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Case studies in national experiences

Assessment of environmental radiation monitoring data in Hungary following the Fukushima accident

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ABSTRACT **The unusually strong earthquake in Japan on 11 March 2011 and the following extreme tsunami caused enormous damage in the buildings of Fukushima Dai-ichi Nuclear Power Plant (NPP) situated on the Pacific coastline of Japan. The accident led to the release of a large amount of radioactive material into the environment. According to the measurements of the Radiological Monitoring and Data Acquisitions Network (RAMDAN) the radioactive plume reached Hungary on 24 March 2011. The main volatile fission products – ^{131}I, ^{134}Cs, and ^{137}Cs radioisotopes – were measurable in aerosol and fallout samples in Hungary. Their activity concentration in air reached the maximum value in the last days of March and returned to the background level in the first half of May. As a consequence of respiration of contaminated air, a maximum of 1 Bq per capita of ^{131}I could be accumulated in the thyroid gland of the Hungarian population during the given period. The calculated upper limits of the committed effective dose from inhalation of ^{131}I were 4 nSv and 10 nSv to the Hungarian adults and infants, respectively. These values are a hundred thousand times less than the annual radiation dose from natural sources to the Hungarian population. The radiation dose from radioactive caesium isotopes originating from Fukushima was even less, around 1 nSv on average, to Hungarian residents. No health deterioration can be expected from this radiation burden.**

Keywords: Environmental monitoring / radiological emergency / nuclear accident

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1. Background of the study

Following the accident of the Fukushima Dai-ichi Nuclear Power Plant (NPP) of Japan, the Radiological Monitoring and Data Acquisition Network (RAMDAN) of Hungary expanded the object and frequency of its examinations to check the concentration of the artificial radioisotopes in environmental and food samples. The coordinator of RAMDAN is the National Research Institute for Radiobiology and Radiohygiene (NRIRR) at Budapest. Based on the radiological monitoring data, the NRIRR has made a continuous assessment on the public health effects.

2. Sampling methods

The objects of the sampling within RAMDAN in Hungary were: air samples (aerosols and fallout), fresh harvested grass, green vegetables (leek, spinach, sorrel) and raw milk.

The aerosol samples were taken by a medium volume air sampler (HUNTER, Senya Oy, Finland). The sampling period was 3–8 days (depending on environmental and weather conditions). During these periods (10–28) $\times 10^3$ m^3 air was filtered by a glass fiber filter with 1.6 µm average pore size (Whatman type 934-AH and GF/A). The middle of the sampling period was chosen as reference date.

The activity of the samples was measured by gamma-ray spectrometry (HPGe detectors, Canberra). The uncertainty of the measurements was in the range of 3 up to 20%.

Environmental dose rate was measured three times per day.

3. Results and discussion

In the previous years there was no measurable amount of ^{131}I in aerosol samples taken in Hungary except for Budapest. In Budapest there is an institute which produces radioiodines for medical use and for medical centers which use radioiodines. During the last decade the average activity concentrations of radioiodines (^{125}I and ^{131}I) in aerosol samples measured by NRIRR were about 10 µBq • m^{-3}. In the national sampling network the detection limit for ^{131}I isotope in aerosols is a few µBq • m^{-3} (Homoki *et al.*, 2011).

According to measurements of RAMDAN, 12 days after the first explosion in the Fukushima Dai-ichi NPP, the amount of ^{131}I in aerosols increased and became measurable. Results of the air monitoring (Fig. 1.) showed that the radioactive plume reached the territory of Hungary on 24 March 2011. The maximum value of its activity concentration in aerosols was detected by four countryside radiation monitoring laboratories in the last days of March (except Budapest, as discussed later) and it fell below the minimum detectable activity in the middle of May.

Before the Fukushima NPP accident only traces of ^{137}Cs originating from the resuspension of the fallout of Chernobyl NPP accident and of earlier nuclear weapon tests was measurable in aerosols.

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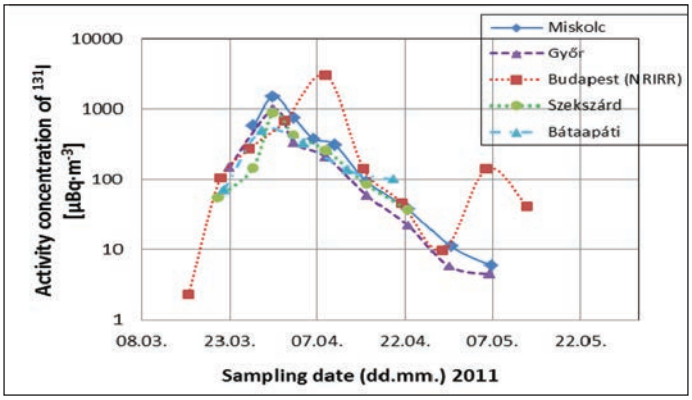

Figure 1 – ***Time series of activity concentration of ^{131}I in aerosol samples at five different locations in Hungary.***

However, a few days after the first appearance of ^{131}I in the air, both ^{134}Cs and ^{137}Cs were detected. The values of their activity concentration reached the maximum at the same time as the ^{131}I isotope at the end of March and then decreased continuously until the middle of May (Homoki *et al.*, 2011). There was no difference in the changes in activity concentrations of ^{134}Cs and ^{137}Cs within the national sampling network. Our results are within the range of values measured in Europe (IRSN, 2011; Masson, 2011).

Maximum activity concentrations of ^{131}I were about one order of magnitude higher than the maximum level of caesium isotopes. It is important to mention that the activity concentrations of natural isotope ^{7}Be are much higher than that of the Fukushima originating volatile fission products (^{131}I, ^{134}Cs and ^{137}Cs) as it was measured in the north-eastern part of Hungary, in Miskolc (Fig. 2).

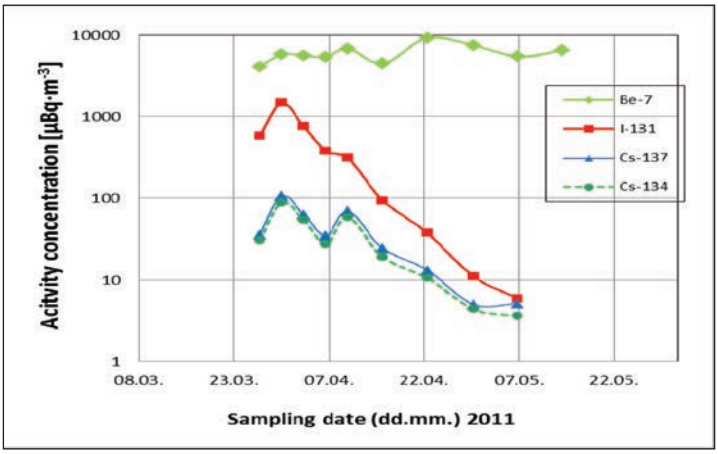

Figure 2 – ***Time series of activity concentration of ^{7}Be, ^{131}I, ^{134}Cs and ^{137}Cs in aerosols collected at Miskolc.***

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In all sampling stations (except Budapest) the values of the activity concentrations of ^{131}I and both caesium isotopes showed similar maximum concentrations and kinetics. This fact refers that these volatile artificial isotopes came simultaneously within a radioactive cloud. We concluded that the radioisotopes (^{131}I, ^{134}Cs and ^{137}Cs) measured by RAMDAN were originated from the Fukushima NPP (Homoki *et al.*, 2011).

Comparing the results of the ^{131}I measurements at Budapest (Fig. 3) with the results of any other countryside sampling station (see Figs. 1 and 2), a difference can be observed in the duration of the detection period and the levels of the maximum values of activity concentrations.

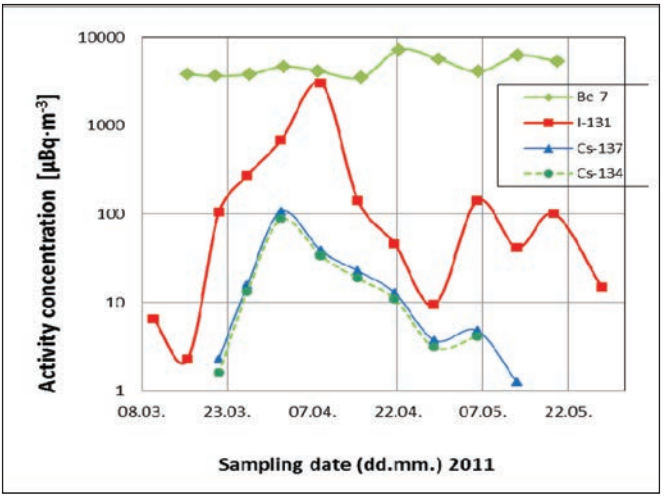

Figure 3 – ***Time series of activity concentration of ^{7}Be, ^{131}I, ^{134}Cs and ^{137}Cs in aerosols collected at Budapest.***

At the sampling station in Budapest both ^{125}I and ^{131}I radioiodine isotopes had been regularly detected in a minor quantity. The average activity concentration of ^{131}I was 10.3 µBq·m^{-3} (SD: 13.7 µBq·m^{-3}) for the period of April of 2003 to March of 2011 and the minimum and maximum values were 0.8 and 96 µBq·m^{-3}, respectively. The average activity concentration of ^{125}I was 8.5 µBq·m^{-3} (SD: 9.6 µBq·m^{-3}) for the period of March of 2008 to March of 2011. The minimum and maximum values were 1.3 and 33.6 µBq·m^{-3}, respectively.

The increase of the ^{125}I activity concentration in spring of 2011 at Budapest suggests that it could be also an increase in ^{131}I activity concentration without being in connection with the nuclear accident. As ^{125}I is not a primary fission product it could not be discharged by the Fukushima NPP.

After the Fukushima accident the activity concentration of ^{131}I reached the maximum value one week later than the caesium isotopes and was significantly higher than the nationwide levels of radioiodine in aerosols (see Figs. 1 and 3).

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To further evaluate the data, we compared the changes of activity concentration of ^{125}I, ^{131}I and ^{137}Cs measured by NRIRR for the period of January to June in 2011 (see Fig. 4). The results show that the activity concentration of ^{125}I increased by about one order of magnitude at the beginning of April. The activity concentration remained at this level until the end of June, when it fell to the earlier level (about 10 μBq·m^{-3}). The activity concentration of ^{131}I continuously decreased during April and at the end of April its level returned to the earlier detected low level. But after that its level increased slightly again two times during May. The countryside laboratories could not detect radioiodine in the air at this time. The ^{137}Cs was not detected after the middle of May at any sampling station.

These observations suggest that part of the ^{131}I measured at Budapest has another origin, other than Fukushima (Homoki *et al.*, 2011).

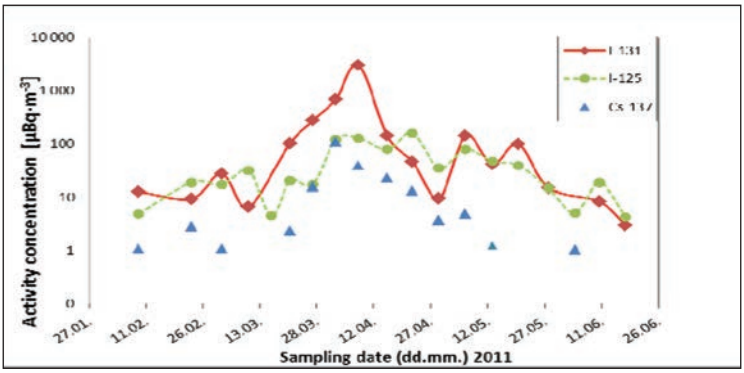

Figure 4 – ***Time series of activity concentrations of ^{137}Cs, ^{125}I and ^{131}I in aerosols at Budapest.***

The supposed second emission source should have had impact on the activity concentration of ^{131}I in air of the whole country, therefore we analysed the decrease of ^{131}I and ^{137}Cs activity concentrations measured in aerosols collected in the countryside. We calculated the "effective half-live" for ^{137}Cs and ^{131}I activity concentrations and we find 8.1 days for the ^{131}I and 4.8 days for the ^{137}Cs respectively.

The decrease rate of ^{137}Cs activity concentration provides information about the migration velocity of the contaminated cloud since the radioactive decay of ^{137}Cs (T½ = 30.2 years) is negligible during the analyzed period. The decrease of ^{131}I activity concentration is given by the contaminated cloud migration velocity and by the radioactive decay (T½ = 8.02 days). These two effects result the "expected effective half-life" for ^{131}I to be 4.0 days. This value is lower than the "effective half-life" which describes the migration velocity calculated from ^{137}Cs measurements (4.8 days). Consequently, the ^{131}I activity concentration decreased slower than expected in collected aerosols.

In another approach we calculated the "effective half-life" of the ^{131}I/^{137}Cs rate decrease, and we found 10.5 days. We assumed that the influence of precipitation (dry and wet) was the same for the ^{137}Cs and ^{131}I. This value is higher than the

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half-life of ^{131}I (T½ = 8.02 days), which fact also shows that the activity concentration of ^{131}I decreased more slowly than expected. The explanation may be that another radioactive ^{131}I wave reached the air of the country, which was not enough significant to cause a visible peak in the time series of ^{131}I activity concentration measured by countryside laboratories.

In the monthly fallout samples the ^{134}Cs and ^{137}Cs were detected in amounts of less than 0.2 Bq·m^{-2}·30 day^{-1}. The activity concentration of ^{131}I in rainwater did not exceed 1.1 Bq·l^{-1}. In the fresh harvested grass (which gives information about the dry deposition) and in green vegetable and milk samples only the ^{131}I isotope was measurable in amounts up to 3 Bq·kg^{-1}.

The dose rate measurements did not show any changes compared to earlier measurements' results.

4. Doses to populations

As a consequence of inhalation of contaminated air, a maximum of 1 Bq per capita of ^{131}I could be accumulated in the thyroid gland of the Hungarian population from the end of March to the middle of May 2011. This value is a hundred times less than the average ^{131}I uptake by the Hungarian population following the Chernobyl accident.

The dose of the thyroid from this estimated uptake of ^{131}I have not exceeded the value of 4 μSv dose equivalent for infants, 1 μSv for children, nor 0.4 μSv for adult persons. These minor doses are a million times smaller than the doses caused by the activity of radioiodines used in nuclear medicine for diagnostic purposes of the thyroid gland, hence, they cannot have health damaging effects.

The calculated upper limit of the committed effective dose from inhalation of ^{131}I were 4 and 10 nSv for the residents of the Hungarian countryside and were 10 and 30 nSv for those who reside in Budapest, for adults and infants, respectively. These values are from one hundred thousand to million times less than the annual radiation dose from natural sources of the Hungarian population (ICRP, 1995).

The radiation dose from radioactive caesium isotopes originated from Fukushima was even less, around 1 nSv in average, to Hungarian residents following the accident.

It can be established that this radiation burden cannot cause any health deteriorations.

5. Conclusions

The radioactive plume originating from the Fukushima accident reached Hungary on 24 March of 2011. Until the end of March the activity concentration of the volatile fission products (^{131}I, ^{134}Cs and ^{137}Cs respectively) in aerosol samples increased

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and the polluted air left Hungary one and a half month later, at middle of May. The activity concentrations of these artificial isotopes were much lower than the activity concentrations of natural isotopes, like ^{7}Be.

In fresh harvested grass and in green vegetables only the ^{131}I isotope could be detected in a very little amount as a consequence of its precipitation.

No health damaging effects can be expected from the calculated minor doses.

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