STUDIES ON THE AEROSOL RADIOACTIVITY LEVEL AND AIR QUALITY AROUND NUCLEAR RESEARCH INSTITUTE AREA

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Abstract. The paper presents studies on the evolution of the aerosol radioactivity level due to the nuclear research activities performed by the Horia Hulubei National Institute for R&D in Physics and Nuclear Engineering, IFIN-HH, during the period 2007–2014. The radiometric method was used to determine the radiation activity concentration from the aerosol samples. The measurements were performed as follows the gross beta activity measured after 3 minutes, 20 hours and 5 days from the sample collection, using a high sensitivity gross alpha-beta activity system and the gross gamma activity measured after 5 days of sampling using a gamma measurement system. The maximum mean values of the radioactivity concentration regarding the years from studied period were 3.78 ± 2.22 Bq m⁻³, 0.19 ± 0.11 Bq m⁻³ and 0.013 ± 0.006 Bq m⁻³ at 3 minutes, 20 hours and 5 days respectively, from the sample collection for gross beta. The values obtained by gross gamma and gamma spectrometry ray were below the detection limit of the systems of measurements. The artificial radionuclide content in the aerosols samples around IFIN-HH for the studied period is situated within the range of aerosols radiation concentration recorded in other Romanian areas where there are no nuclear activities.

Key words: Radioactivity, air quality, aerosols.

1. INTRODUCTION

From the beginning the radiation effects have been used in industry, medicine, research field, university studies and power plant. Newest, radiations have applications in agriculture, space exploration, low enforcement, archaeology (carbon dating), geology and many others and the nuclear research institutes work in order to improve the radiation applications. All flora and fauna are daily exposed to natural and man-made radiations. The natural radiation consists of low levels of uranium, thorium and their decay products concentrated in soil, water and vegetation.

The man-made radiations are produced by space aircraft, radioactive waste, X-ray medical diagnostics, nuclear medicine and others. Technically Enhanced Natural Occurring Radioactive Materials (TENORM), from uranium cycle, general mining, building materials. Natural sources of radiation account for about 82% ± 88% while man-made sources account is of 12% ± 18% depending on the place and country infrastructure [1]. Besides, the nuclear accidents and dirty bombs contribute to the increase of the radioactivity concentration in air, soil, water, vegetation. After many years of the Chernobyl nuclear incident the 137Cs traces are still present in the environmental samples. The population, environment and occupational exposure must be protected against the harmful effects of ionizing radiation by an elaborated system of radioprotection. Referring to man-made radionuclide, the Romanian legislation admits a maximum radioactivity concentration of 3.7 Bq m⁻³ of aerosols in environmental air.

IFIN-HH, located in the town of Măgurele, Romania performs research activities in different nuclear area, as follows VVR-S reactor decommissioning, Tandem Van der Graaff particle accelerator, radioisotopes production for medical application, storage of radioactive waste, multi-purpose irradiation using high activity gamma radiation sources. The nuclear research activities make use of the following unsealed and sealed radioactive sources of different activities: ¹⁹²Ir, ¹³¹I, ⁶⁰Co, ¹³⁷Cs, ¹³⁴Cs, ²⁴¹Am, ¹⁰⁸mAg, ¹⁵²Eu, ¹⁵⁴Eu, ¹⁵⁵Eu. These are the component radionuclide emitted into the atmosphere due to the decommission activity of the nuclear research reactor and radionuclide production. In 2012 the ¹³¹I radionuclide production has been stopped. The specific activities from IFIN-HH require an environmental radioactivity monitoring program which is approved by the National Commission for Nuclear Activities Control, CNCAN, the nuclear authority of Romania in accordance with nuclear legislation [2, 3].

At the beginning IFIN-HH assured the environment nuclear security by basic measurement methods of gross alpha, beta, gamma radiation levels for water and soil samples within a radius of about 10 km around institute and with active dosimeters inside of nuclear laboratories. Since 2005, new methods and technologies of environmental radioactivity measurements have been applied in the institute and all data are centralized in the nuclear monitoring program. As such, the environmental radioactivity monitoring program includes data obtained by the following radiation measurement systems and equipment with particular methods. The TLD environmental monitoring is used for assessment of the radioactivity level in air at one meter above ground. With the gross alpha, beta, gamma global radiation measurement systems are performed multiple types of environmental samples (soil, surface and underground water, natural and cultivated vegetation, sediment, atmospheric deposition, aerosols). The gamma-ray spectrometry analyses are used for cumulated sample measurement. The Real-On Time Survey System METEOROLOGY and RADIATION IFIN-HH is the weather station [http://meteo.nipne.ro/logger/months2012.html] and records IFIN-HH around area.
The environmental radioactivity data obtained by nuclear methods were published in different scientific papers [4–6] and aerosol data were annually communicated by IFIN-HH to the nuclear authority and environmental agency of Romania. The majority of the published studies on the aerosols radioactivity lead to the environmental protection by a continuous radiation monitoring [4, 7, 8]. The nuclear activities from IFIN-HH generate aerosols that could lead to the radioactivity concentration increasing at the workplace and around the institute. The workers and the population around IFIN-HH area risk external exposure and internal contamination caused by aerosol inhalation and external beta and gamma radiations.

In order to take into account the harmful effect of the radiations, it is important to know the evolution in time of the radioactivity level. In this paper, there are presented the levels of radioactivity in the atmosphere around IFIN – HH nuclear institute obtained by measurement of gross alpha, beta and gamma on distinct samples, as well as gamma spectrometry measurements on cumulated samples during the period 2007–2014.

In this paper, the atmospheric radioactivity levels around IFIN – HH nuclear institute will be presented. These were obtained by the measuring of gross alpha, beta and gamma on distinct samples, as well as gamma spectrometry measurements on cumulated samples. The data were obtained on the 2007–2014.

2. METHODS

The aerosols radioactivity level studies were performed on two different days of the same month in the courtyard of the institute. The interest sampling point was chosen depending on the radioactive emission sources and the wind direction. The sampling procedure was performed at a height of 10 m from the ground using a specific suction pump with specific filters for 5:00 hours per sample. The volume of aspirated air is between 18 m$^3$ and 20 m$^3$ per sample. The radiometric method for to determining the radioactive aerosol density (volumetric density) was used [9, 10].

The equipment used for sampling and measurements is presented in detail in the paper [8]. The aerosol pump used for sampling has a flow rate of least 5 m$^3$ h$^{-1}$. The high sensitivity gross alpha-beta activity system without window with P10 gas circulation (mixture of 90% methane and 10% argon) is made of Protean Instrument Corporation – USA. The alpha, beta and gamma gross activity system with automatic samples uses P10 type gas. For gross gamma activity the minim detectable activity (MDA) is 1.25 Bq.

The equipment is verified before starting the measurements using $^{241}$Am, $^{90}$Sr-Y and $^{137}$Cs reference sources made of the Radionuclide Metrology
Laboratory from the IFIN-HH with radioactivity values of about 30 ÷ 300 Bq. The standard radioactive sources have diameters similar to the aerosols suction filters. The equipment of measurement is calibrated at each procedure using the standard sources.

The filters are handled with tweezers and are carefully examined before starting to collect the samples in order to exclude any damage or irregularities.

The aerosol radioactivity is caused by i) short-lived progeny of $^{222}\text{Rn}$ natural radionuclide with half-life of 30 minutes; ii) thoron natural radionuclide, $^{220}\text{Rn}$, for which the half-time is 10 h 30 min; iii) artificial radionuclide with long half-life, as $^{137}\text{Cs}$, $^{125}\text{Sb}$, $^{90}\text{Sr}$ etc.

In order to establish the concentration of radioactivity on artificial radionuclide emitting beta and gamma radiation the gross beta activity measured after 3 minutes, 20 hours and 5 days from the sample collection were performed. For all measurement the high sensitivity gross alpha-beta activity system was used. The gross gamma activity was measured after 5 days of sampling with the gamma measurement system.

The radiometric method was described in detail in [7, 8] papers which report the measurements performed during 2006 and for 2010 and other three months in 2011.

Each filter was measured in order to determine alpha, beta, gamma global radioactivity and the gamma spectrometric analysis was performed on the sets of filters representing the sampling on months.

3. RESULTS AND DISCUSSIONS

The experimental results show the aerosols radioactivity concentration in the IFIN-HH area of influence over the 2007–2014. Table 1 shows the mean annual concentration values of radioactivity determined for all sampling periods. The samples were taken of twice per month, usually between 9:00 and 14:00 from both interest points.

The data presented in the Table 1, columns 1 and 2 and 3 show the radioactivity concentrations given by the natural and man-made radionuclide. The gross gamma radioactivity concentration values were below the detection limit of the measurement system.

The first column presents the radioactive concentration due to short-live progeny of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ natural radionuclide. In the second column there are only $^{220}\text{Rn}$ radionuclide daughters. In the third column the radioactivity values are given by gamma and beta emitting artificial radionuclide. In 2007 and 2008 the radioactivity concentrations given by the natural and man-made radionuclide were higher than the rest of the studied period.
### Table 1

Mean values of the aerosol gross beta radioactivity concentration from IFIN-HH area in the 2007–2014

<table>
<thead>
<tr>
<th>Year</th>
<th>Gross beta activity after 3 min of sampling [10^{-1} \text{Bq/m}^3] (1)</th>
<th>Gross beta activity after 20 hours of sampling [10^{-3} \text{Bq/m}^3] (2)</th>
<th>Gross beta activity after 5 days of sampling [10^{-3} \text{Bq/m}^3] (3)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2007</td>
<td>37.84 ± 22.17</td>
<td>1.60 ± 0.66</td>
<td>7.43 ± 0.73</td>
</tr>
<tr>
<td>2008</td>
<td>34.56 ± 25.58</td>
<td>1.91 ± 1.08</td>
<td>12.51 ± 5.84</td>
</tr>
<tr>
<td>2009</td>
<td>21.58 ± 16.54</td>
<td>1.31 ± 0.50</td>
<td>4.78 ± 1.54</td>
</tr>
<tr>
<td>2010</td>
<td>17.81 ± 8.23</td>
<td>1.28 ± 0.50</td>
<td>4.89 ± 1.20</td>
</tr>
<tr>
<td>2011</td>
<td>19.36 ± 13.14</td>
<td>1.19 ± 0.58</td>
<td>5.98 ± 1.97</td>
</tr>
<tr>
<td>2012</td>
<td>25.21 ± 14.78</td>
<td>1.30 ± 0.69</td>
<td>5.71 ± 3.27</td>
</tr>
<tr>
<td>2013</td>
<td>22.71 ± 13.73</td>
<td>1.13 ± 0.39</td>
<td>5.05 ± 1.42</td>
</tr>
<tr>
<td>2014</td>
<td>26.07 ± 13.98</td>
<td>1.42 ± 0.43</td>
<td>4.65 ± 1.08</td>
</tr>
<tr>
<td>Mean</td>
<td>26.13 ± 7.48</td>
<td>1.44 ± 0.3</td>
<td>7.55 ± 2.81</td>
</tr>
</tbody>
</table>

The 2007 and 2008 were rich in rainfall and these conditions caused the aerosol particles deposition on the sample filters. For example, in case of gross beta activity measured after 3 minutes the radioactivity concentration in 2007 is 53% higher than in 2010. Regarding the man-made radionuclide activity refer to the gross gamma activity all measured values were below the minimum detectable activity of the equipment. All of these artificial radioactivity values measured at 5 days after sampling were very low comparing with the maximum radioactivity of 3.85 Bq m$^{-3}$ allowed by the nuclear legislation.

Although in the studied period the nuclear research reactor was in the decommissioning procedure and the radionuclide center produced unsealed radioactive sources for medical applications the aerosol radioactivity was well below the radioactivity maximum allowed limit in air samples.

Regarding the high standard deviation of the measured values, each year high variations were recorded for the gross beta activity measured at 3 minutes of sampling because the values were very high in cloud season as compared with warm weather. This case is presented in Fig. 1.

In the years of 2007 and 2008, the levels of radioactivity were over 9.00 Bq m$^{-3}$ for January and October. Apart from these, the concentration in the remaining months of the year is very low comparing with radioactivity concentration limit allowed by the nuclear legislation. The gross beta activity level of the aerosol samples measured after 3 minutes of sampling was significantly influenced by the atmospheric conditions (temperature, precipitation), and this is highlighted in the graphs. Also, a similar situation was recorded for gross beta activity measured at 20 minutes after sampling. Gross beta activity values...
measured after 20 hours of sampling were published in work [7] for first two months of 2010 and 2011. The measurements of radioactivity concentration from March, 2011 were daily due to the Fukushima event [7].

![Fig. 1 – Monthly radioactivity level in aerosol samples measured after 3 minutes of sampling.](image1)

Figure 2 shows that the values of artificial radioactivity determined after 5 days of sampling have a homogenous distribution over the entire period and are significantly lower than the maximum allowed values. The exception is 2008 when the nuclear research activities were more intense and the measurements of radioactivity concentration are much higher.

![Fig. 2 – The level of monthly radioactivity in aerosol samples measured after 5 days of sampling.](image2)
These measurements are in completion of some existing measurements conducted at the institute such as, the tritium and iodine the measured on production place, from the radioisotope Department. The cumulated values for this time frame have resulted in a smaller value than the established derived limits for gaseous effluent release approved by CNCAN, of 100 μSv·year\(^{-1}\).

4. CONCLUSIONS

The work highlights the air quality around the nuclear research institute taking into consideration the radiation protection norms issued by the Romanian nuclear authority.

The investigations performed during the decommissioning of the VVR-S nuclear reactor show that the air radioactivity concentration is well below the limits allowed by the radiation protection legislation.

These data are recorded through the radiation protection program approved by Romanian nuclear authority and achieved 30 years for any future reference.

The level of radioactivity for aerosols collected from the influence area of IFIN-HH is comparable with the national value.

Working at the IFIN-HH and in other institutes on the Magurele Research Area has not revealed any radiological impact on the population and environment.

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REFERENCES

