simultaneously and thus an accurate view of the behaviour of aerosol and gaseous fractions was obtained. Large variations between different fractions were observed with the gaseous fraction representing 65–98% of the total ^{131}I. The ^{134}Cs/^{137}Cs ratio was determined to be 0.99 ± 0.10, which indicates a fuel burnup of approximately 30 MWd/t. The ^{136}Cs/^{137}Cs and ^{129m}Te/^{132}Te ratios were used to estimate the time lapse after the accident. The differences between true time lapse and the ones deduced from the isotope ratios were from the correct time lapse to 0–3 days for ^{136}Cs/^{137}Cs and 5 days for ^{129m}Te/^{132}Te, respectively. Radionuclides from the Fukuhisma Dai-ichi nuclear power plant were also observed in deposition samples. In Norther Finland, the total deposition of 0.28–0.62 Bq/m^2 for ^{137}Cs and 0.21–0.57 Bq/m^2 for ^{134}Cs was determined during March–May 2011. For ^{131}I the deposition of 8.5 ± 2.9 Bq/m^2 was determined at Rovaniemi from the samples from the sample collected during April 1st – 12th.

1. Introduction

The Great East Japan Earthquake on March 11th, 2011 at magnitude 9 generated a series of large tsunami waves that struck the east coast of Japan, the highest being 38 m at Aneyoshi, Miyako (EERI, 2011). The tsunami waves hit the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) causing a loss of backup electric generation. The FDNPP had executed an emergency shutdown during the earthquake, but without cooling, the reactor cores overheated due to the decay heat in the nuclear fuel. Radioactive emissions into the atmosphere from the damaged reactors of the FDNPP started on March 12th. Despite the rescue efforts, large quantities of radioactive nuclides were emitted into the atmosphere and sea. The estimates of the released activity vary. According to the IAEA June 2012 Fukushima Dai-ichi status report, approximately 150 PBq of ^{131}I and 8.2 PBq of ^{137}Cs were released into the atmosphere (IAEA, 2012). Of the total radioactivity released, about 20% came from Unit 1, 40% from Unit 2 (peak on March 15th) and 40% from Unit 3 (peak on march 16th) (Thakur et al., 2013). The radioactive release was transported across the Pacific to North America (Bowyer et al., 2011; Diaz et al., 2011), to Europe (Masson et al., 2011) and to Central Asia (Bolsunovsky and Dementyev, 2011). By day 15 after the initial releases of radioactivity, traces fission products from FDNPP were detectable all across the Northern hemisphere (Thakur et al., 2013). Among the various radionuclides released in large amounts, iodine-131 (^{131}I; $T_{1/2}$ = 8.0 d), cesium-134 (^{134}Cs; $T_{1/}$

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$_2$ = 2.1 yr) and cesium-137 (^{137}Cs; $T_{1/2}$ = 30.1 yr) were easily detectable and of major interest for health impact assessments. Other short-lived radionuclides including tellurium-132 (^{132}Te; $T_{1/2}$ = 3.2 d), tellurium-129 (^{129}Te; $T_{1/2}$ = 1.2 h), tellurium-129 in meta-stable state (^{129m}Te; $T_{1/2}$ = 33.6 d), iodine- 132 (^{132}I; $T_{1/2}$ = 2.3 h), and cesium-136 (^{136}Cs; $T_{1/2}$ = 13.16 d) were detected at trace levels. Outside Japan, the released radioactivity did not pose a health risk. In Finland, surface air activity levels were 4–5 orders of magnitude lower than the activity levels encountered in Finland after the Chernobyl accident in April–May 1986 (Paatero, 2010).

In Finland, arrangements for a countrywide radioactivity monitoring programme were initiated in 1956 by the Finnish Meteorological Office (now the Finnish Meteorological Institute), the Defence Forces, the Finnish Marine Research Institute, and the University of Helsinki. The Radiation and Nuclear Safety Authority (STUK) was founded in 1958. The first radiation monitoring programmes in Finland started in the late 1950s. (Koivukoski, 2013 and references therein). The monitoring of airborne radioactivity in Finland started in 1959 with the Finnish Meteorological Institute's ambient air total-beta measurements, and the aerosol filter sampling started a year later in 1960. STUK's current airborne radioactivity monitoring network was built in steps after the Chernobyl accident. From radiation monitoring and data gathering points of view, the situation in 2011 was completely different compared to

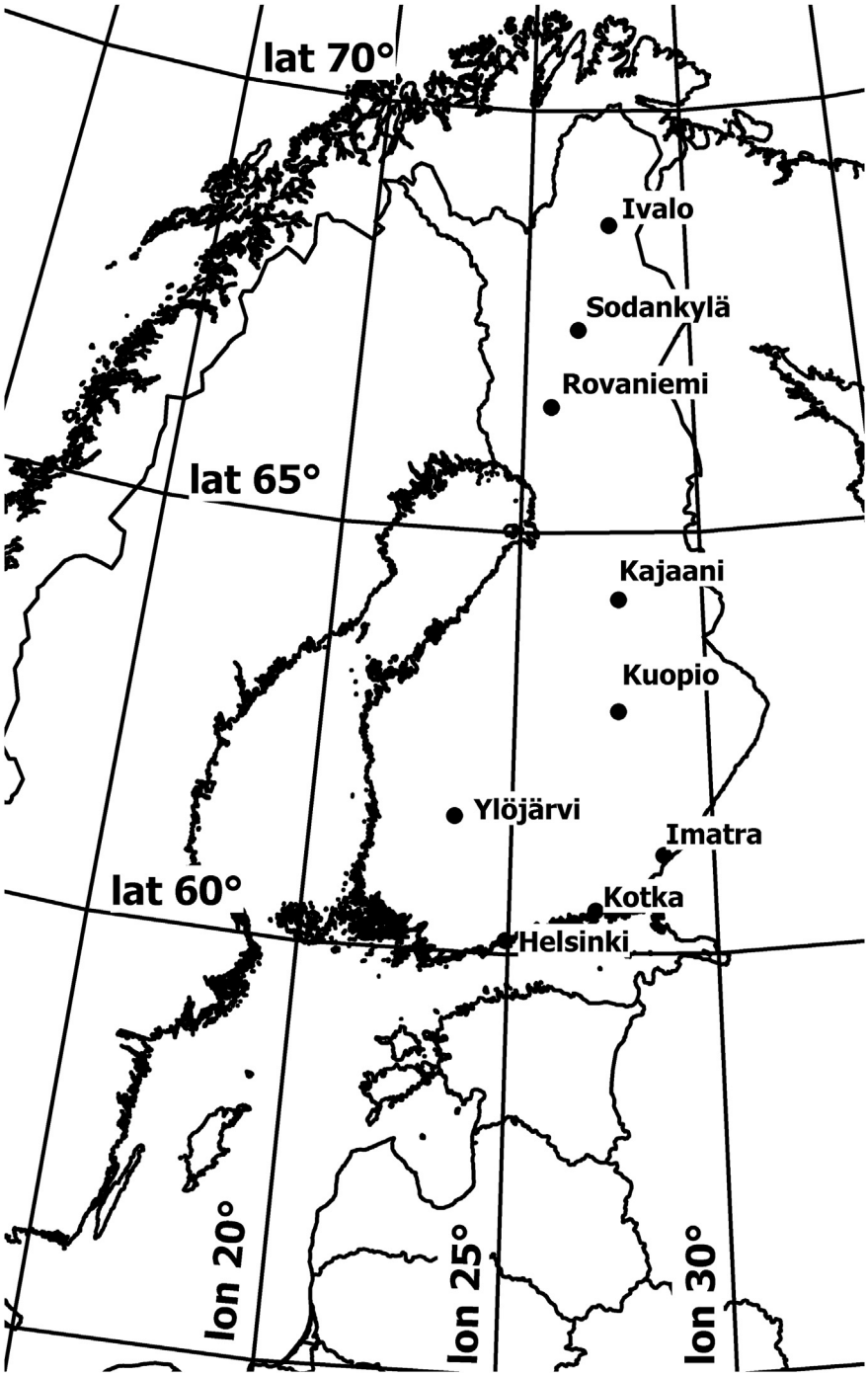

Fig. 1. Locations of the Finnish airborne radioactivity and deposition sampling stations.