GAMMA SPECTROMETRIC ANALYSES OF DIFFERENT SAMPLES FROM CLUJ-NAPOCA, ROMANIA, AFTER THE CHERNOBYL ACCIDENT

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Abstract: After the Chernobyl accident from April 26, 1986, were released into the atmosphere a large variety of fission products. Consequence of the atmosphere contamination and of the wet deposition of the radionuclides occurs into the soil contamination with artificial radionuclides, during May 1986. Romania was one of the most contaminated country from Europe and, therefore, the consequences of this accident still persist in soil and in perennial plants from these territories. After 20 years from the sampling, the sediment from Cluj-Napoca, collected on May 17, 1986, showed a concentration of 203829.8±75 Bq/kg for ^{137}Cs. The smallest caesium activity was found in sand.

Key words: ^{137}Cs, gamma spectrometry, sediment, Chernobyl.

Introduction

Large amounts of radioactivity have been deposited globally due to fallout from the atmospheric nuclear tests conducted by the USA, URSS, UK, France and China from 1945 to 1980. The catastrophy occured on April, 26, 1986 at Chernobyl nuclear power plant released high amounts of radioactive material in environment, from which, caesium, strontium and plutonium were the most important ones, mainly due to their long physical life-time (UNSCEAR 2000). The radioactive cloud passage over Romania, beginning with April 30, 1986 caused fall-out of different intensities in different regions, especially where the cloud passage was accompanied by rainfalls. Romania was one of the most contaminated country from Europe and, therefore, the consequences of this accident still persist in soil and in perennial plants from these territories.

Beginning on the May 1, measurements started in order to identify the radionuclides and also to establish the gamma activity for the environmental impact in Cluj area. The most important radionuclides carried by the radioactive plume over Romania were: ^{132}Te+^{132}I, ^{131}I, ^{140}Ba+^{140}La, ^{103}Ru+^{103}Rh, ^{95}Zr+^{95}Nb, all with relatively short halfe-lives. The radionuclides with an intermediate half-life (as ^{134}Cs, ^{141}Ce, ^{144}Ce, ^{125}Sb and ^{106}Ru+^{106}Rh) were also present in the first years after the accident (Cosma, 2002). After the decay of I-131 and of the other short-lived radionuclides, ^{137}Cs and ^{90}Sr remained the most important contaminants of the Romanian territory. In Romania, the highest deposits were found in Transylvania, for the country center some of these being higher than 80 kBq/m^2 (Sonoc, 1995). A study of the radioactive contamination in different samples collected from Romania, in 1986, is made in the present work. ^{137}Cs was selected to characterize the magnitude of the ground deposition because (i) it is easily measurable, (ii) it was the main contributor to the radiation doses received by the population (once the short-lived ^{131}I had decayed), (iii) being one of the long-lived radionuclides ($T_{1/2}$=30 ani), it still persist in soil and perennial plants.

Materials and methods

The radionuclide composition was measured in a particularly sediment, sampled from a known area of the roof of a building in the Manaştur district from Cluj-Napoca city on May 17, 1986. The soil samples were gathered from undisturbed soils in 1987. The depth of sampling was 15 cm, and in this layer over 95% of ^{137}Cs activity was found. The pollen samples were daily collected between 1-30 May, 1986.

Gamma-spectrometric measurements were carried out using a multichannel system with an ORTEC HP-Ge-detector with an energy resolution of 2.0 keV at 1.33 MeV of ^{60}Co. Materials used in the detector assembly and cryostat was selected for low radioactivity content. The counting time was aproximatively one hour (3600 s) for the most of the samples, but, in some cases, the measurements need more time, until the counting error for ^{137}Cs dropped below 5%. ORTEC Gamma Vision computer program was used to check the peak shape and the radionuclides composition of the samples. Calibration of the experimental setup with primer standards for well-type tube geometry was done initially.

The samples were measured either in cylindrical boxes (100 ml) or in Marinelli Beaker geometry (0.5 l) (used for the water from the roof). At the beginning, all of them were weighed, and then dried at 105 ˚C for 24 hours. Before starting the measurements, the samples were weighed again. The sediments from the roof-water were cleared away by filtration; the water and the filter with the sediments were measured separately.

Results and discussions

In Table 1 is shown the radioisotope composition of the roof-sediment collected during April 30 - May 17, 1986. This sediment is in general similar to other samples from this region. The sample was measured consecutively on May 18 and November 20, 1986, March 4 and July 7, 1988 (Cosma, 2002), respectively March 14, 2006.

The gamma-spectrum recorded with the Gamma Vision program in 14th of March 2006 is shown in Figure 1. As it can be seen, the ^{137}Cs activity is still very high and it remains the main contributor of the total contamination. However, there could be found some traces of ^{125}Sb, ^{134}Cs ^{154}Eu and of course the natural radioisotope ^{40}K, all of this with a long half-life time (more than 100 days). The specific activity of these radionuclides is shown in the last column of Table 1. The error was calculated only for the counting statistics; there were not considered the sample deformation, weighing, the position of the sample on detector, remaining sample inhomogeneity, loss of soil, detector efficiency calibration or others.

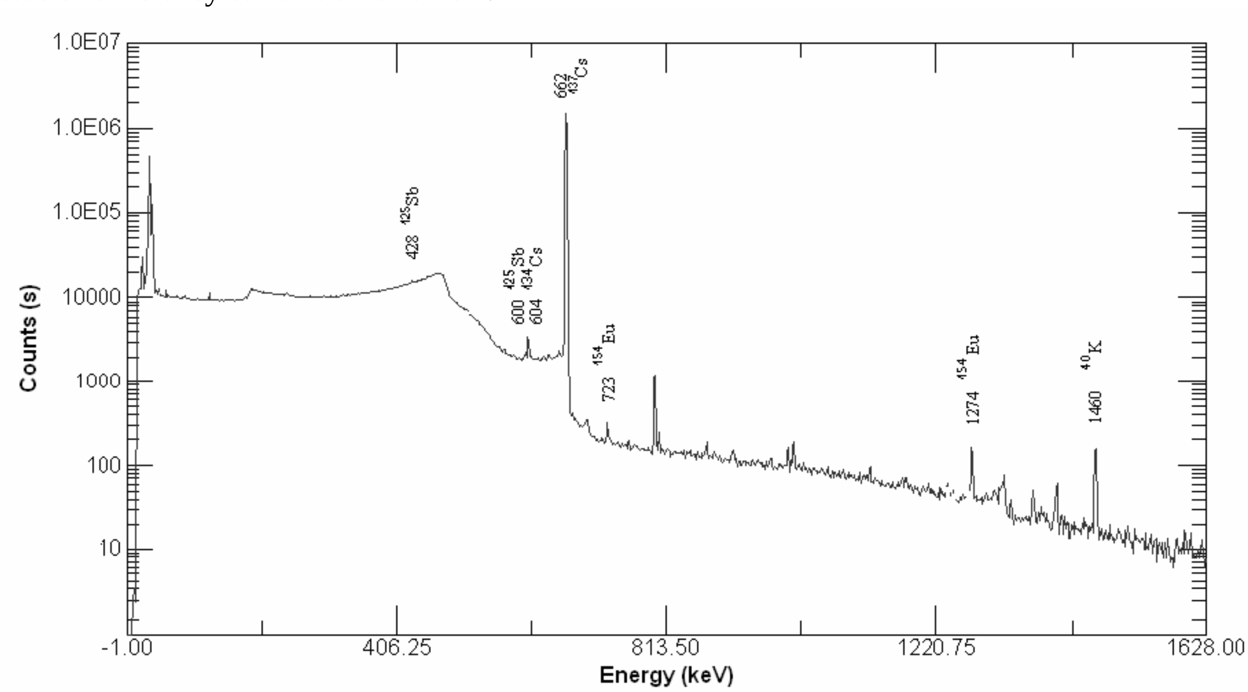

Fig. 1. Gamma-spectrum of the sediment sampled from Cluj-Napoca on May 17, 1986 and registered on March 14, 2006

Although the accident was long time before, its consequences are still present and, unfortunately, ^{137}Cs will persist many years yet.

Table 1
Radionuclides composition of Cluj-Napoca deposit calculated from the spectrum on Figure 1 (March 14, 2006)

Nuclide	**Half-life**	**Specific activity (Bq/kg)**
^{125}Sb	2.758 years	240.3 ± 23
^{134}Cs	2.065 years	163.6 ± 4
^{137}Cs	30.1 years	203829.8±75
^{154}Eu	8.593 years	99.5 ± 5
^{40}K	$1.3 \cdot 10^{9}$ years	357.9 ± 14

Besides the sediment measurements, some other analyses of pollen, soil, roof-water and sediments from the roof-water, street dust and sand samples were made. Except the pollen, which was daily collected during May, 1986 all the samples were collected in august 1986 and measured in June 1988 (Cosma, 2002) and, respectively, in March, 2006. In Table 2, the storage capacity of ^{137}Cs in these samples is presented. All the values were corrected with the date of collection, August 15, 1986 and, respectively, May 7, 1986 for the pollen sample.

Table 2
^{137}Cs activities in samples collected in 1986 and measured 20 years after (the values are corrected with the sampling date)

Sample	**Dry weight (g)**	**^{137}Cs activity (Bq/kg dry weight)**
Roof-water	483.22	85±1.59
Roof-water sediment	2.00	90965±194.25
Roof-sediment	15.00	341332±208.85
Street dust	74.73	6468±32.19
Soil	57.44	3679±26.15
Sand	42.19	5±0.38
Pollen	7.76	9898±146.96

The passage of the radioactive cloud across Cluj-Napoca was accompanied by heavy rains, which caused high, wet deposition of atmospheric radionuclides (Cosma, 2002). When rain occurs, wash-out (entrainment of particles by falling rain drops) and rain-out (formation of rain drops around the particles) of radionuclides from the atmosphere result in a greater contamination of ground surfaces than under dry conditions. Accordingly, in the sediment from the roof water was found a high activity for ^{137}Cs, much higher than in street dust, soil or pollen.

The chemical behaviour of caesium is such that it would tend to be sorbed, much like potassium, by clays, organic matter or other similar materials (Nimis, 1996). The lower value found in sand sample indicate that the sand has a very low cation exchange capacity and there was little organic matter present in the sand to sorb the caesium.

It is believed that the contamination of pollen grains happened during the transport from the gathering place to the beehive. Therefore, the pollen acts like a filter for the radionuclides present in the air. ^{137}Cs activity in the pollen sample was about 9898±146.96 Bq/kg dry weight. This indicates that the pollen might be usable to survey some possible radioactive emissions in the air in close vicinity of nuclear power plants.

Conclusions

The high radioactive contamination of the romanian territory after the Chernobyl accident is confirmed by the values found for caesium in different environmental samples from Cluj-Napoca, even if 20 years passed from the samples collection. The roof-sediment collected on May 17, 1986 showed the highest concentration for caesium. In roof-sediment, traces of ^{125}Sb, ^{134}Cs ^{154}Eu could still be found, after 20 years since the catastrophy, all of this with a long half-life time (more than 100 days). The lowest activity for all the radionuclides is found in sand.

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