Transport of aerosol particles from the Fukushima accident

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Measurements of activity concentrations of ^{131}I; ^{129m}Te, ^{132}Te, ^{134}Cs; ^{136}Cs, ^{137}Cs in aerosol were carried out in daily samples after the Fukushima accident during the period of March – April, 2011 in Vilnius.

The ground level air samples were collected on the outskirts of Vilnius (54°42'N, 25°30'E). FPP-15 filters made of chlorinated polyvinyl chloride (~ 1 m^2 surface) were exposed at the height of 1 m above the ground. High volume samplers with flow rates (2400 m^3 h^{-1} - 6000 m^3 h^{-1}) were operated. The sampling was carried out continuously. The activity concentrations of gamma-emitters were measured in 12 or 24 h samples by gamma-ray spectrometry using a HPGe detector (relative efficiency of 42 %, resolution of 1.9 keV at 1.33 MeV). The combined uncertainty of measurements by gamma-spectrometry was better than ± 7 % (k = 2).

The radiochemical analyses of Am and Pu were performed on monthly samples (total volume ~ 2.0x10^{6}m^3) of aerosol ashes (~ 30 g). The applied radioanalytical procedures are described in detail by Lujanienė (2011). The 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 (k = 2).

Three-dimensional 13-day backward trajectories of air masses that reached Vilnius at 850 hPa and 700 hPa height were calculated employing the Hybrid Single-Particle Lagrangian Integrated Trajectory (Draxler & Rolph, 2011). A meso-scale chemical transport model system (EURAD, http://www.eurad.uni-koeln.de) was used to evaluate the wet and dry deposition of particulate ^{137}Cs, and a potential dispersion of the radioactive cloud after a nuclear accident for three levels

The measured activity concentrations at the site of investigation resulted from a long-range air mass transport and arrival time following downward air mass transport and meteorology (Lujanienė et al., 2012). The activity concentrations of ^{131}I and ^{137}Cs ranged from 2 μBq m^{-3} to 3800 μBq m^{-3} and from 0.2 μBq m^{-3} to 1070 μBq m^{-3}, respectively. In addition to ^{131}I, ^{134}Cs and ^{137}Cs ^{132}I, ^{132}Te, ^{129}Te, ^{129m}Te and ^{136}Cs were detected in the aerosol filters as well. Variations in activity ratios of ^{131}I/^{137}Cs, ^{132}Te/^{129m}Te, ^{134}Cs/^{137}Cs and ^{137}Cs/^{136}Cs were observed. They were mainly due to the decay of the short-lived isotopes (e.g. ^{129m}Te, ^{131}I, ^{136}Cs) while an increase in ^{134}Cs/^{137}Cs was explained by the presence of the Chernobyl-derived ^{137}Cs in the environment. An increase in the activity ratio was observed whereas the highest activity ratio (1 ± 0.05) was found in the most active sample collected on 4 April. Contrary to the ^{134}Cs/^{137}Cs ratio the maxima of ^{131}I/^{137}Cs ratios did not coincide with ^{137}Cs or ^{131}I activities. It was found that different behavior of highly volatile ^{131}I and ^{137}Cs resulted in enrichment of ground level aerosol particles by ^{131}I with respect to ^{137}Cs. Simulated activity concentrations of ^{137}Cs attached to aerosol particles for the Vilnius site reasonably agreed with measured activities in aerosol samples during the Fukushima accident (Lujanienė et al., 2013).

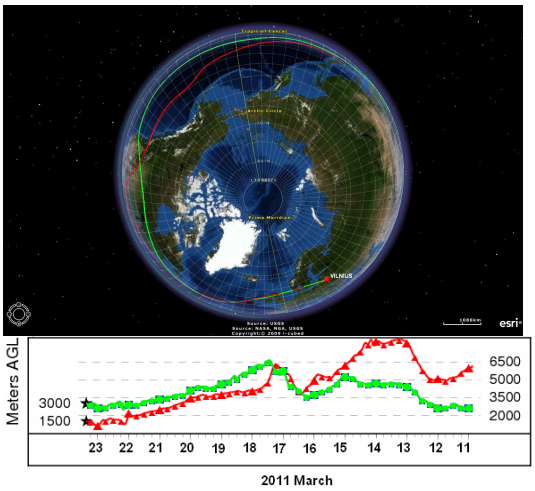

Figure 1. Backward trajectories of air-mass transport ending on 23 March, 2011 in Vilnius.

The activity concentration of actinides measured in the integrated sample collected in March–April, 2011 showed a small contribution of Pu with unusual activity and atom ratios indicating the presence of the spent fuel of different origin than that of the Chernobyl accident (Lujanienė et al., 2012).

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