STUDIES ON THE ENVIRONMENTAL RADIOACTIVITY LEVEL IN THE AREA OF INFLUENCE OF THE HORIA HULUBEI NATIONAL INSTITUTE FOR R & D IN PHYSICS AND NUCLEAR ENGINEERING

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Abstract. The paper presents the radiological studies in the Horia Hulubei National Institute for R & D in Physics and Nuclear Engineering, IFIN-HH influence area. The environmental samples studied are: drinking water, surface and underground water, aerosols, soil, cultivated and spontaneous vegetation, atmospheric deposition and precipitation. The level of air radioactivity is monitored using the environmental thermoluminescence dosimetry (TLD) system. For water, soil and vegetation are used the alpha-beta global analysis with associated methods. The highest ambient dose rate value of (106.25 ÷ 5.82) nSv/h was measured in the third quarter of the 2012 studied year. All values are comparable with those measured at national level and below the maximum radioactivity allowed by the legislation in force. In 2012 the measured values were: i) 0.02 ÷ 0.05 Bq/L global alpha concentration and 0.40 ÷ 0.60 Bq/L global beta concentration in drinking water samples; ii) 0.35 ÷ 0.72 Bq/L gross beta activity for surface water and groundwater; iii) gross beta activity in aerosols of 0.033 ÷ 0.198 Bq/m3 and gamma global concentration between 0.044 Bq/m3 and 0.077 Bq/m3; iv) 670 ±1020 Bq/kg global beta activity in soil samples; v) between 0.15 Bq/m2/day and 0.75 Bq/m2/day global beta activity for atmospheric deposition.

Key words: Ambient dose equivalent, environment, radioactivity, radionuclide.

1. INTRODUCTION

The results obtained on the representative samples collected in 2012 from the area of influence are presented in order to assess the impact of the nuclear activities carried out in the "Horía Hulubei - National Institute for R & D in Physics and Nuclear Engineering", IFIN-HH. The institute develops the following activities: nuclear research, radioisotope production used in medicine for diagnostic or radiotherapy, multiple irradiation activities using the ⁶⁰Co source with high radioactivity, decommissioning of the VVR-S nuclear reactor for research, national center radioactive waste treatment plant e.g. The specifically activity of the nuclear research institute requires the monitoring and assessment of the environmental radioactivity level in the IFIN-HH influence area by different methods: IFIN-HH Weather Tower that records the absorbed dose in air real on time, radiometric analysis for environmental samples[1].

The studies shown in the paper were made in the Laboratory for Personnel and Environment Dosimetry (LDPM) from IFIN-HH that consists in the following units: i) unit for environmental radioactivity measuring (UMRM); ii) dosimetric survey unit with thermoluminiscence dosimeters (USD-TL) for occupational exposure monitoring; iii) photodosimetric survey unit (USF) only for occupational monitoring and iv) unit for measuring radioactivity environmental and food samples using gamma spectrometry method (UMAP).
UMRM performs the environmental radiological monitoring according with the program for environment radioactivity monitoring around the nuclear and radiological plants from IFIN-HH. The program is approved by the National Committee for the Control of Nuclear Activities (CNCAN), from Romania. In this work are analyzed the following types of samples surface water and groundwater, drinking water, soil, spontaneous and cultivated vegetation, atmospheric depositions and precipitations, aerosols. The level of air radioactivity is monitored using the environmental thermoluminescence dosimetry (TLD) system and the associated method. This monitoring is backed by the data from IFIN-HH Weather Tower that records the absorbed dose in air real on time.

1.1 Sampling and sample conditioning

Sampling, conditioning and measurement of samples are made in compliance with the specific laboratory procedures.

The equipment used for sampling and measurement are: i) aerosol pump with a flow of at least 5 m³/h; ii) high sensitivity gross alpha-beta activity system, without window with P10 gas circulation; iii) gross alpha, beta and gamma activity system with automatic change of the samples with P10 gas; iv) high resolution gamma spectrometry system with HP Ge detector, GEM 25 P4 type.

The environmental thermoluminescent dosimeters used for air radioactivity monitoring are placed in the studied area for a period of three months. The sampling points cover the area of influence of the institute of about 5 km.

The water samples are collected from the five points from Ciorogarla River and from the wastewater decanter located around the studied area. Each sample of 2L volume was collected from under the water surface and prepared taking into consideration the LDPM laboratory procedures and scientific papers [4-8]. The samplings are shaken for homogenize the content.

In order to analyze, a quantity of (1.00 ± 0.01) L from the water sample is used. The sample is placed in the porcelain capsule on an electric heater for slow evaporation without boiling. The rest of the sample is stored in the processing room as blank test for a possible revaluation. After the capsule is cooled down, the residue deposited on the bowl walls is transferred and uniformly distributed on the tray used for measurements. For drinking water radioactivity analysis the same procedures were followed.

The soil samples are collected from flat areas and unaffected by corrosion or sedimentation. In an area of 1.00 m², from the first 5.00 cm more individual samples are collected for obtained a sample of about 0.5 kg. After the sample drying at (105 ± 5)°C until a constant mass, a quantity of 0.5 g is evenly distributed on a measuring tray and subjected for alpha-beta global analysis.

The spontaneous vegetation was collected from the same points as the soil samples. The vegetation is cut at a height of around 2 - 3 cm above ground and a known quantity of fresh mass is dried to constant mass. The conditioning process is continued by calcinations that are made gradually, as follows: 6 hours at a temperature of (150 - 200)°C, 4 hours at (300 ± 10)°C and 8 hours at (350 ± 10)°C. For the alpha-beta global analysis, the sample consists of 0.25g of ash that is placed in a stainless steel tray with diameter of (50 ± 2) mm and height of (6 ± 2) mm. The sample is evenly distributed on the tray surface and is fixed with alcoholic solution.
The aerosols are very important samples used to characterize the level of radioactivity in the atmosphere [9] and are taken from two points of interest that are representative for studies. A sampling point is in the institute courtyard and the second is at a distance of 5.00 km from institute.

The radiometric method is used for assessment the density of radioactive aerosols (volumetric density). The sampling is done at a height of 10.00 m from the ground. The suction process is of minimum five hours and the quantity of aspirated air is between 18.00 m$^3$ and 20.00 m$^3$ volumes. The filter placed on the stainless steel tray was measured for to determine the beta global activity level. The atmospheric depositions are weekly collected using a funnel with a surface of 0.302 m$^2$. The funnel is washed with 0.50 L distilled water that is then prepared in the same way with water samples.

All activities of the laboratory have procedures according with the quality management system [10].

2. RESULTS AND DISCUSSIONS

The equipment used to measure the environment samples are following:
- installation for measurement of gross, beta, gamma activity in low background, with automatic sample changer, Model S5 XLB, Soft ECLIPSE. The minimum detection limits for water are 0.01 Bq, 0.030 Bq and 1.12 Bq for beta, alpha and gamma, respectively;
- installation for measurement of gross alpha-beta activity in ultralow background, Model 9300 PC-GFL, Soft VISTA 2000 with background counting rates of 0.500 ± 0.041 cpm and minimum detectable limit (MDL) of 0.010 Bq for gross alpha and 0.600 ± 0.041 cpm background counting rates and 0.029 Bq MDL for gross beta;
  For both measuring equipment is used P10 gas.
- two aerosol pumps used for sampling with a flow of at least 5 m$^3$h$^{-1}$;
- gamma-ray spectrometry installation for the measurement of low activities.

The gamma-ray spectrometry installation was calibrated at the standard secondary laboratory for Radiation Metrology (LMR) from IFIN-HH at radioactive sources of $^{241}$Am, $^{89}$Sr-Y, $^{137}$Cs and $^{40}$K produced and certified by the LMR, [8,9].

The national and international legislation in force is provided the following maximum radioactivity limit values for water samples: 1.85 Bq/L gross beta and 1.00 Bq/L gross alpha for surface water and groundwater; 1.00 Bq/L gross beta and 0.10 Bq/L for gross alpha for drinking water. For drinking water samples, collected from two points from IFIN-HH, the measured radioactivity values were: 0.02 ÷ 0.05 Bq/L global alpha concentrations and 0.4 ÷ 0.6 Bq/L global beta concentration.

The Figure 1 presents the maximum monthly gross beta radioactivity concentration in groundwater and surface water samples collected from the platform Magurele and Ciorogarla River.
The gross beta activity values measured in the water sampled from Ciorogarla River are comparable with very small variations and do not exceed (0.71 ± 0.12) Bq/L. These are considerably lower than the maximum limit value of 1.85 Bq/L. The gross beta values measured in the water upstream collected are lower than those measured in water downstream collected because the wastewater from institute is discharged in river between the two points of sampling.

By analyzing gamma spectrometry performed on cumulative samples of water from Ciorogarla River, the concentration of radioactivity of $^{137}$Cs and $^{40}$K was below the detection limit of the measuring system.

Regarding the aerosols samples analysis the gross beta activity was of 0.033 ÷ 0.198 Bq/m$^3$ and the gamma global concentration was between 0.044 Bq/m$^3$ and 0.077 Bq/m$^3$. In accordance with the current legislation, the maximum allowable value is 3.85 Bq/m$^3$.

The most relevant studies are obtained on the soil samples collected from common points with the samples of Ciorogarla River water and emphasize the natural and artificial radionuclide migration. In Table 1 are given the radioactivity values of the cumulated samples during the studied year, obtained by gamma-ray spectrometry analysis.

**Table 1**

<table>
<thead>
<tr>
<th>Sampling point/ Radionuclides</th>
<th>Upstream Bq/kg dry weight</th>
<th>P. M. channel Bq/kg dry weight</th>
<th>I.W. channel Bq/kg dry weight</th>
<th>Downstream Bq/kg dry weight</th>
<th>Decanter Bq/kg dry weight</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{137}$Cs</td>
<td>13.8 ± 1.5</td>
<td>16.3 ± 1.9</td>
<td>16.3 ± 1.9</td>
<td>14.4 ± 1.7</td>
<td>10.7 ± 1.8</td>
</tr>
<tr>
<td>$^{40}$K</td>
<td>610 ± 27</td>
<td>635 ± 29</td>
<td>635 ± 29</td>
<td>573 ± 27</td>
<td>350 ± 15</td>
</tr>
</tbody>
</table>
The Table 1 shows that without the 40K natural radionuclide, the artificial radionuclide $^{137}\text{Cs}$ is present. Taking into consideration that $^{137}\text{Cs}$ is not produced by any kind of common activity from IFIN-HH their presence is due to the Chernobyl event; the $^{137}\text{Cs}$ concentration is slightly lower and this fact is highlighted and in soil samples collected from common points with those of water and soil. The long half-life of the $^{137}\text{Cs}$ makes this radionuclide to resist over the time. Regarding the global beta activity in soil samples was obtained a value of 670 ±1020 Bq/kg.

The vegetation samples were subjected to measurements of gross beta, gross gamma global and gamma spectrometry. So, the results for vegetation were: 140.00 ± 200.00 Bq/Kg dry mass gross beta activity; gamma global was under detection limit of the measurement system; $^{137}\text{Cs}$ was between 1.23÷ 2.25 Bq/Kg dry mass and $^{40}\text{K}$ activity ranged from 138.30 Bq/Kg dry mass to 180.00 Bq/Kg dry mass.

For samples of atmospheric deposits the monthly average value evolution of the specific gross beta activity is shown in Figure 2. There are two sampling points inside and outside of the institute, in studied area.

![Fig. 2 – Gross beta activity measured on the atmospheric deposition sample](image)

The gross beta radioactivity varied throughout the year and the measured values are comparable for those two points of collection. In February, March, June and October the radioactivity given by gross beta measurement is significantly higher than the rest of the year. In these periods, the weather was characterized by rain, wind, precipitation and these influenced the sample radioactivity. In the rest of the studied year weather conditions were moderate with quite modest rainfall and therefore the gross beta radioactivity values were below 0.55 Bq/m$^2$day. By gamma spectrometric analysis performed on samples of atmospheric depositions and precipitation cumulated quarterly, the $^{137}\text{Cs}$ artificial radionuclide was below the detection limit of the measurement system and properly method. The Environmental TL Dosimetric System, SDTM type that consists of TL Reader, RA 94 type with heating cycle of (25 - 245)$^\circ\text{C}$ and detectors, Gr200-A type was used.
for measurement of the environment (air) radioactivity. The TL method is accredited to measure the H*(10) ambient dose equivalent on the 0.01 ÷ 100.00 mSv range with an uncertainty under 12%. The TL dosimeters were placed in 35 monitoring points from aria of influence inside and outside of the institute. The quarter mean values of the environmental equivalent dose rate recorded in 2012 were: (94.63 ÷ 3.45) nSv/h in the first quarter; (100.29 ÷ 1.98) nSv/h for the second quarter; (106.25 ÷ 5.82) nSv/h for third quarter and (104.81 ÷ 4.22) nSv/h in the fourth quarter.

3. CONCLUSION

In order to evaluate the level of radioactivity several measurement methods were used: radiometric, gamma spectrometry and passive dosimetry; The values determined for the concentration of radioactivity in environmental samples from institute area of influence are comparable with those measured on the samples collected from different nationally places. This led to a value of dose received by people in the critical group of 24.55 µSv/year; the dose constraint is lower than 100 µSv/year approved by the National Commission for Nuclear Activities Control. As conclusion, the specific nuclear activities from IFIN-HH institute have not a radiological impact on the population and environment.

REFERENCES