Traces of radioactive $^{131}$I in rainwater and milk samples in Romania

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Abstract
measurements of $^{131}$I ($T_{1/2}=8.04$ days) activities have been performed in the IFIN HH (Horia Hulubei National Institute of Physics and Nuclear Engineering) underground laboratory situated in Unirea salt mine, Slănic-Prahova, Romania. The rainwater samples were collected starting on 27 March from Brașov and Slănic-Prahova. Also sheep and goat milk samples were collected in the Slănic, Brașov and Iași areas and measurements were subsequently made on them. The measurements on the samples were made at the IFIN HH’s underground laboratory in an ultra-low radiation background, using a high resolution gamma-ray spectrometer equipped with a GeHP (hyperpure) detector having a full width at half-maximum of 1.80 keV at 1332.48 keV for the second $^{60}$Co gamma ray and a relative efficiency of 22.8%. The results show a specific activity of $^{131}$I from <0.063 to 0.75 Bq l$^{-1}$ for rain. In the milk samples the specific activity varied from <0.12 to 5.2 Bq l$^{-1}$.

Keywords: Fukushima accident, $^{131}$I in rainwater and milk, underground laboratory, gamma-ray spectrometry

1. Introduction

The Fukushima accident that started on 11 March 2011 caused the release of significant amounts of $^{131}$I, $^{137}$Cs and other radioactive isotopes into the environment. The atmospheric currents spread the radioactive contamination over all of the northern hemisphere. According to meteorological information, the radioactive cloud reached Romanian territory on 25–26 March [1, 2]. The meteorological conditions over Romania were characterized by little rain. A number of samples of rainwater and of sheep and goat milk were taken for analysis.

The measurements on the samples were made in our institute’s underground laboratory at Slănic-Prahova; see figure 1. The laboratory was constructed and put into operation in 2006 [3]. The equivalent depth of the Unirea mine has been estimated from cosmic ray muon measurements to be 610 mwe (metres of water equivalent) [4].

2. Measurements and results

The measurements were performed with a CANBERRA ultra-low GeHP system, equipped with a detector having a relative efficiency of 22.8%, assisted by an INSPECTOR 2000 multichannel analyser and GENIE 2000 software code. The detector is housed in a 10 cm lead and 2 cm copper shield, which ensures a reduction of the background by a factor of 1600 with respect to that for a spectrum collected outdoors at the surface; see figure 2.

The efficiency of the measurement system was determined with IAEA-444 and IAEA-445 reference materials which, first, a soil from China containing a cocktail of $^{109}$Cd, $^{60}$Co, $^{137}$Cs, $^{54}$Mn and $^{65}$Zn radionuclides, and, second, a spiked water sample containing the same radionuclides. The efficiency was determined using the water sample data. The efficiency dependence of the energy is represented in a log–log graph in figure 3.
Samples of rainwater were collected beginning on 27 March and measurements were made with them in a cylindrical plastic box of 75 mm diameter and 35 mm high. The volume of the samples for measurement was 80 cm$^3$. The $^{131}$I line at 364.48 keV has been seen in all collected spectra. This aspect is illustrated for the rainwater sample collected on the morning of 29 March in Slănic in figure 4. The specific activities of $^{131}$I in the rainwater samples are presented in table 1.

Starting from 5 April, sheep and goat milk samples were collected and measurements subsequently made in the same way as for the rainwater samples. The specific activities of $^{131}$I measured in sheep and goat milk are illustrated in table 2.
3. Comments

From the results, one can observe the presence of $^{131}I$ in very small amounts in the precipitation and milk, recorded starting from 27 March 2011 in Brașov, Slănic-Prahova and Iași regions from Romania. The specific activity in rainwater of $^{131}I$ varies from $<0.063$ to $0.75 \text{ Bq l}^{-1}$. For the milk samples the specific activity varies from $<0.12$ to $5.2 \text{ Bq l}^{-1}$. For a collecting time of $82,500 \text{ s}$, the minimum detectable activity for $^{134}\text{Cs}$ was $0.12 \text{ Bq l}^{-1}$ and for $^{137}\text{Cs}$ it was $0.18 \text{ Bq l}^{-1}$. In all samples on which measurements were made, $^{134}\text{Cs}$ and $^{137}\text{Cs}$ were not observed.

We can suppose that the $^{131}I$ originates from Fukushima nuclear accident and has arrived over Romanian territory around 26–27 March, and the radioactive deposition was observed until the middle of April. After 9 April, the $^{131}I$ level was lower than the minimum detectable activity for rain, and after 16 April, for milk.

The measured activities are far below any intervention limits. For instance in Japan the limit for drinking water was set at $300 \text{ Bq l}^{-1}$ for adults and children and $100 \text{ Bq l}^{-1}$ for infants [6]. The values measured by us are 2–3 orders or magnitude lower than these limits. In sheep milk, the highest specific activity of the $^{131}I$ is more than an order of magnitude lower than the limits.

The presence of $^{131}I$ over Romania demonstrates that evidence of the consequences of a nuclear accident could be obtained even at more than $10,000 \text{ km}$ away, which also demonstrates that the radioactive plume originating in Fukushima spread practically over all of the northern hemisphere.

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