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Radionuclides from the Fukushima accident in the air over Lithuania: measurement and modelling approaches

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ABSTRACT

Analyses of ^{131}I, ^{137}Cs and ^{134}Cs in airborne aerosols were carried out in daily samples in Vilnius, Lithuania after the Fukushima accident during the period of March–April, 2011. The activity concentrations of ^{131}I and ^{137}Cs ranged from 12 μBq/m^3 and 1.4 μBq/m^3 to 3700 μBq/m^3 and 1040 μBq/m^3, respectively. The activity concentration of ^{239,240}Pu in one aerosol sample collected from 23 March to 15 April, 2011 was found to be 44.5 nBq/m^3. The two maxima found in radionuclide concentrations were related to complicated long-range air mass transport from Japan across the Pacific, the North America and the Atlantic Ocean to Central Europe as indicated by modelling. HYSPLIT backward trajectories and meteorological data were applied for interpretation of activity variations of measured radionuclides observed at the site of investigation. ^{7}Be and ^{212}Pb activity concentrations and their ratios were used as tracers of vertical transport of air masses. Fukushima data were compared with the data obtained during the Chernobyl accident and in the post Chernobyl period. The activity concentrations of ^{131}I and ^{137}Cs were found to be by 4 orders of magnitude lower as compared to the Chernobyl accident. The activity ratio of ^{134}Cs/^{137}Cs was around 1 with small variations only. The activity ratio of ^{238}Pu/^{239,240}Pu in the aerosol sample was 1.2, indicating a presence of the spent fuel of different origin than that of the Chernobyl accident.

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1. Introduction

On March 11, 2011 a strong earthquake followed by high tsunami and fires damaged three reactors and a fuel pond at the Fukushima Dai-ichi Nuclear Power Plant (NPP) in Japan with releases of radionuclides to the atmosphere and the sea. According to the NISA (Nuclear and Industrial Safety Agency) report from 1.3×10^{17} Bq to 1.5×10^{17} Bq of ^{131}I and about 6.1×10^{15} to 1.3×10^{16} Bq of ^{137}Cs were released to the atmosphere (NISA, 2011; Chino et al., 2011). The consequences of this accident at the beginning estimated as level 4 were raised to the maximum level of 7 on the INES (International Nuclear and Radiological Event Scale) scale (IAEA, 2011), although the amount of discharged radioactive materials comprised approximately 10% of the Chernobyl accident only. Measurements carried out at Tokushima (about 700 km southwest from the Fukushima NPP) indicated the maximum activity concentration of particulate ^{131}I in the air of ~3 mBq/m^3 which was observed on 6 April (Fushimi et al., 2011).

Worldwide monitoring activities started immediately after the announcement of large radionuclide releases from the Fukushima NPP. The particulate ^{131}I activities of 4.4 mBq/m^3 were detected on 19–21 of March in Seattle (USA) (Diaz Leon et al., 2011). According to the CTBTO (Comprehensive Test-Ben Treaty Organization) data the first signs of diluted airborne activities appeared over Europe after 12 days of the Fukushima accident (Wotawa, 2011). The elevated levels of radionuclides on aerosols derived from the Fukushima NPP were detected at several sampling stations in Spain (Lozano et al., 2011), Germany (Pittauerová et al., 2011), Greece (Manolopoulou et al., 2011), Russia (Bolsunovsky and Dementyev, 2011). The most comprehensive radionuclide data over the Europe has been compiled by Masson et al. (2011).

Anthropogenic radionuclides were introduced into the terrestrial and marine environments primarily after the atmospheric nuclear weapon tests carried out by the United States and the former Soviet Union from the 1940s to the early 1960s (Livingston and Povinec, 2002). Another source of anthropogenic radionuclides is related to nuclear accidents. The most severe of them was the Chernobyl accident when among other radionuclides about 1760 PBq of ^{131}I, 47 PBq of ^{134}Cs and 85 PBq of ^{137}Cs were released into the environment (IAEA, 2006). The consequences of the

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Chernobyl accident on the environment and human health were estimated as the worst in the nuclear accident history by its rating to the highest level 7 on the INES scale. Until 12th March, 2011 the second largest accident was the Kyshtym accident which occurred on 29th September, 1957, when due to problems in a cooling system and followed explosion about 7.4×10^5 TBq of radioactive materials were released into the environment. As a result of this accident, more than 10,000 people received significant radiation doses (Hu et al., 2010).

The aim of the present study has been to estimate activity concentrations of Fukushima airborne radioactive aerosols over Lithuania with special emphasis on particle transport from Fukushima to Europe (preliminary results were published by Lujanienė et al., 2011), and to compare the obtained results with data gathered during the investigations of the Chernobyl accident.

2. Samples and methods

2.1. Sampling

The ground level air samples were collected in a forested area on the outskirts of Vilnius (54°42′N, 25°30′E). Perchlorvinyl filters FPP-15 (~1 m^2 surface) were exposed in a special building with blind walls at the height of 1 m above the ground. High volume samplers with flow rates from 2400 m^3/h to about 6000 m^3/h were used. The sampling was carried out continuously. ^{131}I, ^{137}Cs and ^{134}Cs were measured by gamma-ray spectrometry using a HPGe detector (relative efficiency of 42%, resolution of 1.9 keV at 1.33Mev). The precision of ^{137}Cs measurements by gamma-spectrometry was better than ±7% at 2σ level.

The radiochemical analyses of Am and Pu were performed on monthly samples (total volume ~2.0×10^6 m^3) of aerosol ashes (about 30 g), which were dissolved in strong acids (HNO_3, HCl, HF and $HClO_4$). The TOPO/cyclohexane extraction and radiochemical purification using UTEVA, TRU and TEVA resins (100–150 μm) were used for separation of Am and Pu isotopes. ^{242}Pu and ^{243}Am were used as yield tracers in the separation procedure (Lujanienė et al., 2006). The alpha-spectrometry measurements of Pu and Am isotopes deposited on stainless-steel discs were carried out with the Alphaquattro (Silena) spectrometer. Accuracy and precision of analysis were tested using reference materials IAEA-135, NIST SRM No 4350B and 4357, as well as in intercomparison exercises, organized by the Risø National Laboratory (Denmark), and the National Physical Laboratory (UK). The precision of Pu and Am measurements was better than ±8% and ±10%, respectively (at 2σ level).

2.2. Modelling

The transport of radionuclides was simulated using a Lagrangian particle model which calculates trajectories of particles that follow the instantaneous flow in the particle position (Závodský, 2011). The output particle velocity is a sum of deterministic velocity and semi-random stochastic velocity, generated by the Monte Carlo technique. The probability density function for the random component, which simulates the atmospheric turbulence, is dependent on the state of the atmospheric boundary layer. The model also takes into account the radioactive decay of particles (e.g. in the case of ^{131}I), as well as their scavenging by dry and wet deposition. For the meteorological input, the Integrated Monitoring System – IMS Model Suite Lagrangian dispersion model (MicroStep-MIS, 2011) has been used. It calculates the spreading of radioactive materials with special regard to changes in atmospheric conditions, especially changes in wind direction. The meteorological input for the dispersion model was a time sequence analysis of atmospheric state in GRIB format (WMO, 2009). The GFS global weather model was used in simulation time span from 12 March to 27 March, 2011. The 3D wind (u, v, vertical velocity) at upper air model levels was needed to simulate dispersion due to large-scale winds.

A characterization of radionuclide activities with respect to categorized air mass backward trajectories was carried out for estimation of potential location of the radioactivity source. Air mass backward trajectories were generated using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT4) model (Rolph, 2011) with the Final Analyses (FNL, year 2011) and the Global Data Assimilation System (GDAS) meteorological databases at the NOAA Air Resources Laboratory server (Rolph, 2011).

3. Results and discussion

3.1. Modelling of the Fukushima plume

For the assessment of contamination after the accident and prediction of radioactive particle transport the Lagrangian modelling was applied. In order to describe the atmospheric processes realistically, the vertical velocity, particle dissipation and turbulence during the particle trajectory were considered. A single release of 10^{15} Bq of ^{137}Cs, which occurred on March 12, 2011 from damaged Fukushima NPP was analyzed. The initial plume height, as a result of initial vertical velocity and buoyancy, was kept to be at 2000–3000 m. The meteorological data and simulated trajectories revealed that the arrival times of particles released on 11 March, 2011 and 12 March, 2011 were different, and the particles were transported at different altitudes. It was also obvious that the jet stream affected the transport of emitted particles at upper atmospheric levels. Examples of the trajectories simulated using the Lagrangian dispersion model show (Fig. 1) that the first signs of Fukushima released radionuclides could be detected in the European countries (e.g. Island) on 20 March, 2011. The performed simulation indicated that particles released on 11 March, 2011 mainly appeared over Europe on 850 hPa on 13 April, 2011, at 700 hPa on 30 March, 2011 and at 500 hPa on 20 March, 2011. Similar situation was observed for particles released on 12 March, 2011 which arrived to Europe at 700 hPa on 1 April, 2011, at 500 hPa on 21 March, 2011, and the particles at 850 hPa did not reach the European territory. The particle arriving times are in a reasonable agreement with experimental radionuclide data obtained for Vilnius (Lithuania), as discussed later.

3.2. Radionuclide data

The time course of ^{131}I (aerosol component) and ^{137}Cs concentrations measured in March–April of 2011 in Vilnius is shown in Fig. 2, compared with the course of the cosmogenic ^{7}Be. The activity concentrations of ^{131}I and ^{137}Cs ranged from 2 to 3800 μBq/m^3 and from 0.2 to 1070 μBq/m^3, respectively. The first traces of ^{131}I in aerosol filters in Vilnius were found on 23 March. A considerable increase in the ^{131}I activity concentrations (up to about 2.4 mBq/m^3) was observed during the period of 28 March–1 April. The second maximum was detected on 3–4 April, when up to 3.7 mBq/m^3 of ^{131}I was measured in the atmosphere. Activities of ^{137}Cs in aerosol during this period increased up to 0.5 mBq/m^3 and 1.0 mBq/m^3, respectively. In addition to ^{131}I and ^{137}Cs, traces of other radionuclides were detected in the aerosol filters as well. Their concentrations in the most active sample collected on 3–4 April 2011 14:00–06:50 UTC were: ^{132}I – 0.12 ± 0.01 mBq/m^3, ^{132}Te – 0.13 ± 0.01 mBq/m^3, ^{129}Te – 0.40 ± 0.04 mBq/m^3, ^{129m}Te–0.75 ± 0.25 mBq/m^3 and ^{136}Cs – 0.080 ± 0.008′mBq/m^3.

The variations in the activity concentrations of studied radionuclides can be due to the different arrival time of radioactive particles predicted by the Lagrangian modelling. The particles

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