DYNAMICS OF OUTDOOR RADON AND THORON PROGENY CONCENTRATIONS IN SOME GEOGRAPHICAL AREAS OF ROMANIA*

E. SIMION1,2, I. MIHALCEA2, V. CUCULEANU3,4, F. SIMION1,3

1National Environmental Protection Agency, Radioactivity Laboratory, Splaiul Independentei 294, RO-060031, Bucuresti, Romania
2Faculty of Chemistry, University of Bucuresti, 4-12 Regina Elisabeta Av., 030017-Bucuresti, Romania
3Faculty of Physics, University of Bucuresti, Magurele, RO-077125, Bucuresti, Romania
4Academy of Romanian Scientists, 54 Splaiul Independentei, RO-050094, Bucharest, Romania

Received November 15, 2012

The variation of outdoor $^{222}$Rn and $^{220}$Rn progeny concentrations were investigated for 5 years (2002 - 2006). The results presented in this paper were obtained within the framework of the monitoring program performed by the Environmental Radioactivity Monitoring Stations (ERMS) situated in Toaca, Iasi, Cluj Napoca, Craiova, Bucuresti and Constanta, part of the National Environmental Radioactivity Survey Network (NERSN), coordinated by National Environmental Protection Agency (NEPA). The measuring method is based on the total beta measurements of atmospheric aerosols filters, using a low background total beta counter and ($^{90}$Sr/$^{90}$Y) reference standard. Analysis of the time series of progeny concentrations in the low atmosphere makes evident different patterns of variation of these concentrations: diurnal, seasonal and annual variations.

Key words: radon, thoron, progeny concentration, patterns of variations.

1. INTRODUCTION

Radon ($^{222}$Rn) and thoron ($^{220}$Rn) are considered the main natural sources of radiation that contribute to human exposure. $^{222}$Rn (half-life of 3.824 days) and $^{220}$Rn (half-life of 55.6 seconds) are inert radioactive gases produced by the Earth's crust as decay products of $^{226}$Ra and $^{224}$Ra, from decay series of $^{238}$U, respectively $^{232}$Th, radionuclides present in all terrestrial materials.

Both $^{222}$Rn and $^{220}$Rn are noble gases emanating from the soil and the concentration is depending on soil type and the physical conditions such as moisture, frost, snow and weather conditions (pressure, wind and air temperature) [4, 5, 6].

In normal conditions, natural background radiation is a significant component given by radon, thoron and progeny, approximately 43% of the annual effective dose received by the population [2].

In Romania, measurements made over a period of 9 years (1980-1988) led to monthly averages 1-9 Bq/m³ for ²²²Rn progeny and 0.001 to 0.5 Bq/m³ for ²²⁰Rn progeny [1].

2. RADON AND THORON PROGENY MEASURING METHOD

In the National Network of Environmental Radioactivity Surveillance at all Environmental Radioactivity Surveillance Stations for the time periods analyzed, in order to determine the variation in time of ²²²Rn and ²²⁰Rn progeny concentrations, it was used a unified working program for sampling, measuring and calculating the activity of these concentrations.

Principle of measurements of ²²²Rn and ²²⁰Rn progeny concentrations was based on atmospheric aerosols collected through a nozzle located 2 feet from the Earth's surface, using high volume aerosol samplers, paper glass microfiber filter with pores size between 1.0 ÷ 1.2 mm and retention efficiency of 80%.

Aerosol samples were obtained by aspirating a specified volume of air through a special filter. Atmospheric aerosol sample collection is done daily at regular intervals with suction period of 5 hours as follows: the A1 aspiration, 02:00 ÷ 07:00; the aspiration A2, 08:00 ÷ 13:00; the aspiration A3, 14:00 ÷ 19:00; the aspiration A4, 20:00 ÷ 01:00.

Atmospheric aerosol sample collection was performed using nozzle sampling system (filter holder), made from a rigid material to ensure flatness during cleaning filter for easy access of air through the entire filter surface, thus ensuring uniform deposition over the entire surface of the filter. Nozzle was located at a distance of 2 m from the ground in an open space outside urban areas. Suction pump airflow setting was done automatically and the switching of the 4 aerosol filters was made at predetermined time. According to the sampling procedure of the NERSN, air flow aspiration, with mounted filter, was 5 m³/h [3].

To determine ²²²Rn and ²²⁰Rn progeny concentrations in atmospheric aerosol samples there were performed measurements in a unified manner by all ERMS consisting in immediate measurements (3 minutes after aspiration time) for 1000 seconds, then remeasurements to 20 hours after ending of aspiration for 3000 seconds.

Filters were measured using a low background global beta counter with a (Sr/Y)⁹⁰ calibration source [8].
3. RESULTS AND DISCUSSIONS

To characterize the time evolution of $^{222}$Rn and $^{220}$Rn progeny concentrations it was studied the diurnal, monthly, seasonal and annual variation for continuous measurements during 2002-2006 for some geographical areas of Romania: mountain (ERMS Toaca), hill (ERMS Cluj Napoca and ERMS Iasi), Plain (ERMS Craiova and ERMS Bucuresti) and maritime (ERMS Constanta).

3.1. ANNUAL DYNAMICS PER ASPIRATIONS

Considering the A1 (02-07) aspiration, the maximum annual average values for radon concentrations were at similar levels for the hill and plain areas (Bucuresti 14.52 Bq/m$^3$ in 2006, Cluj Napoca 12.72 Bq/m$^3$ in 2002, Iasi 13.06 Bq/m$^3$ in 2002, Craiova 10.26 Bq/m$^3$ in 2002) and the minimum values were obtained for mountain areas (Toaca 0.88 Bq/m$^3$ in 2003) and maritime (Constanta 5.13 Bq/m$^3$ in 2003) areas.

In case of thoron progeny concentrations, it can be observed that the maximum annual average concentrations were obtained in the hill and plain areas (Bucuresti 0.620 Bq/m$^3$ in 2006, Cluj Napoca 0.272 Bq/m$^3$ in 2003, Iasi 0.440 Bq/m$^3$ in 2002, Craiova 0.318 Bq/m$^3$ in 2002) and that the minimum annual average values were obtained in mountain and maritime areas (Toaca 0.017 Bq/m$^3$ in 2003 and Constanta 0.190 Bq/m$^3$ in 2002).

As far as A2 (08-13) aspiration is concerned, it can be clearly seen that the annual average concentrations for both $^{222}$Rn and $^{220}$Rn were much lower than the values obtain in case of A1 (02-07) aspiration. These values can be explained by the fact that soil heating by sun causes the air convection process which is intensifying the diffusion of radon and thoron accumulated near Earth’s surface during the night due to stable atmosphere and calm.

Diffusion and transport intensification of air masses in the planetary boundary layer determines the decrease of $^{222}$Rn and $^{220}$Rn progeny concentrations in air.

Maximum values of radon progeny were approximately of the same order of magnitude for stations located in plain areas (Bucuresti 5.64 Bq/m$^3$ in 2006, Cluj Napoca 6.2 Bq/m$^3$ in 2003, Iasi 6.68 Bq/m$^3$ in 2002, Craiova 5.89 Bq/m$^3$ in 2002) and the minimum values were obtained for the mountain station Toaca 1.15 Bq/m$^3$ in 2003 and the maritime station Constanta 3.18 Bq/m$^3$ in 2003.

For the thoron progeny concentrations, similarly to the radon pattern, at stations located in plain areas, the values were comparable (Bucuresti 0.210 Bq/m$^3$ in 2006, Cluj Napoca 0.148 Bq/m$^3$ in 2002, Iasi 0.234 Bq/m$^3$ in 2002, Craiova 0.184 Bq/m$^3$ in 2002) and minimum values were obtained for stations located in the mountains, Toaca 0.016 Bq/m$^3$ in 2003, and the maritime area, Constanta 0.09 Bq/m$^3$ in 2003.

On aspiration A3 (14-19), radon and thoron progeny concentrations were minimum because during period of day the air temperature has maximum values
which results in a particular increase of turbulent diffusion and transport what is reducing the concentrations.

Fig. 1 – The annual dynamics of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ progeny concentrations per aspirations.
High average annual values for the progenies of radon are found in plain located stations Cluj Napoca (5.11 Bq/m³ in 2003), Iasi (5.89 Bq/m³ in 2002), Craiova (4.5 Bq/m³ in 2002) and Bucuresti (5.8 Bq/m³ in 2006), and low average annual values were obtained for stations in mountain area, Toaca (1.09 Bq/m³ in 2003), and large area, constant (2.8 Bq/m³ in 2003).

Similar with radon progeny trends, thoron progeny have maximum annual mean values in stations located in plain areas Bucuresti (0.152 Bq/m³ in 2006), Cluj Napoca (0.089 Bq/m³ in 2003), Iasi (0.147 Bq/m³ in 2002), Craiova (0.114 Bq/m³ in 2002), and low average annual values were obtained for stations in mountain areas, Toaca (0.017 Bq/m³ in 2003) and from the maritime area, Constanta (0.05 Bq/m³ in 2003).

On aspiration A4 (20-01), the maximum average annual values for radon progenies are found in plain area stations Cluj Napoca (10.95 Bq/m³ in 2003), Iasi (9.39 Bq/m³ in 2002), Craiova (7.05 Bq/m³ in 2002) and Bucuresti (11.23 Bq/m³ in 2006), and low average annual values were obtained for stations in mountain area Toaca (0.97 Bq/m³ in 2003) and in the maritime area Constanta (4.1 Bq/m³ in 2003).

From the analysis of annual average concentrations of $^{222}\text{Rn}$ and $^{220}\text{Rn}$ progeny obtained at the six stations studied, in 2002–2006, it can be seen that the mean annual radon and thoron progeny during daytime (aspirations A2 and A3) are lower than those obtained in the aspirations of the night (aspirations A1 and A4). Physical explanation of these variations is that in daytime turbulent diffusion is enhanced by increased temperature specific planetary boundary layer and atmospheric stability at night is more pronounced because of thermal inversions and calm atmosphere [3].

In Toaca case, average annual values per aspirations of radon and thoron progeny concentrations are much lower than those specific to the other stations that can be explained by the fact that Toaca is located in the mountain area at 1904 m altitude, where thickness of the soil is extremely small, predominantly rocky, so has a reduced exhalation rate. Also, the thermal convection from the lower air layers contributes to the increase of radon progeny levels during the day [1].

In case of Constanta, $^{222}\text{Rn}$ and $^{220}\text{Rn}$ progeny concentrations, annual average values were relatively low compared to the rest of the stations analyzed (except Toaca where were the lowest values), these concentrations and trends are similar and can be explained by marine air, which has a concentration of progenies much lower than in terrestrial areas. Thus, because the marine air is transported by wind in the area where the station is located, the results of measurements indicated a decrease of progeny concentrations [1].
3.2. SEASONAL VARIATIONS

As shown in the case of annual variations, Toaca station being located in a mountain higher concentrations corresponding to A2 and A3 aspirations can be explained by the air mass transport by thermal convection from the low-lying areas, characterized by higher progeny concentrations.

In case of radon progeny, the seasonal average values highlight the variation with maximum in spring and autumn minimum. Regarding thoron progeny, the curves have pronounced variation from month to month, except during winter, where average concentrations are lower than in other seasons. Explanation for lowering monthly averages for thoron progeny in winter consists in a diminishing of exhalation rate in winter compared to the summer.

![Fig. 2 – Seasonal variations of radon and thoron progeny concentrations.](image)

Seasonal variation, represented in Figure 2, is characterized for radon progeny through a minimum in spring (1,173 Bq/m³ registered in Toaca), a maximum in autumn (9.75 Bq/m³ registered in Bucuresti) and for thoron progeny a winter minimum (0.017 Bq/m³, registered in Toaca) and a maximum in autumn (0.36 Bq/m³) registered in Bucuresti.

3.3. MULTIANNUAL VARIATIONS PER DAILY ASPIRATIONS

All stations analyzed, as can be seen in Figure 3, had similar trend during the day (except Toaca), the annual average concentrations on $^{222}$Rn and $^{220}$Rn progeny aspirations had maximum values during the night (02-07), a decrease during the day (08-13) by reaching a minimum in A3 (14-19) and then begin to rise again, thus forming diurnal variation.
3.4. MONTHLY MULTIANNUAL VARIATIONS

In Table 1 are presented multiannual trends for monthly average radon progeny. Minimum monthly variation was 1,144 Bq/m$^3$ and obtained for mountain station Toaca (minimum value in December 1,021 Bq/m$^3$ and 2,165 Bq/m$^3$ maximum in August) and maximum was 5,520 Bq/m$^3$ and was recorded at plain station Bucuresti (4,590 Bq/m$^3$ minimum in March and maximum 10,110 Bq/m$^3$ in October).

<table>
<thead>
<tr>
<th>Month</th>
<th>Bucuresti</th>
<th>Constanta</th>
<th>Toaca</th>
<th>Cluj Napoca</th>
<th>Iasi</th>
<th>Craiova</th>
</tr>
</thead>
<tbody>
<tr>
<td>January</td>
<td>7,06</td>
<td>5,47</td>
<td>1,177</td>
<td>7,241</td>
<td>6,809</td>
<td>6,910</td>
</tr>
<tr>
<td>February</td>
<td>6,61</td>
<td>4,42</td>
<td>1,322</td>
<td>6,674</td>
<td>5,893</td>
<td>5,868</td>
</tr>
<tr>
<td>March</td>
<td>4,59</td>
<td>2,97</td>
<td>1,421</td>
<td>4,888</td>
<td>3,854</td>
<td>3,847</td>
</tr>
<tr>
<td>April</td>
<td>5,17</td>
<td>2,89</td>
<td>1,491</td>
<td>4,592</td>
<td>4,501</td>
<td>3,606</td>
</tr>
<tr>
<td>Mai</td>
<td>5,79</td>
<td>3,11</td>
<td>1,751</td>
<td>5,961</td>
<td>4,690</td>
<td>4,716</td>
</tr>
<tr>
<td>June</td>
<td>6,00</td>
<td>3,46</td>
<td>1,611</td>
<td>5,811</td>
<td>5,027</td>
<td>5,049</td>
</tr>
<tr>
<td>July</td>
<td>7,23</td>
<td>4,05</td>
<td>2,143</td>
<td>6,788</td>
<td>5,891</td>
<td>5,620</td>
</tr>
<tr>
<td>August</td>
<td>9,04</td>
<td>4,73</td>
<td>2,165</td>
<td>7,651</td>
<td>6,946</td>
<td>6,226</td>
</tr>
<tr>
<td>September</td>
<td>9,63</td>
<td>4,99</td>
<td>2,157</td>
<td>7,953</td>
<td>8,068</td>
<td>6,479</td>
</tr>
<tr>
<td>October</td>
<td>10,11</td>
<td>4,88</td>
<td>1,990</td>
<td>8,378</td>
<td>7,809</td>
<td>7,221</td>
</tr>
<tr>
<td>November</td>
<td>9,50</td>
<td>5,27</td>
<td>1,270</td>
<td>9,411</td>
<td>7,763</td>
<td>7,284</td>
</tr>
<tr>
<td>December</td>
<td>9,24</td>
<td>5,97</td>
<td>1,021</td>
<td>8,775</td>
<td>7,748</td>
<td>7,410</td>
</tr>
</tbody>
</table>
For analyzed stations were revealed minimum values for progenies of radon in spring season, in March, April and maximum values were recorded in winter season, November, December, except Toaca station, with peaks in July-September.

<table>
<thead>
<tr>
<th>Month</th>
<th>Bucuresti</th>
<th>Constanta</th>
<th>Toaca</th>
<th>Cluj Napoca</th>
<th>Iasi</th>
<th>Craiova</th>
</tr>
</thead>
<tbody>
<tr>
<td>January</td>
<td>0,113</td>
<td>0,103</td>
<td>0,017</td>
<td>0,056</td>
<td>0,068</td>
<td>0,091</td>
</tr>
<tr>
<td>February</td>
<td>0,230</td>
<td>0,112</td>
<td>0,017</td>
<td>0,069</td>
<td>0,106</td>
<td>0,127</td>
</tr>
<tr>
<td>March</td>
<td>0,263</td>
<td>0,121</td>
<td>0,018</td>
<td>0,117</td>
<td>0,156</td>
<td>0,160</td>
</tr>
<tr>
<td>April</td>
<td>0,386</td>
<td>0,139</td>
<td>0,023</td>
<td>0,152</td>
<td>0,233</td>
<td>0,229</td>
</tr>
<tr>
<td>Mai</td>
<td>0,355</td>
<td>0,122</td>
<td>0,027</td>
<td>0,152</td>
<td>0,212</td>
<td>0,216</td>
</tr>
<tr>
<td>June</td>
<td>0,273</td>
<td>0,135</td>
<td>0,020</td>
<td>0,118</td>
<td>0,189</td>
<td>0,212</td>
</tr>
<tr>
<td>July</td>
<td>0,312</td>
<td>0,139</td>
<td>0,021</td>
<td>0,116</td>
<td>0,177</td>
<td>0,210</td>
</tr>
<tr>
<td>August</td>
<td>0,365</td>
<td>0,157</td>
<td>0,021</td>
<td>0,138</td>
<td>0,181</td>
<td>0,218</td>
</tr>
<tr>
<td>September</td>
<td>0,392</td>
<td>0,171</td>
<td>0,023</td>
<td>0,229</td>
<td>0,253</td>
<td>0,221</td>
</tr>
<tr>
<td>October</td>
<td>0,349</td>
<td>0,147</td>
<td>0,021</td>
<td>0,151</td>
<td>0,213</td>
<td>0,198</td>
</tr>
<tr>
<td>November</td>
<td>0,350</td>
<td>0,159</td>
<td>0,016</td>
<td>0,149</td>
<td>0,207</td>
<td>0,224</td>
</tr>
<tr>
<td>December</td>
<td>0,278</td>
<td>0,179</td>
<td>0,016</td>
<td>0,098</td>
<td>0,226</td>
<td>0,168</td>
</tr>
</tbody>
</table>

In Table 2 are presented trends for multiannual monthly average thoron progeny. Minimum variation was obtained in the mountain station Toaca (minimum 0,016 Bq/m³ in December, and the maximum 0,027 Bq/m³ in May), and the maximum was 0,279 Bq/m³, recorded at the plain station Bucuresti (0,113 Bq/m³ minimum in January and maximum 0,392 Bq/m³ in September).

Lowest monthly variations for thoron progeny were obtained for mountain station Toaca (minimum 0,016 Bq/m³ in December and the maximum 0,027 Bq/m³ in May). The largest variations were recorded for plain station, Bucuresti (0,113 Bq/m³ minimum in January and maximum 0,392 Bq/m³ in September), the minimum values were recorded in winter (minimum values recorded for: Bucuresti, Constanta, Iasi and Cluj Napoca) and the maximum values were recorded in autumn (maximum values: Bucuresti, Cluj Napoca, Constanta and Iasi).

3.5. ANNUAL VARIATION FOR CONCENTRATIONS WITH 2 AND 4 ASPIRATIONS PER DAY

In Figure 4 there are represented the annual average concentrations for 2 and 4 aspirations per day reveals the similarity of annual variations between the concentrations obtained with two and 4 aspirations per day as well as the fact that the aspirations of A3 and A4 are diminishing the diurnal average of the progeny concentrations.
4. CONCLUSIONS

As far as the annual dynamics per aspiration is concerned the least values from the mountain area may be explained by the fact that the exhalation rate of the
specific soil (rock) is very small, the only contribution being from the lower atmospheric layers by means of the vertical transport due to the convection process [10]. As for the maritime area, the lower values of concentrations (with respect to the plain areas ones) may be explained by the influence of the air above the sea has the progeny concentrations with two order of magnitude lower than the respective concentrations from the land area. The plain area station has the greatest values which can be explained by the normal exhalation rate of soil and the usual atmospheric diffusion and transport conditions [7].

Multiannual variations per daily aspirations concentrations for both, radon and thoron progeny, may be explained as follows: the night aspirations A1 and A4 have the greatest values because during the night the atmosphere becomes stable and thermal inversion and atmospheric calm occur quite frequently and for day aspirations A2 and A3 have the least values because during the day the atmospheric diffusion are intensified due to convection process and wind transport. As for the mountain area concentrations, they have a opposite dynamics (the greatest values are during the day and the least values are during the night) [12].

In seasonal variation one may see in case of radon progeny, the values from the spring season are the least and the greatest values are in autumn season. This variation is due to the fact that during the spring, the atmospheric diffusion and transport are more intense as compared with the autumn season. In case of the mountain the variations from season to season are not significant. In case of thoron, the seasonal variation is not significant between maritime and plain areas and the least values are in the mountain area [3].

For monthly multiannual variations the least values for radon and thoron progeny are in March and the greatest during the winter months (because the more intensified stability of the atmosphere). Thoron is much more sensitive to the exhalation rate which is decreasing significantly during the winter.

Acknowledgments. Our colleagues from Environmental Radioactivity Monitoring Stations are acknowledged for their effort in sample collection. The authors would like to thank the reviewer of Romanian Journal of Physics for the useful comments and suggestions made in order to improve the paper.

REFERENCES