Study on atmospheric radioactivity in Suceava Metropolitan Area

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ABSTRACT: Ionising radiation from different environments (sun, celestial bodies, soil and geological substrate, etc.) influences the whole living world as a hidden factor. About 87% of the radiation dose received by humans is due to natural radiation. It is essential to assess radiation doses in order to control possible health effects from such natural sources. In this context we were interested in assessing the atmospheric radioactivity in Suceava Metropolitan Area by performing an experimental field monitoring with gamma dose monitoring, between 15 - 17 July 2021, and an analysis based on gamma dose rate measurements, global specific beta activity and radon/thoron emissions in the period (2009 - 2020) performed by the Environmental Radioactivity Monitoring Laboratory of Suceava Environmental Protection Agency. The obtained results for the external gamma dose, on hourly data show that the warning threshold of 0.250 µSvh-1 was exceeded, on cumulative annual data the external gamma dose rate was between 0.53 mSv in 2012 and 0.97 mSv in 2019, so it did not rise above the threshold of 1 mSv, the threshold from which health problems may occur. For atmospheric aerosols the warning threshold of 10 Bq m^{-3} was not exceeded in any case, and for atmospheric deposition (on the ground) the maximum in observations rose to 61.3 Bq m^{-3}, less than 1/3 of the warning threshold value of 200 Bq m^{-3}. The assessment of Radon and Thoron emissions showed a maximum of 28.4 Bq m^{-3}, a quarter of the human health threshold of 100 Bq m^{-3}.

KEY WORDS: radioactivity, atmosphere, gamma dose, radon, thoron, Suceava.

1. Introduction

In recent decades, entire society has become increasingly concerned on environmental issues on their health and lifestyle. Various studies on environmental polluants have been published. Humans are constantly influenced by the action of multiple environmental agents, including ionising radiation. Environmental radioactivity is represented by cosmic radiation, gamma-earth radiation, water and food products radioactivity, as well as some radioactive gases: radon, thoron, etc. exhaled from the earth's crust, radiation from hygienically significant technogenic radionuclides radiation (137Cs, 90Sr) as a result of nuclear weapons tests and accidents at atomic power plants (PPAs), e.g. the Chernobyl accident (Bahnarel et al, 2011).

Natural radioactivity exposure varies according to radioactive elements present in each location, so researchers have studied the levels of natural radioactivity in the atmosphere, soil and water to provide background information and to detect environmental radioactivity (Radhakrishna et al., 1993). Radioactivity levels can be used to assess the doses of radioactive contamination to which population is exposed and to predict environmental radioactivity changes caused by nuclear accidents, industrial activities or other human activities (UNSCEAR, 2000). Knowledge of radioactivity levels is required radiation interaction damages cells, causing cell death and alterations that result in organ and tissue malfunction and stochastic health impacts (cancer and hereditary effects) in the end.

Radionuclides have been present in nature since the Earth was formed. About 340 radionuclides are known, of which about 70 are radioactive. Radioactivity initiated by the successive decay of uranium and thorium leads to the appearance of radioisotopes of polonium, bismuth, radon, radium, etc. distributed in different layers of the Earth's crust and in all types of rocks and soils (Paschoa et al., 2010) as well as in rivers, marine waters and biological organisms (Ouseph, 1975). Through various geological and environmental events, such as volcanic and seismic activity, droughts, floods, earthquakes, fires, etc., various processes take place whereby the radioactive equilibrium is disrupted, the decayed radioactive elements of uranium and thorium gradually disintegrate and are released into the atmosphere or form secondary sediments. Radium is a radioactive element found in the composition of brown clays, so it is present in soil, ocean and river waters, and its decay products (radon, thoron and actinon) can be found in all natural environments (Burghele, 2013).

In this study we are concerned with terrestrial radionuclides gamma radiation, without considering cosmic radiation. Kendall et al. (2016) estimated in a study for UK that the average dose from cosmic radiation would be 43 nGy h^{-1} outdoors and 34 nGy h^{-1} indoors. Most studies focus on indoor radiation. Although indoor cosmic ionising radiation penetrates more slowly, there is a higher concentration of radiation, building materials being a source of radiation (Kendall et al., 2018, 2021).

At some sites/locations in Romania, researchers have focused their attention on the outdoor absorbed gamma dose rate in relation to the natural radioactivity and meteorological conditions in Bucharest (Baciu, 2006). Dolha et al. (2014) produced a high-resolution gamma dose map for Cluj County, this study completing the research carried out a year earlier to determine the gamma dose in 7 sites with different geology: Arad, Cluj-Napoca, Bologa, Miercurea-Ciuc, Predeal, Beliș, Sinaia (Dolha et al., 2013). Encianu et al. (2005) measured outdoor radon and thoron concentrations during 1994-1997 in the municipality of Bistrita. Studies were then directed towards determining inside dwellings radiation levels in uranium mining areas of Bihor (Sandor et al., 1999), Băița-Ștei (Cosma et al., 2013), in wells in Băița-Ștei (Burghele et al., 2012), in soil (Cosma et al., 2010; Papp, 2010). Cuculeanu et al., 2011 investigated the radon concentrations in the low atmosphere and their correlations with meteorological parameters for Bacău city. Also, the north area of Mures county was analysed, the authours aimed to determine residential radon concentrations, radon concentrations in soil and in waters (Papp et al., 2017.).

The main objective of this study is to assess the exposure of AMSv population to natural and artificial atmospheric ionizing radiation. To quantify the environmental radiation background in a location and to calculate the risk to human health from both natural and artificial sources, measurements of gamma dose rate in air are crucial.

2. Study area

Our study focused on determining of alpha, beta, and gamma radiation levels in 30 points (Figure 1) located in the Suceava Metropolitan Area (AMSv), which includes the municipality of Suceava and 6 neighbouring localities: Șcheia, Moara, Ipotești, Salcea, Adâncata, Mitocu Dragomirnei and Pătrăuți. The examined area is about 408.39 km^2 and the Suceava river valley runs through its centre.

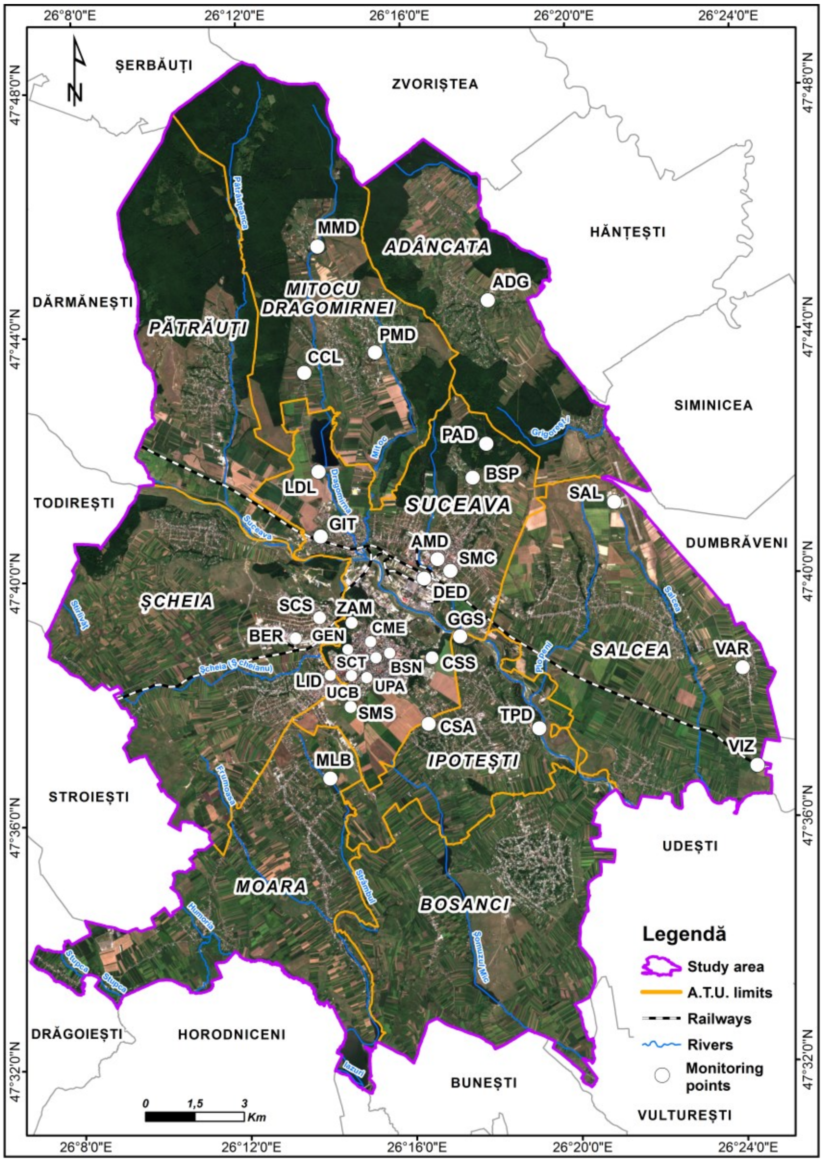


Figure 1 Airborne gamma dose rate monitoring points.

The studied territory presents geological formations that are located within the platform unit, Moldavian Platform, showing an old crystalline bedrock of Precambrian age, later covered by a sedimentary stack. Thus, the investigated area consists of a succession of Volhynian clay marls with sandy interlayers, gravels, and recent alluvial sediments of the Holocene age (Figure 2).

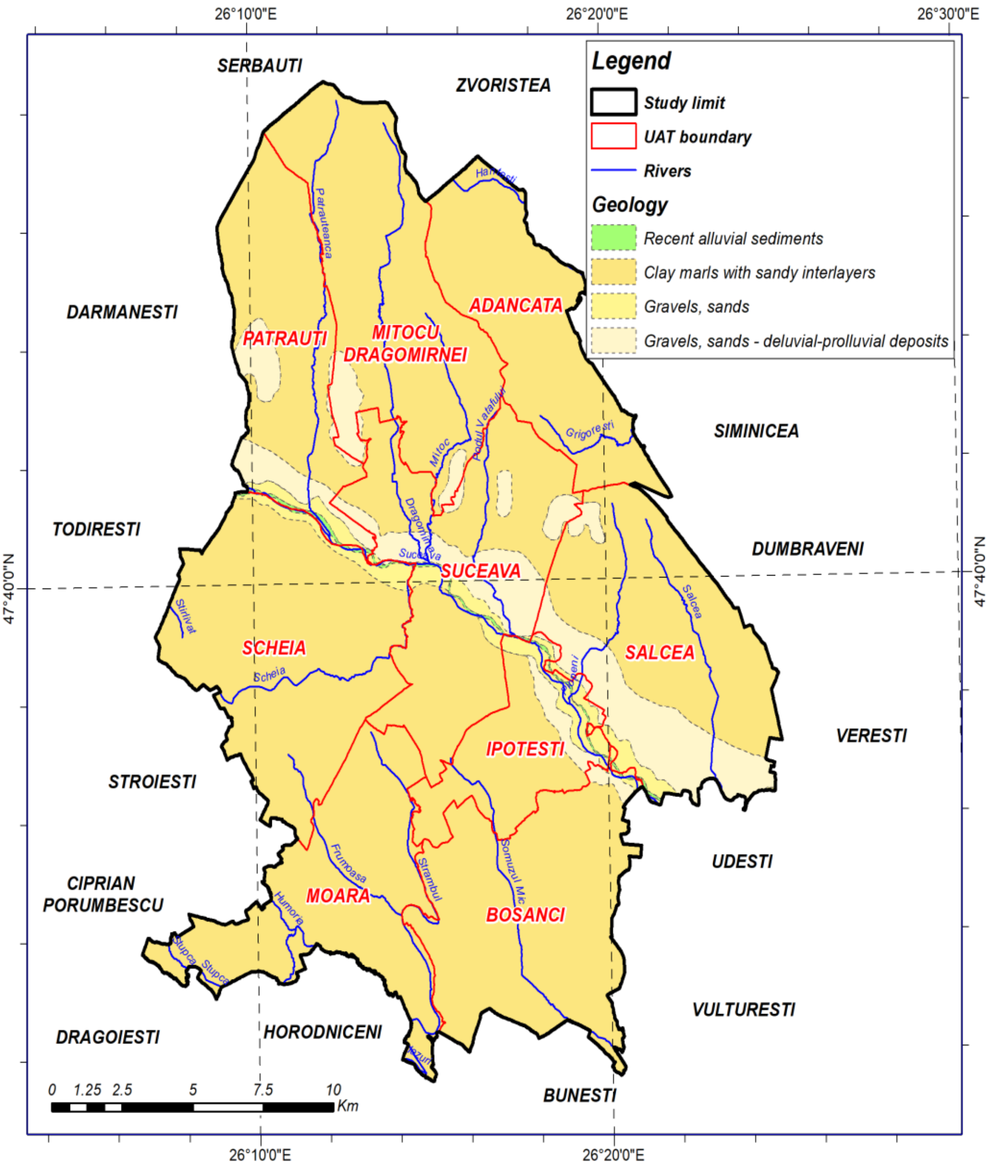


Figure 2 Geological map of Suceava Metropolitan Area.

The north ÷ north-west territory of the valley overlaps with a part of the Dragomirna Plateau, and the south ÷ south-west partially overlaps with the Falticeni Plateau. Both subunits belong to Suceava Plateau (Băcăuanu, 1983; Efros et al., 2004). The dominant landforms are interfluvial plateaus, generally smooth and gently sloping in the general NW-SE direction, separated by relatively deep valleys, bordered by slopes affected by ongoing geomorphological processes (landslides, linear and surface erosion).

The climate is transitional temperate (Ciulache, 2004), specific to the low hills and foothills (300-500 m). The westerly circulation is dominant (westerly and northwesterly winds account for 39.2% of the year), and there is calm air for 34.8% of the year. The average multiannual temperature is 7.9°C, and the average precipitation is 620.1 mm (Tănasă, 2011). The atmosphere over Suceava is undergoing a significant warming process (Mihăilă et al., 2017).

The analysed area is crossed by the Suceava River, whose multiannual average flow is 16.5 mc s^{-1}, but whose maximum flow reached 1946 mc s^{-1} on 27 July 2008 (Mihăilă et al., 2009). According to INS Romania, 186603 inhabitants live in the AMSv (456.9 km^{-2}).

3. Methods

The assessment of atmospheric radioactivity was based on self measurements data set as well as on data provided by the Environmental Protection Agency of Suceava. First part of this study, we present the field campaigns results between 15 and 17 July 2021, when measurements were made at 30 points in the AMSv. Their code and geographical coordinates are given in Table 1 and the positioning within the AMSv is shown in Figure 1.

Table 1 Geographical coordinates and radioactivity monitoring point code.

Site No.	Monitoring sites	Site code	Coordinates	
			Longitude (°E)	Latitude (°N)
1	Moara Bulai Lake	MLB	26° 14' 2.130"" E	47° 36' 43.949"" N
2	Lidl Obcini	LID	26° 14' 8.615"" E	47° 38' 25.779"" N
3	University Park	UPA	26° 14' 51.685"" E	47° 38' 25.475"" N
4	University Corp B	UCB	26° 14' 41.118"" E	47° 38' 28.038"" N
5	George Enescu Park	GEN	26° 14' 30.834"" E	47° 38' 44.261"" N
6	Sf. Nicolae Church	BSN	26° 15' 29.504"" E	47° 38' 46.295"" N
7	Mihai Eminescu College	CME	26° 15' 5.802"" E	47° 38' 58.012"" N
8	The Seat Fortress of Suceava	CSS	26° 16' 23.202"" E	47° 38' 39.595"" N
9	Landfill	GGS	26° 17' 13.655"" E	47° 39' 1.636"" N
10	Tișăuți bridge	TPD	26° 19' 6.602"" E	47° 37' 29.844"" N
11	Dedeman	DED	26° 16' 22.737"" E	47° 39' 58.930"" N
12	Acoperământul Maicii Domnului Church	AMD	26° 16' 42.692"" E	47° 40' 17.882"" N
13	Burdujeni village	BSP	26° 17' 35.711"" E	47° 41' 37.128"" N
14	Adâncata forest	PAD	26° 17' 56.370"" E	47° 42' 10.281"" N
15	Adâncata Girigan	ADG	26° 18' 1.999"" E	47° 44' 31.250"" N
16	Mitocu Dragomirnei Hall	PMD	26° 15' 16.751"" E	47° 43' 41.800"" N
17	Mitocu Dragomirnei Monastery	MMD	26° 13' 55.302"" E	47° 45' 26.657"" N
18	Lipoveni Lake	LDL	26° 13' 51.955"" E	47° 41' 45.486"" N
19	Lipoveni Community Center	CCL	26° 13' 33.364"" E	47° 43' 22.929"" N
20	Ițcani train station	GIT	26° 13' 53.445"" E	47° 40' 41.671"" N
21	Șcheia School	SCS	26° 13' 52.752"" E	47° 39' 21.148"" N
22	Bermas	BER	26° 13' 23.602"" E	47° 39' 1.630"" N
23	Zamca Monastery	ZAM	26° 14' 39.350"" E	47° 39' 10.522"" N
24	Secondary School 3	SCT	26° 15' 6.053"" E	47° 38' 44.799"" N
25	Wather station	SMS	26° 14' 32.226"" E	47° 37' 57.228"" N
26	Sainiuc home	CSA	26° 16' 25.938"" E	47° 37' 36.036"" N
27	Miron Costin School	SMC	26° 17' 1.108"" E	47° 40' 6.187"" N
28	The International Airport Ștefan cel Mare Suceava	SAL	26° 21' 1.106"" E	47° 41' 11.000"" N
29	Văratec	VAR	26° 24' 3.797"" E	47° 38' 26.428"" N
30	Văratec Spring	VIZ	26° 24' 22.810"" E	47° 36' 50.283"" N

Our aim was to perform measurements on the predominant active surface types of AMSv. These types include asphalt, concrete, stone pavement, stony or sandy soil, sandy soil, agricultural soils, forest soils and railway infrastructure.

From 15 to 17 July 2021, in AMSv the weather was beautiful, evolving into an anticyclonic regime. The atmospheric pressure values at sea level were 1008.8 hPa at Suceava Weather Station, 1009.4 hPa at SV1 and 1011.6 hPa at SV2 (SV1 and SV2 are automatic air quality monitoring stations belonging to the National Air Quality Monitoring Network). Total cloudiness values were low (3.1 tenths) and global radiation values were high (5174.4 W/mp at SV1, 4658.9 W/mp at SV2 - as daily mean values). No precipitation occurred. The wind speed was 2.4 m/s measured at anemometer height of the Suceava Weather Station, and in the city the wind speed was 0.2 m/s at SV1 and 0.4 m/s at SV2. Temperature values were high (average 25.3°C at the Suceava Weather Station, 25.6°C at SV1 and 26.3°C at SV2). Air humidity was low (68 % at the Suceava Weather Station, 56 % at SV1 and 57 % at SV2).

The measurements were carried out with the help of the ISU Suceava CBRN (chemical, biological, radiological, nuclear) truck, using a Berthold LB125 portable device for alpha, beta, and gamma radiation measurements. The calibration of the instrument was performed using a Cs-137 source positioned in front of the detector in the 2000 keV range. Two measurements (2 min) were made for each of the 30 points, one at ground level and the other at 1.8 m height, following a rigorous protocol of observations, which gives us the conviction that the accuracy of the determined values was very close to the maximum technical precision of the instrument.

A second, more detailed and comprehensive analysis was carried out on the basis of a database of the gamma dose rate absorbed in the air, obtained at 60-minute intervals after continuous monitoring in the period 2009 - 2020 by the automatic station belonging to the National Environmental Radioactivity Warning/Alerting System of the Environmental Protection Agency of Suceava (station coordinates lat: 47.617, long: 26.250). Global beta activity has also been determined by sampling atmospheric aerosols and atmospheric deposition. Suceava Environmental Radioactivity Monitoring Station (SSRM) carries out two daily atmospheric aerosol samplings, each lasting 5 hours, hourly ranged 02÷07, 08÷13, 14÷19, 20÷01, by aspiration on high retention efficiency filters, and performs global beta measurements of the filter retained aerosols, as followed:

- immediately after sampling (3 minutes after the end of sampling) - immediate measurements

- after 25 hours - for the determination of radon (Rn) and thoron (Tn);

- 5 days after the end of sampling - delayed measurements.

Atmospheric deposition samples are obtained by daily sampling of 0.3 m² sedimentable dust (dry deposition) and atmospheric precipitation (wet deposition). After sampling and preparation, the total deposition samples are measured on the same day for determining of the immediate global beta activity and 5 days after sampling for determinaing of the delayed global beta activity, according to the Order of the Ministry of Environment and Forests No. 1978/19.11.2010. Graphical and cartographic processing was done using Microsoft Office - Excel and ArcGis v.10.2.2. Inverse distance weighted (IDW) interpolation was used to make the absorbed gamma dose rate maps.

4. Results and discussion

4.1. Experimental field monitoring of the absorbed gamma dose rate

The results of the two days of experimental field monitoring of the absorbed gamma dose rate in air are shown in Figures 3 (a and b) and 4 (a and b).

Table 2 Warning, caution and alarm limits - immediate measurements (Order 1978 / 19.11.2010).

Environmental factor	Type of measurement	Limit		
		Warning	Alerting	Alarming
Air / atmospheric aerosols	Global beta activity (Bq m^{-3})	10	50	200
	External gamma dose rate ($\mu Sv\ h^{-1}$)	0,250	1,0	10
Atmospheric deposition	Global beta activity (Bq m^{-2} day^{-1})	200	1000	2000

Figure 3a shows an hourly momentary value resulting from 2-minute gamma dose values at ground level compared with the annual cumulative dose calculated for the same level. The annual cumulative gamma dose values were obtained by multiplying the hourly values by 24 hours in a day and 365 days in a year. The points where the radioactivity was higher are Adâncata, Dragomirna Monastery, Șcheia School, but these values do not exceed the 1 microSieverti year (mSv $year^{-1}$) threshold, so they do not endanger human health, the limit set by Order 1978 / 19.11.2010 (Table 2).

Due to specific characteristics of the soil (composition, structure, degree of pollution), the radiation concentration emitted by different types of active surfaces can vary considerably from one place to another. The radiation dilution concentration in soil is directly proportional to the above-ground surface height (at 5 cm height as an average of the points where measurements were made, a radiative flux of 0.060 µSv/h and at 1.8 m height of 0.049 µSv/h resulted). Measurements taken at 1.8 m level reflect slightly lower values than at ground level due to the scattering effect of vertical air currents or wind (Figure 4) (Moore at al., 1973).

According to the category of active surfaces, the radiation flux was 0.125 µSv/h at 5 cm and 0.090 µSv/h at 1.8 m above the surface covered with natural granite (pedestrian infrastructure around the Dragomirna Monastery). Ploughed, cultivated, and forested soils were in second place in terms of radioactivity (0.062 µSv/h at 5 cm and 0.0492 µSv/h at 1.8 m). This was followed by sandy soils (0.061 µSv/h at 5 cm; 0.049 µSv/h at 1.8 m), paved surfaces (0.046 µSv/h at 5 cm; 0.040 µSv/h at 1.8 m), stony soils (0.045 µSv/h at 5 cm; 0.035 µSv/h at 1.8 m), and asphalted surfaces (0.040 µSv/h at 5 cm and 0.039 at 1.8 m).

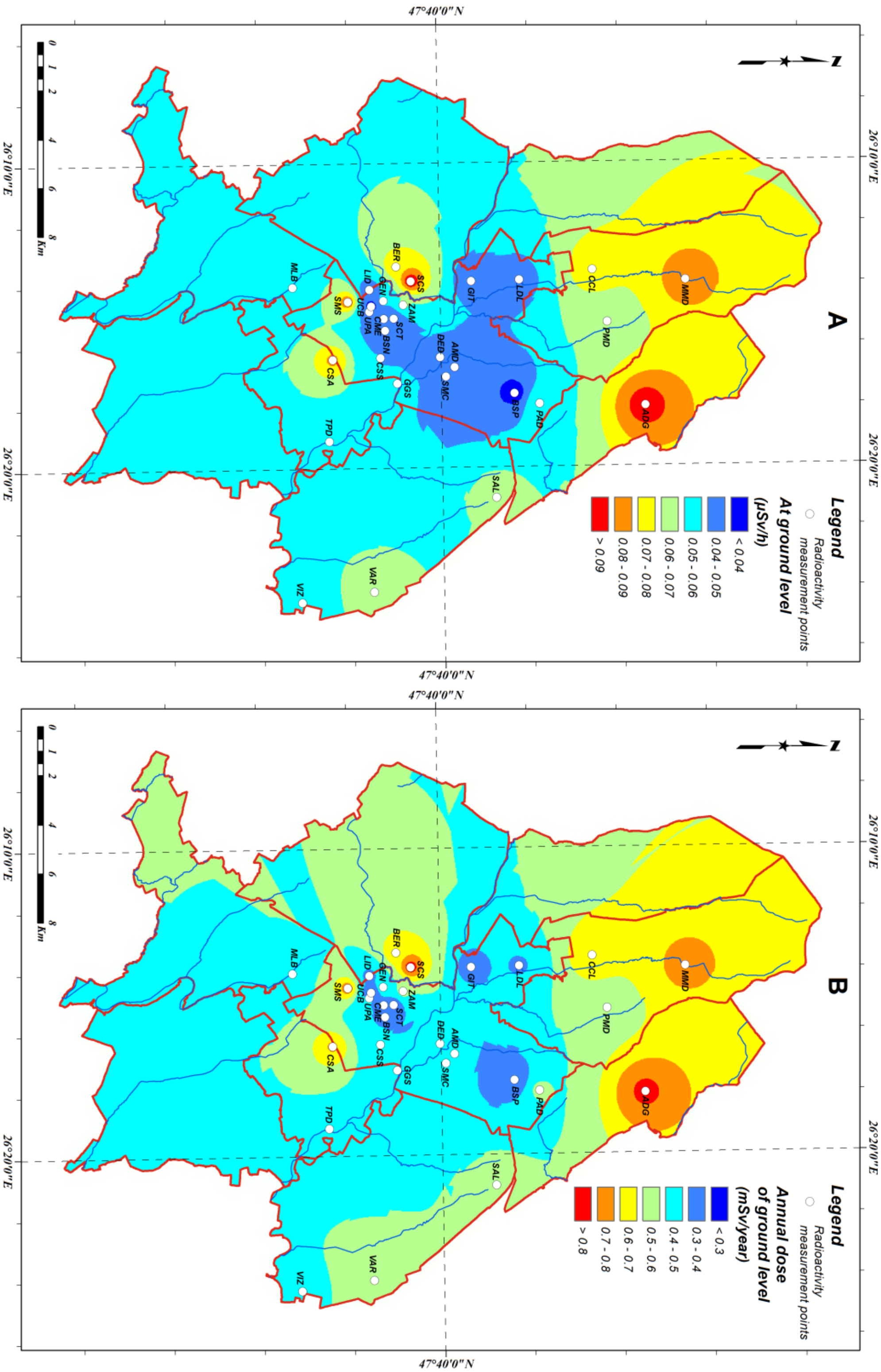


Figure 3 Absorbed gamma dose rate at ground level (µSv h^{-1}) - A, measured on 15 and 17 July 2021; simulated calculations for an average year were based on these measurements (mSv $year^{-1}$) -B.

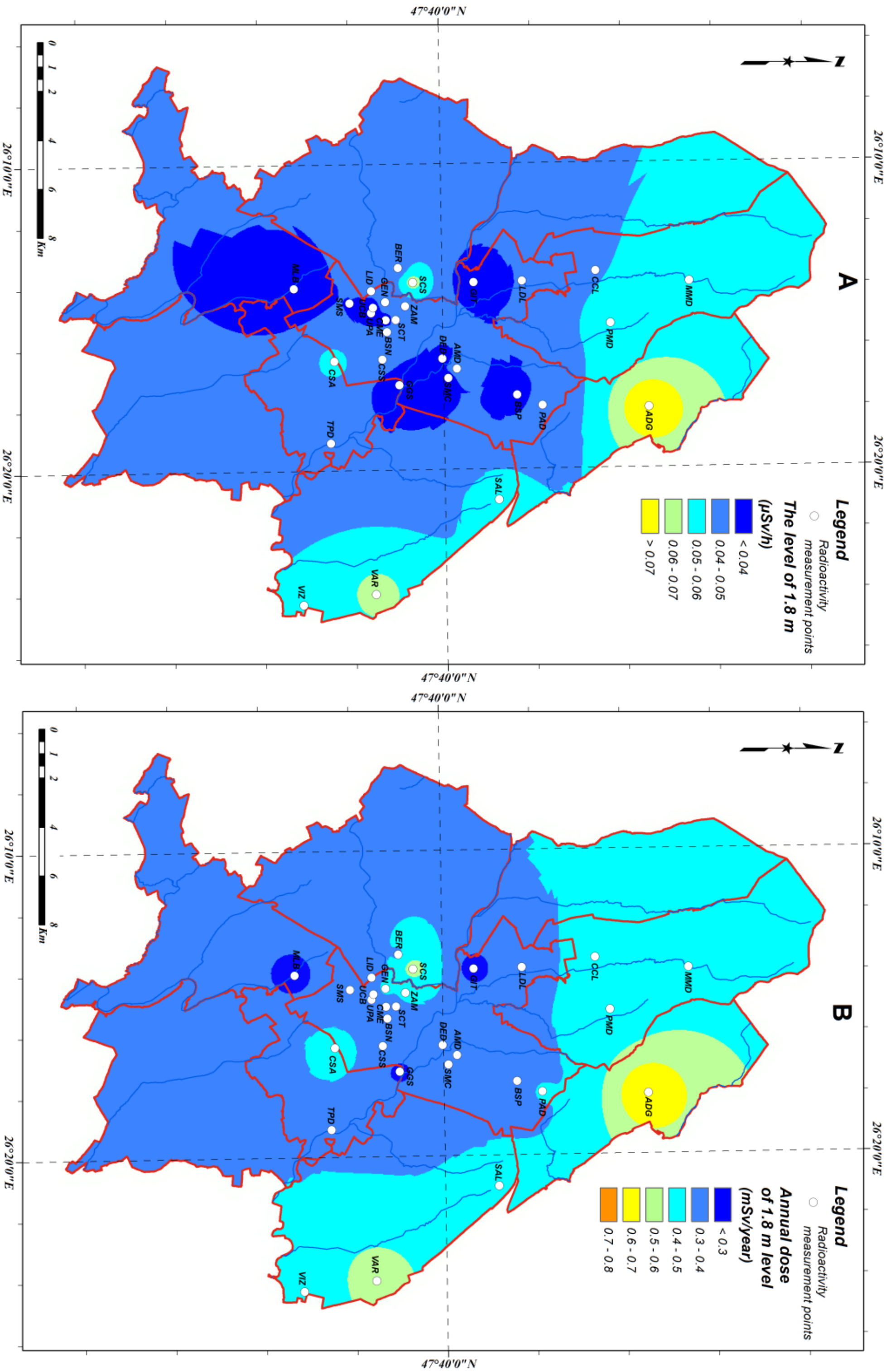


Figure 4 Absorbed gamma dose rate at 1.8 meters level (µSv h^{-1}) - A, measured on 15 and 17 July 2021; simulated calculations for an average year were based on these measurements (mSv $year^{-1}$) -B.

4.2. Assessment of atmospheric radioactivity based on institutional measurements

4.2.1. In the air absorbed gamma dose

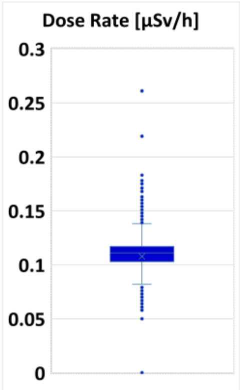


Figure 5 Boxplot representation of hourly gamma dose values in Suceava (2009-2020).

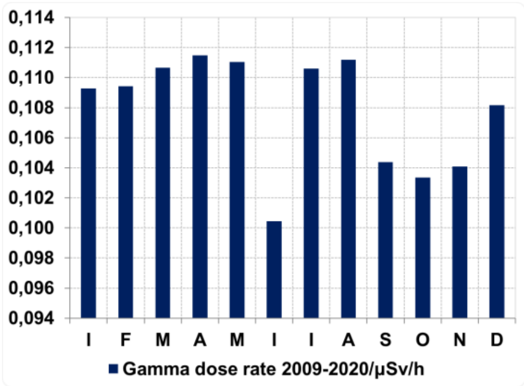


Figure 6 Monthly average gamma dose rate absorbed in the air determined on the basis of measurements in the specialized laboratory of the Suceava Environmental Protection Agency in the period 2009-2020.

The entire hourly gamma dose database (over 85000 determinations) is summarised in figure 5. Analyzing the Suceava Environmental Protection Agency specialized laboratory database between 2009-2020, we found that atmospheric gamma dose rate recorded a maximum of 0.261 µSv h^{-1}, thus exceeding the warning limit. The annual averages ranged from 0.53 mSv in 2012 to 0.97 mSv in 2019.

Monthly rated, the lowest gamma dose level value has been recorded in June with values up to 0.102 µSv h^{-1}, the values of this parameter were between 0.103 and 0.105 µSv h^{-1} in September - November, and in the remaining months of the year values were ranged at 0.108 - 0.112 µSv h^{-1} (Figure 6).

The lowest hourly values were recorded in June. Values up to 0.105 µSv h^{-1} also characterise the months of September, October, November (Figure 7).

	1	2	3	4	5	6	7	8	9	10	11	12
00:00	0,109	0,110	0,112	0,111	0,112	0,100	0,110	0,112	0,104	0,103	0,104	0,108
01:00	0,109	0,109	0,110	0,111	0,110	0,100	0,110	0,111	0,104	0,104	0,104	0,107
02:00	0,110	0,109	0,111	0,112	0,111	0,100	0,111	0,111	0,104	0,104	0,105	0,109
03:00	0,109	0,109	0,110	0,111	0,111	0,101	0,110	0,112	0,105	0,103	0,103	0,108
04:00	0,109	0,109	0,113	0,112	0,111	0,100	0,111	0,113	0,105	0,103	0,104	0,107
05:00	0,109	0,109	0,112	0,112	0,110	0,101	0,111	0,112	0,105	0,103	0,105	0,108
06:00	0,111	0,110	0,111	0,114	0,111	0,101	0,111	0,112	0,105	0,104	0,104	0,108
07:00	0,110	0,110	0,111	0,111	0,111	0,100	0,110	0,111	0,106	0,104	0,103	0,108
08:00	0,109	0,110	0,111	0,112	0,110	0,101	0,110	0,111	0,105	0,103	0,104	0,107
09:00	0,109	0,109	0,110	0,112	0,110	0,101	0,111	0,111	0,105	0,103	0,105	0,108
10:00	0,109	0,110	0,110	0,112	0,111	0,101	0,111	0,112	0,104	0,103	0,104	0,108
11:00	0,109	0,109	0,110	0,111	0,111	0,100	0,110	0,111	0,104	0,104	0,104	0,108
12:00	0,108	0,109	0,111	0,111	0,110	0,101	0,111	0,110	0,103	0,103	0,104	0,108
13:00	0,109	0,110	0,110	0,112	0,111	0,099	0,111	0,110	0,104	0,104	0,104	0,109
14:00	0,110	0,111	0,109	0,112	0,110	0,101	0,111	0,112	0,104	0,102	0,104	0,109
15:00	0,110	0,109	0,110	0,111	0,112	0,101	0,111	0,111	0,105	0,104	0,104	0,109
16:00	0,109	0,109	0,110	0,111	0,112	0,100	0,110	0,111	0,104	0,103	0,105	0,108
17:00	0,109	0,110	0,110	0,110	0,113	0,100	0,110	0,110	0,105	0,103	0,104	0,109
18:00	0,108	0,109	0,110	0,110	0,111	0,101	0,112	0,110	0,105	0,103	0,105	0,108
19:00	0,109	0,109	0,111	0,112	0,112	0,101	0,112	0,112	0,103	0,103	0,104	0,109
20:00	0,110	0,109	0,111	0,111	0,111	0,101	0,111	0,111	0,105	0,103	0,103	0,109
21:00	0,110	0,109	0,111	0,111	0,111	0,101	0,110	0,111	0,105	0,103	0,103	0,108
22:00	0,110	0,109	0,110	0,111	0,111	0,101	0,111	0,111	0,103	0,104	0,104	0,108
23:00	0,109	0,109	0,111	0,111	0,111	0,101	0,111	0,111	0,104	0,104	0,104	0,108

Figure 7 Hourly gamma dose rate absorbed in the air (µSv h^{-1}) determined on the basis of measurements at the specialized laboratory of the Suceava Environmental Protection Agency in the period 2009-2020.

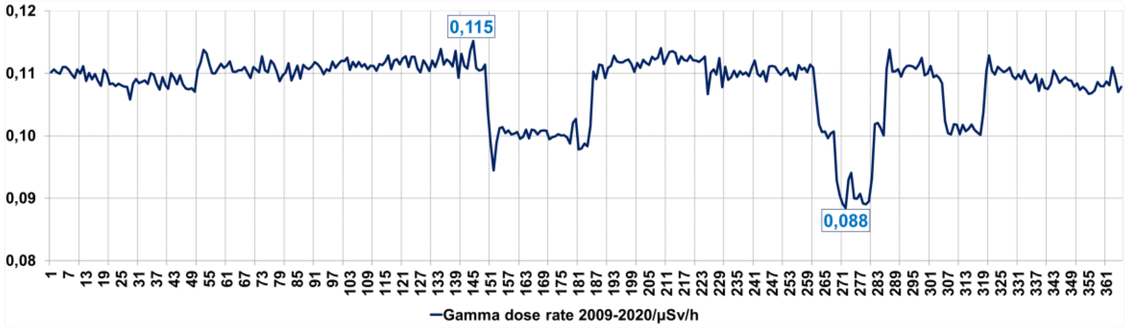
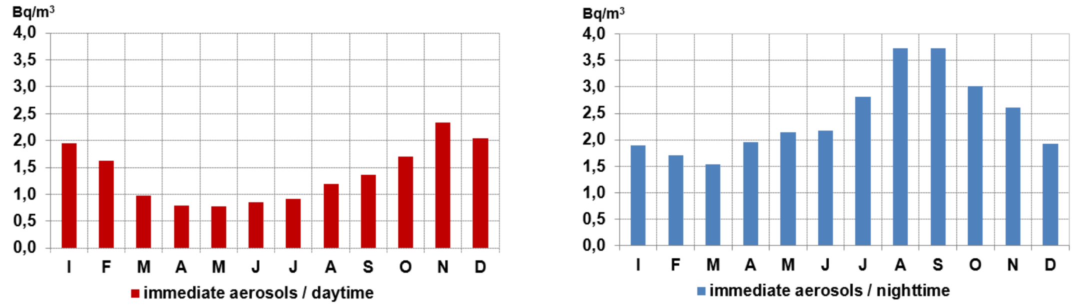


Figure 8 Interdiurnal regime of hourly average values calculated for the days in an average year of the gamma dose rate absorbed in the air (µSv h^{-1}), determined on the basis of measurements in the specialized laboratory of the Suceava Environmental Protection Agency in the interval 2009-2020.

A careful interdiurnal regime analysis of the average hourly values calculated for all days of the year of the absorbed gamma dose shows that on most days of the year (more than 2/3) it oscillates with small positive or negative deviations around the threshold of 0.11 µSv h^{-1} (Figure 8). It should be noted that on certain groups of days in one year there is a number of decreases in the hourly averages of gamma dose, which can be associated w excess rainfall (for the month of June) episodes or increased atmospheric pressure (days in October and November). Although these large decreases appear on the graphical representation scale, they are in fact low in absolute value (maximum 0.01 µSv h^{-1}).

4.2.2. Global beta measurements of atmospheric aerosols

The immediate daytime global beta activity measurements ranged from 0.1 to 8.9 Bq m^{-3}. From figure 9 a it is to be noticed that there is a seasonal variation in the immediate measured global beta radioactivity. Immediate global beta activity measurements of atmospheric aerosols show that the highest daytime values occur in autumn-winter months, when aerosol dispersion conditions at the surface are hindered by low atmospheric dynamics and the presence of stable liquid aerosol layers forming haze or fog in the lower atmosphere.



a b

Figure 9 Variation of monthly averages of the immediate global beta activity of atmospheric aerosols in Suceava (2009-2020).

The immediate nighttime global beta activity ranged from 0.2 to 9.9 Bq m^{-3}. The time series of global beta activity parameters is summarized in figure 10.

Diagrams 1 and 2 show the obvious difference between immediate nightime global beta activity and during the day (decreased in value by 30-40%, but at the same time more variable), compared to delayed global beta activity (diagrams 3 and 4) which is higher in value, but has a more attenuated temporal variability. The highest values of the immediate nighttime global beta activity are recorded in July-November interval (Figure 9b). Very frequently cold nights thermal inversions followed by very warm days in July-November interval explain the accumulation of aerosols at ground level. In August and September the day/night thermal contrast is maximum, as well as night thermal inversions intensity, which explains the production of highest air aerosol concentrations at the bottom of the atmosphere (microclimatic layer) during these nights.

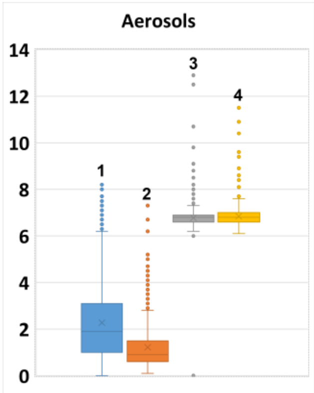
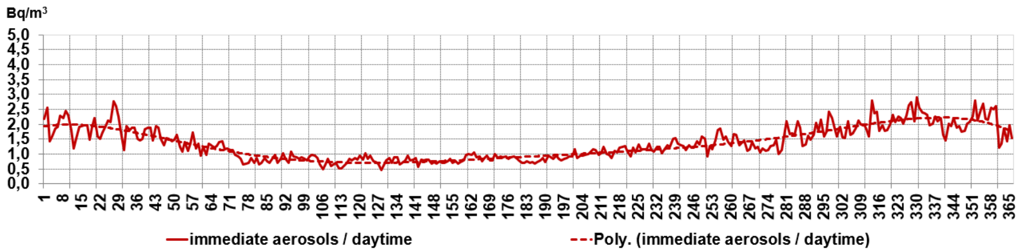


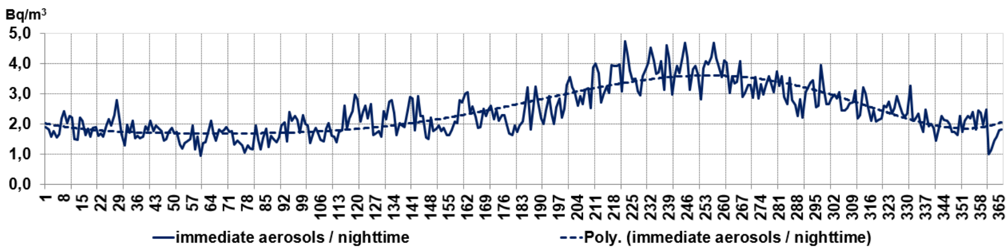
Figure 10 Value representation (in Bq m^{-3}) of atmospheric aerosol radioactivity (half-day values) by boxplots: 1- immediate night aerosols, 2 - immediate day aerosols, 3 - delayed night aerosols, 4 - delayed day aerosols.

From figure 11a and b its noticeable that there is a clear interdiurnal variation in the immediate global radioactivity. The patterns of daytime and nighttime regime of this parameter (Figures 11a and 11b) follow those of the intermonthly regime (Figures 9a and 9b), but are much more detailed.

During the analysed period, delayed global beta activity measured during the day was between 6.1 and 41.2 Bq m^{-3}, exceeding the warning threshold of 10 Bq m^{-3} on 46 days, the maximum being recorded on 13 December 2012. During the night, the measured values ranged from 6.0 to 104.6 Bq m^{-3}, exceeding the alert threshold on 4 days, the maximum being recorded on 1 October 2017.



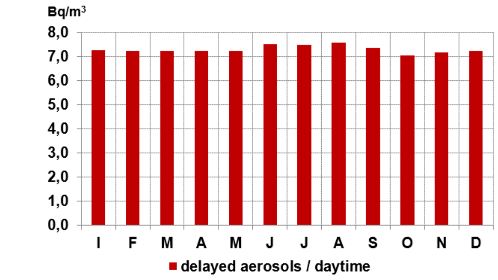
a



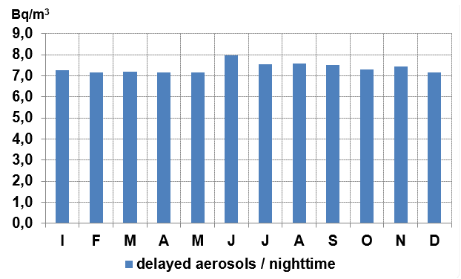
b

Figure 11 Variation of daily averages of immediate global beta activity of atmospheric aerosols in Suceava (2009-2020).

In the case of the delayed daytime and delayed nighttime global beta activity determinations there is a flattening of the intermonthly (Figures 12a and 12b) and interdiurnal (Figures 13a and 13b) variability.

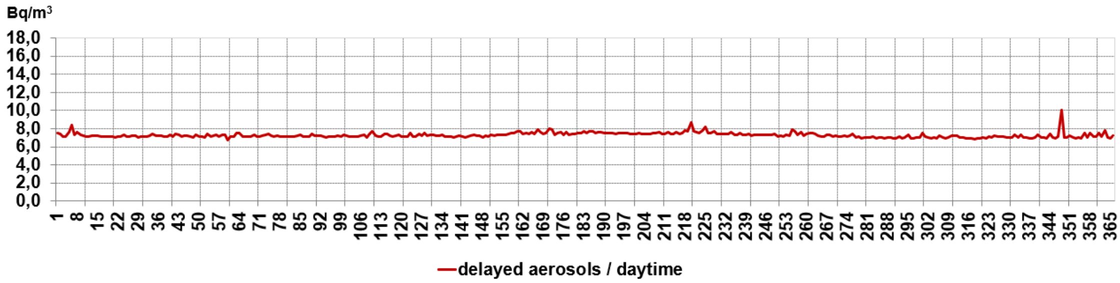


a

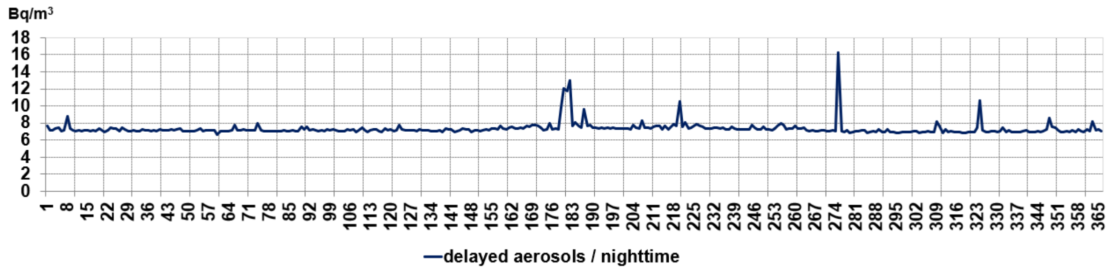


b

Figure 12 Variation of the monthly averages of the global mean beta activity of atmospheric aerosols in Suceava (2009-2020).



a



b

Figure 13 Variation of daily averages of global delayed beta activity of atmospheric aerosols in Suceava (2009-2020).

In the context of this flattening, however, we note an increase in the total global beta activity, measured on daytime or nighttime in summer months (Figures 12a and 12b) and although less obvious than for months, for days corresponding to these months (Figures 13a and 13b).

The annual diurnal and nocturnal regime is very attenuated, taking the form of an almost straight line (with a slight convexity in the summer days), with some peaks due to the increase aerosols concentration, and therefore their short intervals radioactivity in the atmosphere.

4.2.3. Total atmospheric depositions

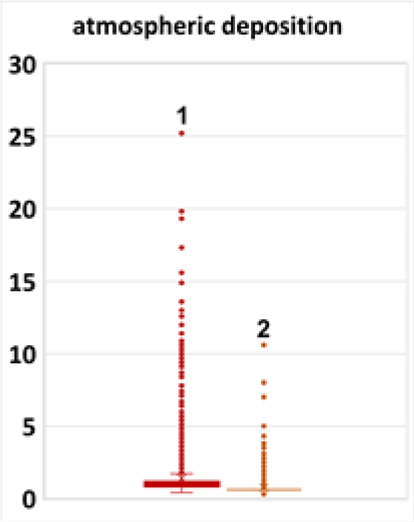


Figure 14 Value representation (in Bq m^{-3}) of the radioactivity of the deposition (diurnal values) by boxplots: 1: immediate atmospheric deposition, 2: delayed atmospheric deposition.

The global beta activity of daily atmospheric deposition measured immediately recorded values between 0.4 and 61.3 Bq m^{-3}. In figure 14 of Dataset 1, we have removed, for better graphical visibility, 4 outliers of the global beta activity of daily atmospheric deposition. We observe the massive clustering of most of the values composing the series around the middle quartile, with the exception of some outliers (quite significant numerically) placed above quartile 3.
Delayed atmospheric deposition ranged from 0.3 to 10.6 Bq m^{-3}. Based on time series analysis of global beta activity we identified that the maximum hourly values recorded for both immediate and delayed deposition were well below the warning threshold of 200 Bq m^{-3} or the alert threshold of 1000 Bq m^{-3}.

For the monthly average values of immediate/delayed deposition, it is observed that in May-July there is a maximum related to the dynamization of social-economic activities in the area of representativeness of the monitoring point (Figure 15 a and b).

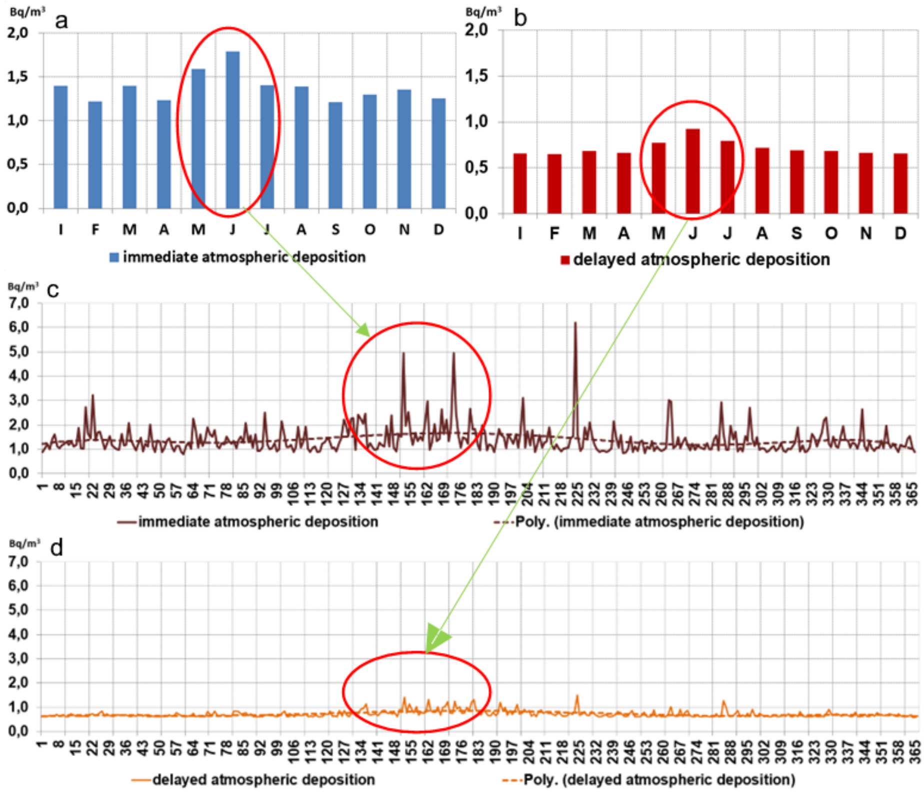


Figure 15 Variation of monthly / daily averages of immediate / delayed atmospheric deposition in Suceava (2009-2020).

We observe that, due to the decreasing values of delayed deposition at the intermonthly and interdiurnal levels of analysis, the interdiurnal variability of delayed deposition decreases significantly, especiall (Figures 15 c and d).

4.2.4. Radon and Thoron emissions

Knowing the levels of radon and thoron emissions is an important objective for European countries, as radon is one of the major contributors to natural irradiation of the population. Radon is a naturally occurring radioactive gas found in earth's crust that cannot be detected by humans. Thoron is another gas with the same effects on the human body as radon. According to the World Health Organisation, the risk of lung cancer increases by about 16% per 100 Bq/m3 increase in long time average radon concentration. The worldwide average annual radiation dose from exposure due to naturally occurring radiation sources, including radon, is 2.4 mSv. In anylarge population, about 65% would be expected to have annual doses of between 1 and 3 mSv (UNSCEAR, 2000). These gases enter the atmosphere as a result of soil and rocks emanations, where they are subject to atmospheric dispersion phenomena.

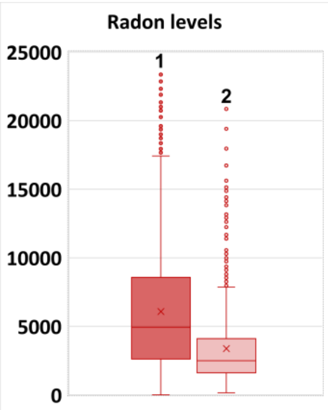


Figure 16 Value representation (in mBq m^{-3}) of radon radioactivity (half-day values) by boxplots: 1: night-time radioactivity, 2: day-time radioactivity

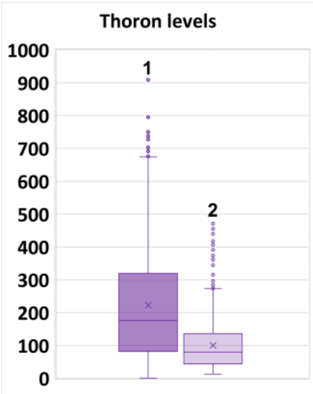


Figure 17 Value representation (in mBq m^{-3}) of thoron radioactivity (half-day values) by boxplots: 1: night-time radioactivity, 2: day-time radioactivity

Daytime radon concentrations ranged from 271.9 to 25837.9 mBq m^{-3} and nighttime values from 156 to 28350.4 mBq m^{-3}. For thoron, the daytime minimum was 6.5 mBq m^{-3} and the maximum was 670.6 mBq m^{-3}. During the night the minimum was 17.6 mBq m^{-3} and the maximum 1109.7 mBq m^{-3}.

Figures 16 and 17 show the half-day radon and thoron levels at Suceava, with the exception of 8 positive radon outliers, which are not plotted for reasons of better visibility. However, they were not excluded from the statistical calculations and the construction of the diagrams.

Both radon and thoron show lower concentrations during the day and in spring-summer (Figure 18 a and 18 b), when atmospheric diffusion is enhanced by conventional processes. The highest radon/thoron concentrations in the atmosphere occur by night, towards the morning, in autumn-winter months (Figures 18 a and 18 b), because the aerosol dispersion at the ground surface has the lowest parameters, during which there are frequent situations of calming/reversal of air streams. Similar results were obtained by Cosma (2005) in a study carried out in Bucharest between 9 May 1996 and 30 April 1999.

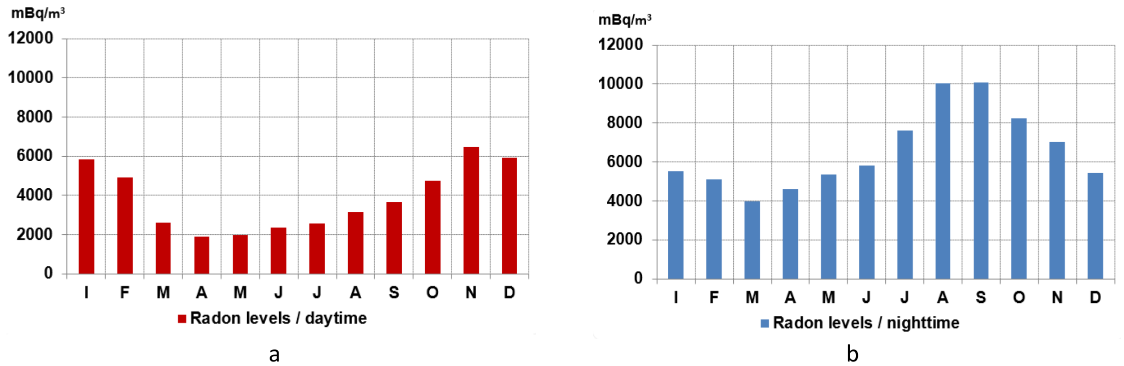


Figure 18 Monthly average radon variation in Suceava (2009-2020).

The high concentrations of these gases are enhanced by high relative humidity, hazy air intervals and haze by facilitating the attraction of radon molecules to aerosol particles. Increased daytime temperatures (especially in summer) lead to convection currents that contribute to the dispersion of radon and thoron accumulated overnight in the atmospheric lower layers (Figure 18). Radon and thoron concentrations in the atmosphere are influenced by turbulent diffusion, radioactive decay and removal processes such as dry deposition, rain and flushing (Baciu, 2005).

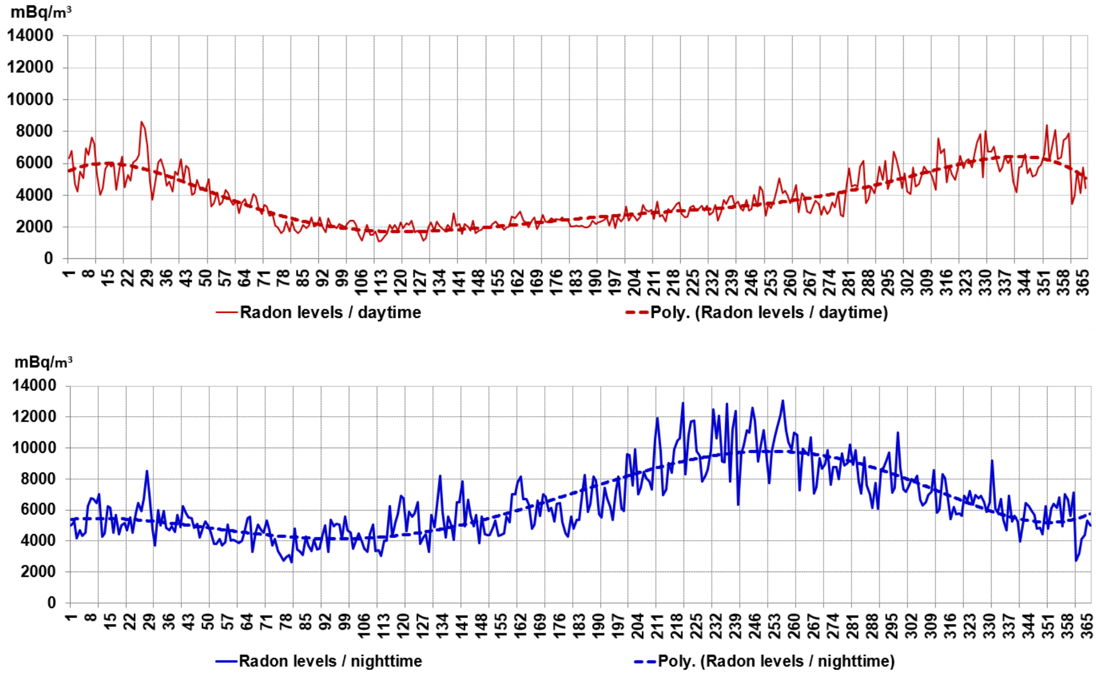


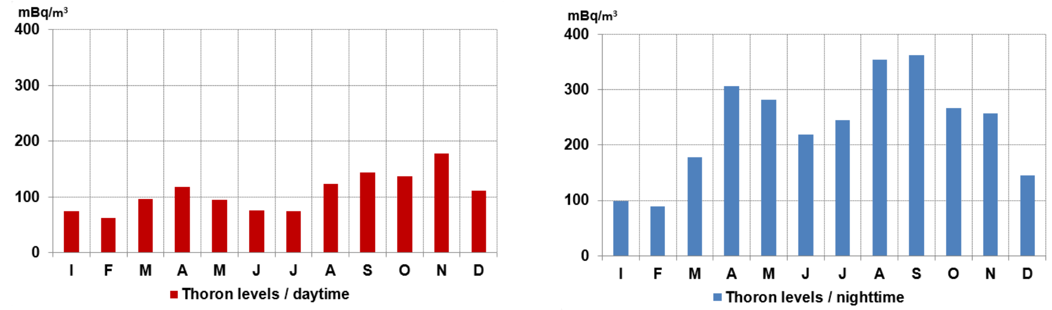
Figure 19 Daily average radon variation in Suceava (2009-2020).

The patterns of mean daytime interdiurnal variability (Figure 19a) and nighttime (Figure 19b) radon concentrations are consistent with the patterns of monthly daytime annual regime (Figure 18a) and nighttime values (Figure 18b) values and show that on most days and nights in spring and early summer months, when thermoconvection processes reach their annual maximum, the risk of human exposure to high radon concentrations is lower. On cloudy winter days with stratiform clouds, fog and atmospheric stability, radon concentrations increase (Figure 19a). On late summer and autumn nights (Figure 19b), when anticyclonic baryonal fields cause downward air movement and compression of pollutants in the lower layer of the atmosphere, the risk of exposure to high radon concentrations is higher (Figure 19b).

Summing up the average concentrations of radon and thoron in the Suceava atmosphere for the period 2009-2020, it can be noticed that by night radon accounts for 96.6% and thoron for 3.4% of these radioactive gases, while during the day radon accounts for 97.2% and thoron for 2.8%. These statistics clearly show that radon is a more important atmospheric pollutant than thoron.

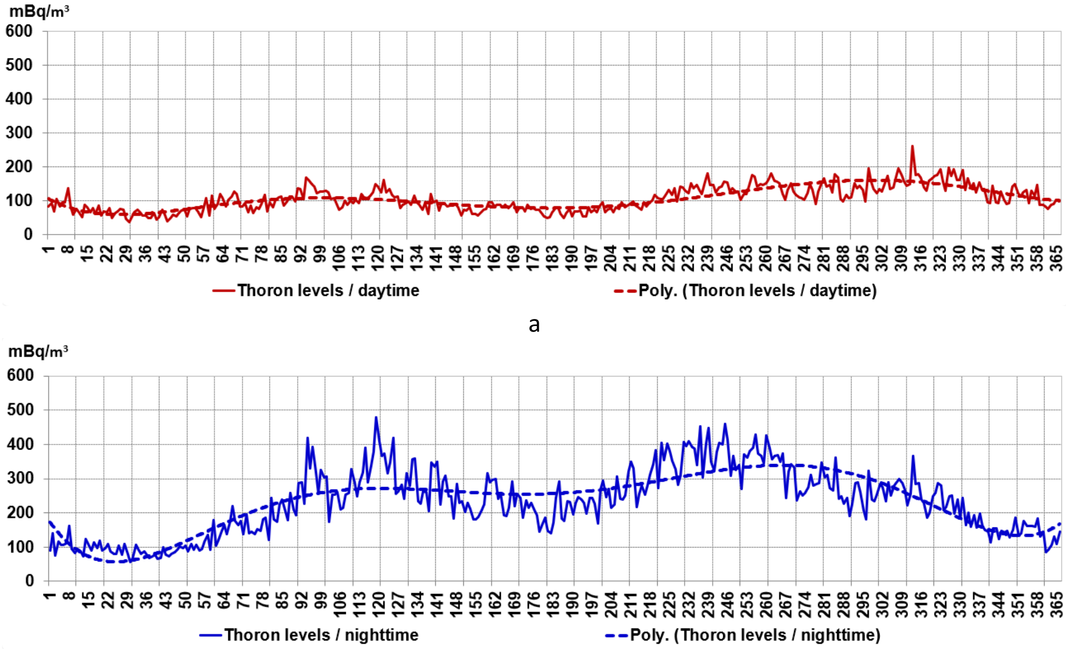
For both monthly and daily values, thoron concentrations in the atmosphere are significantly higher at night than during the day, regardless of the monthly (Figure 20 a and b) or diurnal (Figure 21 a and b) unit to which we refer.

Thus, for both day and night observations, thoron shows an annual regime which, based on monthly (Figure 20 a and b) and diurnal (Figure 21 a and b) values, outlines the same peak, trough or inflection aspects: a main summer-autumn maximum (August-November) and a secondary spring maximum (March-May), separated by the main winter minimum (January-February) and the secondary summer minimum (June-July).



a b

Figure 20 Variation of monthly averages of thoron in Suceava (2009-2020).



b

Figure 21 Variation of daily averages of thoron in Suceava (2009-2020).

During the warm season of the year, thermoconvection generally favours the release of thoron from the soil, whereas during the cold season, frost and snow cover on the soil surface limit the reach of thoron into the microclimatic layer. The lower humidity in April - early May and the more active atmospheric circulation favour the thoron peak in these months. In the second half of May and June, the wettest period of the year, the atmosphere is washed out and wet soils do not favour thoron emissions. In August-September, the predominance of anticyclonic baric fields causes the phenomenon of drought, which affects the soils, causing them to crack; the soil cover is strongly heated and thoron reaches higher concentrations in the drier atmosphere. In December-February, the thermal and snow conditions do not favour the release of thoron from the soil into the overlying atmosphere.

5. Conclusion

Field measurements included in the mini-campaign between 15 and 17 July 2021 showed some differentiation in the levels of radioactivity on the ground and in the air in Suceava. The spatial variability of the values is within the limits that do not endanger human health. Gamma dose values on the ground were slightly higher (by 0.01-0.02 µSv h-1) than at the 1.8 m level.

The obtained results in the case of external gamma dose, through the analysis of hourly data, show that during the monitoring period (2009-2020) the warning threshold of 0.250 µSv h-1 was exceeded in Suceava by only 0.011 µSv h-1. On the basis of cumulated annual data, the external gamma dose rate was between 0.53 mSv in 2012 and 0.97 mSv in 2019, i.e. it did not exceed the threshold of 1 mSv, the level at which health problems can occur.

For atmospheric aerosols, the warning threshold of 10 Bq m^{-3} was not exceeded in any case, and for atmospheric deposition (on the ground) the observed maximum rose to 61.3 Bq m-3, less than 1/3 of the warning threshold of 200 Bq m^{-3}.

The assessment of radon and thoron emissions showed a maximum of 28.4 Bq m^{-3}, a quarter of the human health threshold of 100 Bq m^{-3}. Starting from the premise that radon and thoron emissions summed up to 100%, analyzing the averaged radon and thoron concentrations over the period 2009-2020, we found that radon accounts for about 97%, and thoron for about 3% of the emissions, demonstrating the more important role played by radon in the radioactive emissions reaching the atmosphere of AMSv. The mean radon and thoron concentrations in the 2009-2020 period show that radon accounts for about 97% and thoron for about 3%, which demonstrates the more important role played by radon in the process of air pollution in Suceava.On cloudy winter days with stratiform clouds, fog and reduced atmospheric dynamics, radon concentrations increase. The radon concentration in the open atmosphere increases especially in the late summer and autumn nights, when the anticyclonic baric fields force the dynamic descent with the compression of the pollutants in the lower layer of the atmosphere. In these specific situations, the risk of exposing the body to high radon concentrations is higher.

After analysing the results, we can say that the population of the area studied enjoys optimal living conditions in the open atmosphere, which are not endangered by the values of the air quality indicators studied in this study. Based on this existing reality in the free atmosphere of AMSv a next step in the research will include the monitoring of radon emissions from indoor air (individual dwellings, public institutions).

References

Appleton, J.D., Kendall, G.M., 2022. Gamma-radiation levels outdoors in Great Britain based on K, Th and U geochemical data. *Journal of Environmental Radioactivity*, 251–252,. https://doi.org/10.1016/j.jenvrad.2022.106948.

Baciu, A.C. 2006. Outdoor absorbed dose rate in air in relation to airborne natural radioactivity and meteorological conditions at Bucharest (Romania). *Journal of Radioanalytical and Nuclear Chemistry*, 268, 3-14. https://doi.org/10.1007/s10967-006-0116-7.

Bahnarel, I., Coreţchi, L. 2011. Contribuţii la monitorizarea radioactivităţii mediului. *Akademos*, Nr.1 (20).

Băcăuanu, V. 1983. *Geografia României, I.* Editura Academiei Române, București.

Burghele, B., Cosma, C. 2012. Intercomparison between radon passive and active measurements and problems related to thoron measurements. *Romanian Journal of Physics*, 58, 56–61.

Burghele B.D., 2013. Thoron contribution to the natural irradiation of the Romanian population and problems related to thoron and radon measurements. Ph.D. thesis, Babeș-Bolyai University of Cluj Napoca.

Ciulache, S., 2002. *Meteorologie şi Climatologie*. Editura Universitară, Bucureşti.

Cosma, C., Cozar, O., Jurcut, T., Baciu, C., Pop, I., 2005 Simultaneous measurement of radon and thoron exhalation rate from soil and building materials, Mc Laughlin, J. P., Simopoulos, S.E,. Steinhausler, E. The natural radiation environment, Elsevier, 699-705.

Cosma, C., Papp, B., Moldovan, M., Cosma, V., Cindea, C., Suciu, L., Apostu, A. 2010. Measurement of radon potential from soil using a special method of sampling. *Acta Geophysica,* 58, 947-956.

Cosma, C., Cucoș-Dinu, A., Papp, B., Begy, R., Sainz, C. 2013. Soil and building material as main sources of indoor radon in Băița-Ștei radone prone area (Romania). *Journal of Environmental Radioactivity*, 116, 174-179. http://dx.doi.org/10.1016/j.jenvrad.2012.09.006.

Cosma, C.; Papp, B.; Cucoș, A.; Sainz, C., 2015. Testing radon mitigation techniques in a pilot house from Baița-Ștei radon prone area (Romania). Journal of Environmental Radioactivity, 140, 141–147.

Cuculeanu, V., Simion, F., Simion, E., Geicu, A. 2011. Dynamics, deterministic nature and correlations of outdoor (222)Rn and (220)Rn progeny concentrations measured at Bacău, Romania. *Journal of Environmental Radioactivity,* 102, 703-712, https://doi.org/10.1016/j.jenvrad.2011.04.005.

Dolha, M., Timar-Gabor, A., Cameniță, A.,Costin, D. and Cosma, C. 2013. Gamma background measurements by TL method: applications in locations with varied geological context. Carpathian. *Journal of Earth and Environmental Sciences*, 8 (4), 109–114.

Dolha, M., Timar-Gabor, A., Dicu, T., Begy, R., Anton, M., Cosma, C. 2014. A high-resolution map of gamma dose rates in Cluj County, Romania using LiF: Mg, Cu, P detectors. *Radiation Protection Dosimetry*, 162, 14–19, doi:10.1093/rpd/ncu209.

Encian, I., Cosma, C., Damian, G., 2005. Studii asupra radonului și toronului din aerul atmosferic în Municipiul Bistrița, în perioada 1994-1997. *Environment & Progress,* 3, 155-159.

Kendall, G.M., Wakeford, R., Athanson, M., Vincent, T.J., Carter, E.J., McColl, N.P., Little, M.P. 2016. Levels of naturally occurring gamma radiation measured in British homes and their prediction in particular residences. *Radiation and Environmental Biophysics*, 55, 103–124.

Kendall, G.M., Chernyavskiy, P., Appleton, J.D., Miles, J.C.H., Wakeford, R., Athanson, M., Vincent, T.J., McColl, N.P., Little, M.P. 2018. Modelling the bimodal distribution of indoor gamma-ray dose-rates in Great Britain. *Radiation and Environmental Biophysics*, 57 (4), 321–347. https://doi.org/10.1007/s00411-018-0752-7 .

Mihăilă, D., Bostan D.C., Tanasă I. 2009. Les précipitations abondantes de l'ouest d'Ukraine et du nord de Moldavie enregistrées dans la période 23-28 juillet 2008. Causes et Conséquences” Geographia Technica, pag. 317 - 322, Numero special dedie au XXII - eme Colloque de L Association Internationale de Climatologie ,,Extremes Climatiques: genese, modelisation et impact”, Cluj University Press http://technicalgeography.org/pdf/sp_i_2009/gt_sp2009. pdf

Mihăilă, D., Bistricean P.I., Lazurca L. G., Briciu A.E. 2017. Climatic water deficit and surplus between the Carpathian Mountains and the Dniester River (1961–2012). *Environmental Monitoring and Assessment,* 189: 545, DOI 10.1007/s10661-017-6253-3.

Moore, H.E., Poet, S.E., Martell, E.A., 1973. 222Rnl 210Pbl ‘loBi and Po profiles and aerosol residence times versus altitude. *Journal of Geophysical Research*, 78(30), 7065-7072.

Efros, V., Popescu D. A., Popescu L. G. 2004. Geologia, exploatarea şi valorificarea nivelelor de gresii din zona Adâncata şi implicaţiile asupra utilizării terenurilor, *Analele Universităţii „Ştefan cel Mare” Suceava,* 17-24.

Ordinul Ministerului Mediului și Pădurilor nr. 1978 din 19.11.2010 privind aprobare Regulamentului de organizare și funcționare a Rețelei Naționale de Supraveghere a Radioactivității Mediului

Ouseph, P.J. 1975. Laboratory Instrumentation and techniques. Volume 2: Introduction to Nuclear Radiation Detectors. Plenum Press, New York and London.

Papp, B., Szakács, A., Néda, T., Papp, Sz. Cosma, C. 2010. Soil radon and thoron studies near the mofettes at Harghita Bai (Romania) and their relation to the field location of fault zone, *Geofluids,* 10, 586-593.

Papp, B., Cucoş (Dinu), A., Cosma, C., 2017. A comprehensive study of residential, geogenic and water radon in the North area of Mures county, Romania. *Radiation Protection Dosimetry*, 179 (1), 80–86.

Paschoa, A.S., Steinhäusler, F. 2010. Terrestrial, atmospheric, and aquatic natural radioactivity. *Radioactivity in the Environment*, 17, 29-85.

Radhakrishna, A. P., Somashekarappa, H. M., Narayana, Y., Siddappa, K. 1993. A new natural background radiation area on the southwest coast of India, *Health Physics*, 65, 390–395.

Sandor, GN., Poffijn, A., Cosma, C. 1999. Indoor radon survey in a uranium area from Romania, Radon in the living environment, 19-23 April, Athens, Greece, 207−208.

Tănasă I. 2011. Climate of the Suceava Plateau - risk phenomena, implications for sustainable development, Ph.D. thesis, Ștefan cel Mare University of Suceava.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation), 2000. Sources and Effects of Ionizing Radiation, United Nations, New York.

World Health Organization, 2009. Handbook on Indoor Radon: A Public Health Perspective, Geneva.