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Environmental and Human Health Issues from the Perspective of the Natural and Artificial Radioactivity in Covasna County

-*Summary of the PhD thesis*-

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CONTENT

Keywords: radioactivity, radon (^{222}Rn), mofetta, effective dose, cesium (^{137}Cs), soil, tree bark.

INTRODUCTION

The aim of this thesis is to contribute to environmental and human health issues from the perspective of the artificial and natural radioactivity in Covasna County. The clearly formulated objectives and the chosen methods are the prerequisites for achieving this aim.

The first major objective of the study is the collection of certain data regarding the natural radioactivity from different mofettes, the calculation of certain radon concentrations and effective doses on the human body. The majority of the mofettes are used without continuous monitoring and medical supervision. The data relating to this field are sporadic and often decades old. Therefore, the study of certain physico-chemical properties of mofetta gas is important, especially the study of those parameters, that are significant from the perspective of human health, such as the radioactivity of the gas emission. The specific objectives associated with this topic can be summarized as follows:

- ✓ selecting certain post-volcanic gas emissions from Covasna County frequented for curative purposes and performing radon measurements on different levels inside these mofettes;
- ✓ analyzing data collected on the site under laboratory conditions, obtaining radon concentration, and carrying out calculations in order to find the effective doses;
- ✓ comparing the results obtained within the framework of this research with other studies on radon inside mofettes in Romania;
- ✓ evaluating data obtained by us from the perspective of human health by means of legislative norms in the field of radioprotection.

The second major objective of the thesis deals with the anthropogenic radioactivity problems in the Ciomad-Bálványos protected area and refers to the use of cesium - 137 as an indicator of radioactive pollution in the post-Chernobyl era. In 1986, after the Chernobyl accident, more than 20 artificial radionuclides were identified, which polluted the human population and environment in Romania. Some of these radionuclides (including ^{137}Cs) with their long elimination half-life persist even today. They can be detected and quantified allowing different relevant conclusions in the field of radioprotection, referring especially to the last 30 years. For these reasons the following specific objectives are stated:

- ✓ collecting soil samples and tree bark samples in the protected area of Ciomad - Bálványos;
- ✓ determining the concentration of ^{137}Cs by gamma spectrometry;

- ✓ evaluating the results in the context of the data existing in Romania and in other parts of Europe.

In accordance with the two main objectives, the methodological part of the thesis consists of two segments. For the first part of the research, that deals with natural radioactivity and radon - 222, measurements from the mofettes the integrated method of solid-state nuclear track detectors CR - 39 were used. This approach is one of the most efficient methods for measuring the indoor radon concentration. This method consisted of passive measurements of the indoor air of the mofettes selected from Covasna County over a period of approximately one month with track detectors CR - 39 (RadoSys). In the present study, 6 track detectors CR - 39 were exposed at different heights inside the studied mofettes. After the completion of the exposure period the sampling and the analysis of the detectors took place in the Environmental Radioactivity and Nuclear Dating Center, Babeș - Bolyai University. Based on the ^{222}Rn concentrations, the doses were calculated for a regular treatment period. For the second part of the study, which deals with artificial radioactivity and ^{137}Cs, 31 samples of bark (spruce and oak) and 21 soil samples were collected. The cesium activity was determined using gamma spectrometry methods in Babeș-Bolyai University, Faculty of Environmental Science and Engineering.

The thesis is structured in 5 main chapters:

I. introductory chapter, which presents the purpose and objectives of this research, exposes certain methodological considerations and indicates the structure of the thesis;
II. literature review, this chapter presents data based on scientific literature regarding general aspects of radioactivity, followed by information related to natural and artificial radioactivity and data referring to radon - 222 and cesium - 137;
III. one chapter describes the conducted research and the methodology used in accordance with the stated objectives;
IV. the results are presented in a separate chapter, including the original contributions of the thesis as well;
V. the last chapter summarizes the main conclusions and possibilities for pursuing this research.

Finally, the bibliography is followed by the annexes of the thesis.

^{222} Rn AND ^{137} Cs, ISSUES OF ENVIRONMENTAL RADIOACTIVITY

Radon is present in different concentrations everywhere: in rocks and soil, in surface water and groundwater, in the atmosphere and even indoors. The ^{222}Rn and ^{220}Rn existing in the atmosphere originate primarily from the soil; that is clearly because of the ^{238}U and ^{232}Th content of the soil. The mofettes show radon in high concentrations. The term ”mofette” denotes a post-volcanic phenomenon, that means gas emissions at relatively low temperatures. The main component of the gas occurring in the mofettes is $CO_{2.}$ In addition to the carbon dioxide, mofetta gases contain other components too, for example nitrogen, oxygen, methane, hydrogen sulfide and noble gases, including radon.

Radon is the chemical element with atomic number Z = 86 in the periodic table. It is part of the 8th group, consequently it is an inert gas. It is formed by disintegration of the heavy elements from the earth’s crust and diffuses in soil gases or in water and it is transported in the atmosphere. Radon, under normal conditions, is a colourless gas and it has a density of 9,73 kg/m^3 (being the heaviest gas in nature). It also dissolves in water (according to Henry’s law), but it dissolves more easily in organic solvents. The main isotopes disintegrate by α radiation emissions. Radon is present in all three decay chains of natural disintegration of the uranium. The radon (^{222}Rn) with a half-life of 3,82 days is part of the uranium series (^{238}U; $T_{1/2}$ = 4,47×10^9 years) and it is created by the alfa disintegration of ^{226}Ra ($T_{1/2}$ = 1600 years).

Different organs of the human body are affected in different ways, but nevertheless benefit from following the treatments with mofetta gases that contain radon (Brassai 2000 and 1999, Falkenbach at al. 2005, Néda et al. 2008a, Szabó 2005 and 1978, Mureșan 1974). The positive effect on the endocrine system include thyroid hypofunction, stimulation of the sexual glands and the intensification of the excretion of uric acid. It has also a strong diuretic effect. At the same time, mofetta is recommended for gynecological diseases, infertility and impotence. It has positive results for cardiovascular issues, for example it has beneficial effects on hypotensives as well as hypertensives. By improving the peripheral circulation, the treatment is effective for certain skin diseases too. Through the change in nucleic metabolism, the radiation acts not only on the central nervous system, but on the autonomic nervous system as well. Different types of rheumatisms are successfully treated in the mofettes. An advantage of an adequate radon treatment compared to the drug treatment is the mitigation of secondary effects.

Radon therapy is contraindicated in some cases of (Néda et al. 2008a, Szabó 2005 şi 1978): febrile illnesses, tumours, tuberculosis, pregnancy, psychopaties, depressions, menstruation, respiratory problems etc. There is a tendency to use the mofettes empirically, however appropriate precautions and the specialist advice could mitigate the negative effects occurring because of negligence or failure to obtain correct information. Exaggerated radon exposure attacks first of all the lungs, especially the bronchial epithelium.

Radon may also pose a risk to human health, since radon contributes a certain percentage to the irradiation of the human body (Cosma şi Jurcuț 1996). The International Commission of Radioprotection (ICRP) has recommended different reference levels and these recommendations have formed the basis of the establishment of certain specified dose limits.

In Covasna County there are approximately 40 mofettes in the volcanic and flysch areas, as well as in intramontainous basins. Many localities, which own mofettes, have several dry baths, for example Turia commune or Covasna town. The mofettes are used for therapeutical or recreational purposes. The majority of mofettes, however, are used without continuous monitoring of the physicochemical properties and without medical supervision of use. Therefore, the studies performed on certain parameters of these gas emissions are of importance.

According to the definition given by the Fundamental Norms of Radiological Security, nuclear accident is the event that affects the nuclear installation and causes radiation, contamination of the population and environment above the permitted limits (Romanian NEPA 2015). The Chernobyl accident, that took place on 26th April 1986, is considered the biggest catastrophe in the history of peaceful exploitation of nuclear energy. Following the accident, a large quantity of radionuclides was dispersed in the atmosphere, that later spread all over Europe. The radioactive cloud reached Romania on 29th and 30th April 1986. Back then, the air masses moved in W-SW direction. Although the pollution reached us only after 3-4 days, there were areas with significant deposits (Cosma 2002, Toader şi Vasilache 1995). In Romania there were more than 20 radionuclides revealed by gamma spectrometry, amongst which I-131, Ba-140, La-140, Ru-103, Rh-103, Zr-95, Cs-134, Cs-137, Sb-125, Ce-141, Sr-90 (Romanian NEPA 2015, Cosma 2002). After June 1986, special consideration was given to some of these, such as cesium-137, cesium-134 and strontium-90.

Radionuclide ^{137}Cs had the highest level of contamination, and it was followed by ^{134}Cs and ^{90}Sr. Currently, ^{134}Cs dropped significantly by disintegration (with a half-life of approximately 2 years). Strontium-90, due to its low concentration, is determined in a complex way by radiochemical methods.

The ^{137}Cs element appears only in different artificial radioactive processes (EPA 2016). It has a half-life of 30,17 years, emits beta and gamma radiations. It decays in short-lifetime isotope ^{137m}Ba by beta radiations and the latter reaches a non-radioactive form known as barium, ^{137}Ba. From certain perspectives, this radioactive isotope behaves similarly to the stable one (Williams et al. 2004). It is capable of travelling long distances in the air until it reaches the soil again by means of the precipitation and gravitation. The majority of the chemical compounds of cesium are water-soluble. They form chemical bonds in the soil and, because of this, it doesn't travel long distances.

The highest values regarding soil contamination with ^{137}Cs in Romania were measured on the route of the radioactive cloud, namely on the NE-SW axis of the country, where these values reached up to 80 kBq/m^2 (Cosma 2002). Similar studies have brought results up to 100 kBq/m^2 in Sweden and in Ukraine and Belarus they have measured values even above 200 kBq/m^2 (Romanian NEPA 2015). The majority of the radionuclides have settled in the surface layer of the uncultivated soil, contaminating it long after the accident. By migration into the deeper layers and vegetation and by re-suspension the soil contamination has been reduced.

It has been shown that the forest ecosystems are complex and they show particular characteristics compared to other ecosystems from the perspective of the radionuclides, because the forest also functions as a natural reservoir (Calmon et al. 2008). It has been shown that the main reservoir long term is the soil, contributing in this way to the contamination of the plants. Research conducted in 1986 related to the vegetation of the country showed: radionuclides reached the plants primarily from the atmosphere through the leaves and the penetration through the soil and the root was insignificant (Constantinescu et al. 1988). Later, these considerations were confirmed at an international level as well, for example it has been proved that in case of trees, the main access of the radionuclides is their deposition at canopy level under dry or wet conditions (Fesenko et al. 2003). From the canopy the contaminated elements reached the other parts of the trees through the physiological activities of the plants, and they also reached the soil with the occasion of leaf fall (Shcheglov et al. 2011, Fesenko et al. 2003).

Currently, the soil is still considered a contamination source. The absorption of cesium-137 is slower in the case of the old trees than the young trees (Goor şi Thiry 2004), in accordance with their physiological processes. In the past, however, in the first months after the Chernobyl disaster, the main source of contamination was the deposition of the radionuclides from the atmosphere on the bark and other parts of trees (Kuroda et al. 2013). The structure of the bark and of the rhytidome affects significantly the way in which the woody plants were contaminated. The young individuals always have a smooth bark, which can be coloured according to the species. Over time, the dead tissues of the bark, in cumulation with the parenchyma and phloem, generate the rhytidome (Clinovschi 2005). The rhytidome can exfoliate in a circular way (birch, cherry tree), in longitudinal strips (thuja), in scales (spruce, apple tree, sycamore). The rhytidome does not always exfoliate and then it forms some characteristic cracks (elm, Turkey oak, walnut tree) or it exists in the form of suber excrescences as we encounter it in the case of Amur cork oak. Some species do not form rhytidome, therefore, they have smooth bark throughout their lifespan (hornbeam, beech).

During the years after the Chernobyl disaster, the main changes occurred in the cesium-137 content of the tree's rhytidome because of the radioactive disintegration and after the physical processes: diffusion, wash, bark fell etc. (Rulik et al. 2014, Zhiyanski et al. 2004). Not only the anatomic structure, but the physiological processes of tree also supports the results obtained by Fesenko et al. (2001 and 2003), i.e. in the case of the old individuals contamination through root or leaf plays secondary role. Thus, summarizing the above mentioned issues about the contamination of the trees with ^{137}Cs, we know that currently the concentration of radiocesium in rhytidome is proportional with the initial quantity (Cosma et al. 2016, Suchara et al. 2011).

METHODOLOGICAL CONSIDERATIONS

Radon concentrations inside mofettes and effective dose calculation

In this research the integrated method of solid-state nuclear track detectors CR-39 was used. This consisted of passive measurements of the indoor air of the mofettes (**Figure no. 15**) with track detectors CR-39 (RadoSys). It is recommended to use these detectors for a period of 20-80 days. Each detector has a certain individual ID number and based on this number their identification is performed. The applicability quantifies as follows: 40-8000 kBqh/m^3. Each detector is packed in a special radon proof bag. This is one of the most efficient methods for indoor radon concentration measurements.

Figure no. 15. *Radon detectors, RSKS type (RadoSys)*

There were selected 9 mofettes frequented for medical and recreation purposes from Covasna County. Out of these, there are the following mofettes in Covasna town: Bene, Bardócz, Dacia, Hephaistos, Bradul and the one belonging to the hospital. The other 3 are circulated too, namely Hătuica and Şugaş Băi and the one from Stinky cave (Peştera Puturoasă).

In the present study there were 6 track detectors CR-9 exposed in each of the 9 mofettes from Covasna County within the three periods from 2012 to 2015, according to the **Table no. 6**. In this table, by observing the D-F columns, one can notice that the detectors were exposed for 32-50 days. However, it is also shown that the measurements were repeated in 5 out of the 9 mofettes, namely in Bardócz, Bene, Şugaş Băi, Hătuica and Stinky cave (Peştera Puturoasă). For the calculations the most recent data related to the mofettes was used. All the measurements were performed in the cold period of the year, in the interval from September to January.

The detectors were distributed on levels as follows: 10-20 cm, 50 cm and 100-120 cm from the floor, to determine the vertical distribution of radon activity concentrations. At every height there were 2 detectors deployed. Additionally, detectors were used as controls to ensure accurate results. The distribution, collection of the detectors and the data analysis were carried out in accordance with the measurements protocol and with the quality assurance programme.

A. No.	B. Name	C. Locality	D.3 Exposure period I.	E. Exposure period II.	F. Exposure period III.
1	Hospital's mofetta	Covasna	-	17.09.2013. - 05.11.2013.	-
2	Mofetta Bardócz	Covasna	14.12.2012. - 20.01.2013.	16.09.2013. - 04.11.2013.	-
3	Mofetta Bene	Covasna	-	16.09.2013. - 04.11.2013.	21.12.2014. - 21.01.2015
4	Mofetta Hephaist.	Covasna	-	17.09.2013. - 05.11.2013.	-
5	Mofetta Dacia	Covasna	-	17.09.2013. - 05.11.2013.	-
6	Mofetta Bradul	Covasna	-	02.10.2013. - 05.11.2013.	-
7	Mofetta Hătuica	Hătuica	14.12.2012. - 20.01.2013.	-	21.12.2014. - 21.01.2015.
8	Mofetta Şuga Băi	Sf. Gheorghe	14.12.2012. -24.01.2013.	-	21.12.2014. - 21.01.2015.
9	Stinky cave	Turia	15.12.2012. - 27.01.2013.	-	21.12.2014. - 21.01.2015.

Table no.6. *Mofettes and periods: measurements of radon activity in Covasna County*

The processing and data analysis were performed in the Environmental Radioactivity and Nuclear Dating Center at Babeș Bolyai University from Cluj-Napoca. This phase of the research involved several stages. The solvent development stage included chemical etching in a NaOH concentration of 6,25 molarity for 4,5 hours at a temperature 90^{o}C. This stage was followed by the counting of the alpha particles on the surface of the sensitive plastic film. The tracks of the alpha particles on the surface of the sensitive plastic film were read using the optical microscope RadoMeter 2000 RadoSys. The average radon activity concentration was calculated in Bq/m^3 based on the track density of the alpha particle tracks/mm^2, using the formula below (Cosma et al. 2009, Cosma et al. 2013, Cucoş et al. 2012):

$$C_{Rn} = \cdot\ p \cdot F_c/t$$

C_{Rn} -calculated radon concentration [$Bq \cdot m^{-3}$]

p -measured track density [tracks mm^{-2}]

F_c -calibration factor

t -exposure time [days]

A total treatment lasts for 15 days with 20 minutes/day spent in the mofetta (Cucoş et al. 2014, Neda et al. 2008a). Based on the radon concentrations’ values the effective dose was calculated for a common treatment, using the following formula (Cucoş et al. 2014, Harrison şi Marsh 2012):

$$ED = C_{Rn} \cdot K \cdot F \cdot t$$

C_{Rn}-radon concentration [$Bq \cdot m^{-3}$]

K-conversion factor [$nSv \cdot (Bq \cdot h \cdot m^{-3})^{-1}$],

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F-equilibrium factor [0,4]

t-time spent in the mofetta [h]

^{137}Cs in tree barks and soil samples

In the 17-21 August 2013 period there were 31 tree bark samples collected from the protected area Ciomad-Bálványos, Covasna County. Out of these samples, 16 were sessile oak (*Quercus petraea*) samples and the other 15 were spruce (*Picea abies*) samples. The samples were collected according to the methodology applied at international level (Cosma et al. 2016, Fesenko et al. 2003), namely:

- ✓ trees of min. 50-60 years,
- ✓ height of sampling 1,3 m,
- ✓ NW direction.

The samples were collected simply by hand or by using a screwdriver. The sampling was limited to approx. 3 mm in the case of spruce and 5-8 mm for the sessile oak. The samples were dried, sprayed and homogenized. The dry mass was 30-50 g/sample.

The collection of the soil samples took place on 26.03.2015 in 8 locations in Ciomad-Bálványos area. The collection of the samples was carried out in uncultivated land soils using a cylinder of 20 cm height and a diameter of 6 cm. For each study site the geographical coordinates were established using a GPS. Outside the protected area, in the locations 1, 2, 7, 8, the soil samples were simply treated. However, in locations 3-6 one profile was made for each: 0-5 cm, 5-10 cm, 10-15 cm and 15-20 cm. After sampling, the soil was placed in plastic bags and labelled with the information of sampling. The soil samples were dried and homogenized.

The analysis of the bark and soil samples was performed within the framework of the Faculty of Environmental Science and Engineering. The gamma spectrometry measurements were carried out with a HpGe GMX type detector manufactured by the Ortec company. Maestro-32 software was used in order to determine the cesium-137 (Bq/kg) activity. The concentration of the radionuclides from the samples was calculated with the relative method. For the samples with ^{137}Cs content the IAEA - 375 standard was used that is certified by the International Atomic Energy Agency. Considering the analyzed samples in this thesis, the cesium-137 was determined by a certain peak, namely the 662 keV line.

RESULTS AND DISCUSSIONS

Radon concentrations beneficial for human health

In the first period of data collection from December 2012 to January 2013, as expected, the highest radon activity was measured at 10 cm from the floor, namely 5035 Bq/m^3 in mofetta Bardócz. The profile of the radon activity in general shows a decrease concomitantly with the increase of the height. Therefore, the finding of higher values at 10-20 cm than at 50-70 cm or 100-120 cm was something to be expected. This phenomenon of variation of the radon activity concentration occurs frequently in the scientific literature (Csegzi 2008, Cucoş et al. 2014, Szakács et al. 2006, etc.). This tendency, however, is valid especially in mofettes that are placed directly above gas emissions and not necessarily for those which are supplied from a distance from natural or artificial sources. Additionally, the stability of mofetta gas in a concave space contributes to the emergence of the mentioned profile.

In the first period of data collection, namely from December 2012 and January 2013, as expected, the lowest radon activity concentration was measured at 120 cm from the floor, specifically 280 Bq/m^3 in mofetta Şugaş Băi. In the first period this mofetta shows the following average radon activity value depending on the height: 1709 Bq/m^3 at 10 cm, 1917 Bq/m^3 at 50 cm and 320 Bq/m^3 at 120 cm. The same profile was identified in the case of mofetta Bene (in the second period) and at the Stinky cave (in the third period). A possible explanation may be that the radon does not come just from the bottom of the concave space, but also from its sides.

The values that increase from bottom to top (e.g. Bardócz, Hephaistos) having the minimum value at 10-20 cm and the maximum at 100-120 cm, mark a gas perturbation for different reasons, such as meteorological conditions, planning problems or malfunctioning.

As identified in Table no. 14, we can see that, in similar conditions, in the winter of 2014-2015 significantly higher values were measured in 2012-2013 in the same period of the year. The difference between the two seasons in the three investigated mofettes can be explained by meteorological conditions. The lower the air temperature values, the more unstable the mofetta gas is, therefore the radon concentrations decrease. Similarly, the higher the air humidity, the more stable the mofetta gas is in the concave spaces.

A.Mofettes	B.Height (cm)	Average ^{222}Rn Activity (Bq.m^{-3})		E. Ratio
		C. Winter 2014-2015	D. Winter 2012-2013	C/D
Mofetta Şugaş Băi	120	157	320	0,49
	50	2382	1917	1,24
	10-20	2707	1709	1,58
Mofetta Hătuica	120	5721	3792	1,51
	50	6334	3231	1,96
	10-20	6187	3093	2,00
Stinky Cave, Turia/Băile Bálványos	120	2173	1153	1,88
	50	5104	938	5,44
	10-20	4136	1696	2,44

Table no. 14. *The difference of the ^{222}Rn activity between the two seasons*

The possible explanation of the differences between the radon concentration values from the two seasons investigated in the three mofettes referred to is based on the air temperature. The winter in the 2012-2013 season was a frosty winter and the one from 2014-2015 was a mild one. More precisely, the average temperature deviations (related to the standard period, namely 1961-1990) in December 2012 showed values between -2,9 ºC and -1,5 ºC, and the same values in December 2014 were between +1,1 ºC and +3 ºC. In absolute values, in December 2012, the average temperature of the month ranged from -3,9 ºC to -2 ºC, and in December 2014 these values were between -1,9 ºC and 0 ºC. In January 2013, the average temperature deviations (related to the standard period, namely 1961-1990) were slightly positive, showing values between 0,1 ºC and 1ºC. The same values in December 2015 were between 1,5 ºC şi 2,0 ºC. In absolute values in December 2012, the average temperature of the month was between -3,9 and -2,0 ºC, moreover in December 2014, these values were between -1,9 ºC and 0 ºC. Therefore, in the colder winter, the one from 2012-2013, there were lower radon activity values, whereas the less cold winter determined higher concentrations of radon activity in the mofettes.

A.Mofetta	Effective dose (mSv)					
	I. period Dec.2012-Jan.2013		II. period Sept.-Nov.2013		III. period Dec.2014-Jan.2015	
	B. K = 9	C. K = 12	D. K = 9	E. K = 12	F. K = 9	G. K = 12
Mofetta BARDÓCZ	0,051	0,068	0,141	0,187	-	-
Mofetta BENE	-	-	0,195	0,260	0,129	0,172
Mofetta Spitalului	-	-	0,024	0,032	-	-
Hephaistos Hotel’s Mofetta	-	-	0,148	0,197	-	-
Dacia Hotel’s Mofetta	-	-	0,061	0,081	-	-
Bradul Hotel’s Mofetta	-	-	0,015	0,020	-	-
Mofetta Hătuica	0,063	0,084	-	-	0,108	0,145
Stinky cave (P. Puturoasă)	0,019	0,025	-	-	0,065	0,087
Mofetta Şugaş Băi	0,020	0,027	-	-	0,023	0,030

Table no. 15. *Effective doses in nine mofettes from Covasna County, in three periods*

The time spent in the mofetta was estimated here, too, at 5 hours, taking into consideration that a session lasts for approx. 20-30 minutes and a total treatment has 10-15 days. The conversion factor received was 9 and 12. The corresponding results are shown in the B-G columns in Table no. 15. As one can see in this table, the effective dose contains values between 0,015 and 0,260 mSv. As expected, the 3 minimum values come from the first and the third period, i.e. the winter periods (from Mofetta Bradul, Stinky Cave and from the Şugaş Băi Mofetta). The maximum values were obtained in the second period (in autumn) in Bene and Bardócz mofettes.

Naturally, in the case of the conversion factor K = 12, the doses are slighty higher (columns C, E, G), than in the case of K = 9 (columns B, D, F). The effective dose values, calculated in this study and reflected in the Table no. 15, are under 20 % of the one indicated as additional dose for the population which is 1 mSv (Harrison and Marsh 2012., Incze et al. 2016). There is one single exception in the case of Bene mofetta in the second period. Therefore, according to our studies, the radon from the mofettes in itself does not constitute a risk factor. However, if the patient receives other doses of radiation, the dose received from the mofettes needs to be taken into consideration. Additionally, more attention needs to be focused on the staff members, e.g. guides, medical nurses, who spend a lot more time inside or in the surroundings of the mofetta related to the patients that undergo just a treatment.

^{137}Cs, radioactive contamination indicator

The results of the cesium activity measured from the soil samples belonging to the protected area of Ciomad-Bálványos are comparable with the data from the scientific literature (Begy 2009, De Cort et al. 1998, Cosma 2002). In this case an appropriate method is the comparison with the activity from 1986, because each series of measurements from the publications reflect dynamic data throughout the last 30 years referring to radiocesium. Normally, the activity from the past and future can be calculated on the basis of the fundamental law of radioactive decay: $N(t) = N_0\, e^{-\lambda t}$. In our case, however, a far simpler method is the duplication of the current activity, because 30 years from the Chernobyl disaster have recently passed and the half-time of the ^{137}Cs is 30,17 years. Out of the four soil samples collected in the vicinity of the protected area Ciomad - Bálványos, the minimum current activity value was 24,85 Bq/kg at the sample no. 7, which means 10991 Bq/m^2 deposition in 1986. The maximum value was obtained in the case of sample no. 1, namely 61,64 Bq/kg, corresponding to a deposition of 27263 Bq/m^2 from 1986. We can find similar values in the scientific literature as well.

In the four locations inside the protected area of Ciomad-Bálványos, the minimum value of the current activity measured from the soil samples was found in location no. 3, namely 7833 Bq/m^2, which means 15665 Bq/m^2 deposition from 1986. The maximum value was obtained in location no. 6, namely 11345 Bq/m^2, which corresponds to a deposition of 22690 Bq/m^2 from 1986. We can find similar values in the scientific literature as well. It needs to be mentioned that the values obtained in the location no. 6, and 11345 Bq/m^2, corresponds to a deposition of 22690 Bq/m^2 from 1986. We can find similar values in the scientific literature as well. It needs to be mentioned that the values obtained in the investigated area belong to a high range in the context on Transylvania. This evidence of high activity is reflected in the calculated average for the 8 locations from the Ciomad-Bálványos area: the current one is 9404 Bq/m^2 and the one from 1986 is 18808 Bq/m^2. However, compared to the highest values regarding soil contamination with ^{137}Cs in Romania, which were measured on the route of the radioactive cloud, namely on the NE-SW axis of the country, and which have reached values up to la 80 kBq/m^2 (Cosma 2002), the values obtained in the Ciomad-Bálványos area are relatively low, since the maximum deposition (1986) is 27263 Bq/m^2. The results from this research in a European context are moderate: similar research brought results up to 100 kBq/m^2 in Sweden and in Ukraine and Belarus measured values were above 200 kBq/m^2 (Romanian NEPA 2015). In Denmark, Belgium and Hungary, however, the values are much lower (De Cort et al. 1998). The majority of the radionuclides have settled in the surface layer of the uncultivated soil, contaminating it up to now. Amongst other things, the soil contamination has been reduced by the migration of the radionuclides into the deeper layers. One can notice the pollution reduction and the decrease in activity in parallel with the increasing depth. Although the scientific literature describes the correlation between deposition and geographic height, in our case between these parameters there occurred only weak correlations, namely 0,3. The weak correlation can be explained by the relatively small number of sampling and by the narrow range of the samples in terms of height: 625 m-871 m.

It is known that after the Chernobyl disaster, with regard to trees, the main source of contamination was the deposition of the radionuclides from the atmosphere on the bark and other parts of the woody plants (Kuroda et al. 2013). Additionally, it is known that the bark structure and the rhytidome significantly affected the way the woody plants were contaminated.

In the protected area of Ciomad-Bálványos, bark samples were collected belonging to spruce and oak species. It can be observed that the values of the ^{137}Cs in the the oak are higher than in the spruce, the average value of oak being 44,75 Bq/kg and in the case of the spruce 10,59 Bq/kg. The average error in the spruce is considered 7,5 and for the oak 9,3 (Cosma et al. 2016). The ratio between oak and spruce is 4,22. The difference can be explained by the anatomic and physiologic properties of these tree species. The older trees (min. 50-60 for this study), however, generate rhytidome, in other words they display dead tissues of the bark, resulting from the activity of the phellogen, combined with the parenchyma and the phloem. Rhytidome may exfoliate in scales in the spruce and in the oak it forms some characteristic cracks. The oak has a structure with far more and bigger cracks and does not exfoliate. The spruce has a smoother textured bark and also exfoliates. It is therefore natural for the oak to be more active, in our case approx. four times higher.

The differences between *Quercus sp.* and *Picea sp.* are also reflected in the scientific literature. Moreover, the different tree species show a ranging capacity of cesium accumulation depending on the texture and physiological characteristics of the bark. The accumulation capacity of the ^{137}Cs is ranging as follows (Cosma et al. 2016):

oak > aspen > spruce >cherry tree.

The correlation between the geographic height and ^{137}Cs was analyzed. The results reflect a weak correlation for both the spruce (0,2) and the oak (0,3). At the same time, there is a strong relation between the ^{137}Cs activity in the bark and the location of the sampling compared to the route of the radioactive cloud from 1986 (NE-SW direction). We have higher values in Suceava - Harghita - Mureş - Caraş-Severin, and the neighboring areas (Covasna, Cluj, Sibiu). Lower values come from the Bihor, Maramureş, Bistriţa and Sălaj counties. Thus, in the case of the spruce in Suceava County there were measured 11,9 Bq/kg, in Mureş 20,8 Bq/kg, in Sibiu 16 Bq/kg, and in Bistriţa only 3,1 Bq/kg, in Maramureş 7,7 Bq/kg (Cosma et al. 2016). For the species of *Quercus sp*., the values are generally higher than for spruce, however, they reflect also the position, distance from the NW-SE axis of the country. Thus, the *Quercus sp.* in Suceava measured 58,3 Bq/kg, in Caraş-Severin 73,6 Bq/kg whereas in Bistriţa only 5,9 Bq/kg or in Bihor only 4,9 Bq/kg (Cosma et al. 2016). Consequently, it can be concluded that the values of this research are in accordance with the other values from the country for both the spruce and the oak.

CONCLUSIONS

This current study provides data regarding the **distribution and variation of the radon activity inside the mofettes**. The mofettes, which are formed directly above the emissions (e.g. Stinky Cave, Hătuica) occur at a maximum value at a certain height from the floor (in the case of this study at 50 cm), because the radon penetrates from the sides as well as the depths. The maximum radon concentration values are at the inferior level and these values gradually increase in the case of the mofettes that receive gas from distance (e.g. Hospital's Mofetta, Bradul Hotel's Moffeta). The values that increase from bottom to top (e.g. Bardócz, Hephaistos), have the minimum value at 10-20 cm and the maximum at 100-120 cm, reflects a gas perturbation due to different reasons (e.g. meteorological conditions, planning problems or malfunctioning).

The current study through the measured activities and the calculated doses (0,015-0,260 mSv) confirms that the radon from the mofettes in itself does not constitute a risk for the patients. However, if the patient receives other doses of radiation, the dose received in the mofettes needs to be taken into consideration as well. Far more attention needs to be focused on the staff members (e.g. guides, medical nurses), because they spend a lot more time inside or in the surroundings of the mofetta, than the patients that undergo a certain treatment.

The data obtained in this research are comparable with the data from the scientific literature, the relatively small values are due to the timing of the sampling period (autumn and winter). In similar conditions, during the 2014-2015 winter there were significantly higher values measured in the mofettes, than during 2012-2013 in the same period of the year. The difference between the two seasons was explained by the meteorological conditions: the lower the air temperature values, the more unstable the mofetta gas, therefore the radon concentration became lower.

The results of the cesium-137 deposition measured from the soil samples in the Ciomad-Bálványos protected area are comparable with the data from the scientific literature. The current average calculated for the 8 locations is 9404,25 Bq/m^2, whereas the average from 1986 is 18808,50 Bq/m^2. The values obtained in the investigated area belong to the high range in the context of Transylvania. However, in comparison with the highest values (measured on the route of the radioactive cloud, namely on the NE-SW axis of the country), which reached up to 80 kBq/m^2, the values obtained in the Ciomad-Bálványos are moderate, having a maximum deposition value of 27262,94 Bq/m^2 (1986). The results from this research in a European context have medium values. For example in Denmark, Belgium and Hungary the values are lower and in Sweden, Ukraine and Belarus there are higher depositions.

The ^{137}Cs activity profile is a typical one, namely the maximum values occur close to the surface, that is at 0-5 cm depth, gradually decreasing, and the minimum values are at 15-20 cm. These results are in accordance with the theory by which the majority of the radionuclides settled in the superficial layer of the soil after the 1986 disaster. The radioactive contamination continues to this day, and the reduction is carried out, amongst others, by the migration of the radionuclides in the deeper layers of the soil.

The current research confirms that the cesium-137 activity from the bark is affected significantly by the tree species and the bark texture. The activity values ^{137}Cs in the oak are higher than in the spruce, the average value in the oak is 44,75 Bq/kg, whereas in the the spruce it is 10,59 Bq/kg. The ratio between the oak and the spruce is 4,22. The oak has a structure with more and bigger cracks and, additionally, the species does not exfoliate. However, the spruce has a less cracked surface and exfoliates. Therefore, the activity in the oak is naturally higher than in the spruce.

In the bark of the oak the maximum activity of the ^{137}Cs was measured in the first layer, that is in the exterior, and it was 1,6 Bq/kg. Under 12 mm depth, the ^{137}Cs concentration did not change significantly. These data support the theory that after the Chernobyl accident, with regard to trees, the main source of contamination was the deposition of the radionuclides from the atmosphere on the bark and other parts of the woody plants. It is important to know the ranging capacity of ^{137}Cs accumulation in the layers of the bark, because this explains the accumulation mechanism and it helps us to clarify the nuances of the sampling methodology.

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ANNEXES