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DOCTORAL THESIS

Thoron contribution to the natural irradiation of the Romanian population and problems related to thoron and radon measurements

- Summary -

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Keywords:

Thoron, radon, decay progenies, integrated measurements, passive monitoring, active methods, track detectors, indoor, exposure, natural dose, seasonal variations, diurnal variations, mapping, radon mitigation.

1. Introduction

1.1. Introduction to radioactivity

Radioactivity, the process of nuclei undergoing spontaneous decay or disintegration, is a part of nature as the process of element formation by nuclear reactions taking place in stars; both stable and radioactive isotopes of elements are formed. The isotopic composition of elements is characterized by properties of nuclear reactions that led to the formation of the elements. Elemental composition of the planet Earth, thought to be about 4.5×10^9 years old, although not yet in chemical equilibrium, reflects the composition of the material from which it was borne.

There are about 340 nuclides in nature, of which about 70 are radioactive. The latter are called the naturally occurring radionuclides, and are found mostly as isotopes of heavy elements. Uranium and thorium, for example, known long before the discovery of radioactivity can be found in their natural state, distributed within different layers of the Earth's crust, and in all types of rocks and soils (Paschoa and Steinhäusler, 2010) but also in river and sea waters as well as in biological organisms (Ouseph, 1975). The radioactivity of natural radionuclides as uranium (^{235}U and ^{238}U) and thorium (^{232}Th) series is due, essentially, to alpha and beta decay process plus gamma de-excitation of nuclei. As a result of the processes of breakdown of rocks and their weathering, radioactive elements are migrating, thus, the radioactive equilibrium is broken, the radioactive elements decayed from uranium and thorium, gradually disintegrating. Short-lived elements quickly disappear while long-lived radioactive elements form secondary sediments, such as radium-bearing brown clay. As it is, not long after discovering, in 1896, that uranium emits radiation, in 1898 was singled out an element that would emit an even higher radiation. That element was radium, hence the term radioactivity (Cosma and Jurcut, 1996). Radium occurs in soil, sea and river waters, hence, its decay products (radon, thoron and actinon) can be found in all natural environments.

1.2. Aim of the study

Radon represents one of the most important contributors to the natural radiation of the population. Together with its decay products, it is classified as one of the leading causes of lung cancer development.

World Health Organization (WHO) declared radon to be the second cause of lung cancer after smoking. Beside this increasing concern in regard to radon, the importance of thoron (short-lived isotope of radon) has been recently acknowledged. Thoron gas is present everywhere along with radon and sometimes indoor thoron level can be even higher than radon. Consequently when considering indoor radon exposure a special consideration should be paid to the presence of thoron together with radon, otherwise the risk of exposure to radon will be underestimated. The importance and novelty of this research can be found reflected in the increased interest shown during the last couple of years by worldwide researchers through the multitude of papers which treat the amplitude and mansidedness of thoron investigations worldwide especial as a potential carcinogenic gas.

In the context of increased interest shown by the international scientific community on the importance of monitoring radon and its impact on public health, in Romania, to date, there have been no systematic studies on a national level that would include a mapping of radon or thoron, and therefore does not have any legal basis in terms of residential radon protection. Romania's obligations as a Member State of the European Union, in terms of environmental policy, are clearly set out in the European Commission Treaty, Article 174 (which refers to the necessity of preserving, protecting, and improving the environment; human health's protection; rational use of natural resources; and promoting specific measures aiming at approaching environmental problems at a regional level). Therefore, certain measures are necessary to properly quantify the concentrations of radon and thoron, both inside and outside, to identify risk areas and also their mapping. This process is a necessity and a priority within both national and European standards for the protection of the population and occupational personnel.

Therefore, the main aim of this study is to contribute to the awareness of the Romanian population in regard to their natural radioactive environment. The second objective, aim towards contributing to a radon-thoron map, which will provide a database of informations that could be useful for further research and for highlighting regions with elevated natural radioactivity in Romania.

Within the present thesis are discusses two overlapped topics which present an increased interest worldwide and yet, not so much in Romania. The first subject is focused on thoron measurements in Romania and estimation of its dose contribution to the natural dose received by population. The second research topic is represented by the large variety of inconveniences that often intercept with accomplishing the first task.

1.2.1. Outline of the thesis

The thesis is structured over six chapters. In the first four chapters is described the literature study which represents the theoretical background required in order to properly carry out the set research. **Chapter 1** presents an introduction into the field, the motivation for implementing such topic and the goal of the research. The next two chapters (**Chapter 2**, **Chapter 3**) describe relevant information for the topic in regard to thoron and its decay progeny's gas features and proprieties, available measuring methods and techniques and their applications.

The next two chapters represent the original contributions to the topic. **Chapter 4** includes 3 subchapters and analyses the thoron level in Romania and its contribution to the natural irradiation of the population. **Chapter 5** comprises 7 subchapters and is focused on investigating thoron and radon dynamics, such as, indoor spatial distribution and diurnal and seasonal variations and testing of measuring devices and also describes the research conducted during 4 months mobility training in the Institute of Radiochemistry and Radioecology, Veszprém, Hungary.

All the conclusions that can be drawn from the experimental investigations and which can be an indicator for future research are reunited in the last chapter (**Chapter 6**).

The references used within this thesis are listed in the References.

4. Thoron contribution to the natural irradiation in Romania

4.1. Thoron measurements in Romania throughout the years

The first half of '90's marked the beginning of thoron gas measurements with an isolated outdoor survey (Encian et al., 2005) that used air filtering and gross beta measurement. Further on, thoron soil exhalation tests in the urban area of Cluj-Napoca (Cosma et al., 2005) were performed using activated charcoal and Lucas cell method, the latter being also used for soil-gas exhalation near mofetts (Papp et al, 2010). In the past couple of years the focus turned towards long-term indoor measurements by using radon-thoron discriminative detectors (Burghele et al, 2011; Burghele and Cosma, 2012, 2013, Burghele, 2013) which by the time this paper was written was summing up to 145 indoor and 2 outdoor thoron measurements within the territory of Romania. A list of published data on thoron gas measurements in Romania is presented in Table 2.

Table 1. State of the art for thoron gas measurements in Romania throughout the years

Author	No. of locations	Type of survey (period)	^{220}Rn	Measuring sistem/method	Annual effective dose (mSv/yr)
Encianu et al., 2005	1	outdoor (1994 - 1997)	0.18 (Bq/m ³)	air filtering and gross beta measurements	-
Cosma et al., 2005	3	exhalation from soil	2986±180 (mBq/m ² s)	charcoal	-
			2423±146 (mBq/m ² s)	LUK3C - scintilation cell	-
Burghele et al., 2011	35	indoor	106±77 (Bq/m ³)	RADUET	-
Burghele and Cosma, 2012	35	indoor	70±4(Bq/m ³)		0.02
Burghele and Cosma, 2013	15	indoor	91 ± 57 (Bq/m ³)	RADUET	-
	6	indoor	15±10 (Bq/m ³)	RAD7	-
	7	exhalation - wall - flor	395±96 (Bq/m ³) 66±43 (Bq/m ³)	RAD7	- -

The average thoron activity concentration of 58.7 Bqm⁻³ calculated from the existing indoor measurements is 2.1 times lower than the Romanian radon mean of 126 Bqm⁻³.

4.2. Feasibility study and methodology

Preliminary measurements were started at the beginning of 2011 in three counties (Bihor, Satu Mare and Sălaj) located in north-western part of Romania (Burghele et al., 2011). A number of 35 RADUET detectors have been placed in different types of indoor environments, i.e. dwellings, cellars, schools and offices. The locations were chosen in such manner that would involve different type of building materials and a different occupation factor. All detectors were suspended 30 cm from the wall, with the help of an iron wire, in the lowest occupied level of the building.

After three month exposure, detectors were collected and brought to the Laboratory of Environmental Radioactivity ('Babes-Bolyai' University of Cluj-Napoca, Romania) for processing. The first step of processing starts with preparing the solution and detectors for etching. The second step represents the etching itself followed by the third step, track reading.

The thoron mean concentration for the three counties ranged between 41 Bqm^{-3} and 128 Bqm^{-3} with a mean concentration of 75 Bqm^{-3} representing a lower activity compared to those reported for other countries (Gulan et al., 2012) and a low contribution to the effective dose. Taking into consideration the occupation factor of each location, thoron dose was found ranging between 0.003 mSv/y and 0.07 mSv/y . The thoron decay progeny mean concentration was 1.5 Bqm^{-3} representing a higher concentration than previously measured in Romania (Iacob et al, 2005).

The Tn/Rn ration was calculated to be 0.21 for both Salaj and Bihor County and 0.83 for Satu Mare County.

4.3. Survey of thoron level in schools

A new thoron/radon survey was started in north-western part of Romania covering 35 schools within the same three counties previously investigated

Among 35 radon - thoron paired detectors, 24 showed the presence of thoron with concentrations varying widely in the range of 3 Bqm^{-3} to 235 Bqm^{-3} , thus presenting a higher maximum value than reported for Slovenian schools (Vaupotič et al., 2012).

This study concluded that all investigated schools presented radon and thoron level lower than the action level for workplaces suggested by ICRP 1993 and one location exceeding the radon indoor reference level recommended by ICRP 115. The thoron activities are predominantly below 100 Bqm^{-3} while radon activity is mostly above 100 Bqm^{-3} but without exceeding 500 Bqm^{-3} . The seasonal effective dose calculated from thoron exposures, overlapping all three counties, ranged between 0.004 mSv and 0.05 mSv .

5. Problems related to thoron and radon measurements

5.2. On site thoron and radon monitoring problems

The spatial distribution analysed with the help of RAD7 indicated a slightly higher activity of thoron in the vicinity of the walls and low activity towards the centre of the room, while radon concentration followed a relatively constant pattern indifferent to the distance from the wall.

Surface emission measurements pointed out that the walls (building material) represent the source of thoron and the floor (filling) of radon.

After diurnal measurements in a red brick building it was observed a low activity of Tn in the morning followed by a drop below limit of detection (LD) during afternoon. Thoron developed an accession of activity after midnight when the temperature starts to decrease. On the other hand, Rn activity does not seem to be affected neither by temperature nor humidity: it presents a rather steady behaviour throughout the day and a slight increase of activity during night hours.

Diurnal investigations performed in a 50 years old adobe building provided a rather constant behaviour for Tn activity throughout the whole investigated time interval. Unlike in the red brick building, the radon activity was found to be constant during day time but decreasing consistently after midnight. Neither gas appears to be influenced in any way by the temperature or humidity.

5.3. Seasonal variations of indoor thoron and radon in Vaslui County

The present survey was conducted during a two year period (2012-2013) in which time 15 Raduet detectors were deployed for 3 months during a drought summer season (2012) and 15 during a rainy summer season (2013). In the same locations investigations were also performed with 30 Raduet track detectors deployed during winter season (2012 - 2013).

The summary statistic of indoor thoron and radon concentration measured for each season are presented in *Table 6*. On testing the outdoor thoron and radon level with 2 track detectors in the same area of interest was obtained an average activity concentration of 9 Bqm⁻³ respectively 13 Bqm⁻³.

Table 6. Summary statistic of indoor thoron and radon concentration for each season.

Gas concentration (Bqm ⁻³)	Summer (drought season)	Winter	Summer (rainy season)	Annual mean
Thoron	94	82	79	80
Radon	248	216	146	196

The present survey comes to confirm the previous study conducted in Transylvania regarding radon seasonal variability indoor according to which radon presents high activities during winter season and low activities during the summer season. In case of thoron, the second set of data confirms the international database through the fact that indoor thoron activity concentration decreases during rainy season while the first step of the survey brought to attention a novelty in the field, the fact that indoor thoron activity concentration appears to present an increasing tendency at high atmospheric temperature.

5.4. Mapping of indoor radon and thoron in Vaslui County

Vaslui County is located in central-east side of Romania and has its territory administratively organised into 3 municipalities, 2 towns and 81 communes cumulating in a total area of 5,318 km² and a total population of 375,151 people. Its whole territory was divided into 10 km x 10 km grids in accordance with JRC (Dubois și col., 2010) and the Romanian Radon Map (Cosma et al., 2013). During the last two years (2012-2013) 90 radon and 60 thoron track detectors (type RSKS and RADUET designed by Radosys) were deployed for periods of three months in dwellings, public and private institutions from 15 villages and one city spread over 9 grid cells of Vaslui County hence an area of 900 km². The placement and processing of detectors were made in accordance with HPA-NRPB Measurements Protocol (Miles and Howarth, 2008).

The indoor radon and thoron was mapped over 9 grid cells roughly covering 18 % territory in the south-west of the county. *Figure 13* presents the AM of radon and thoron from the measurements made per grid cell.

Annual indoor radon concentration from 90 measurements performed in Vaslui County concluded in an average indoor level of 186 Bqm⁻³ for this county. Thoron annual concentrations from 60 indoor measurements ranged between 12 Bqm⁻³ and 559 Bqm⁻³ with 18% data above 100 Bqm⁻³ and 5% of measured concentrations above the recommended action limit for radon, cumulating in an average indoor thoron level for Vaslui County of 80 Bqm⁻³. Thoron contribution to the natural dose received by the population of Vaslui County, living within the investigated area, varied between 0.06 mSv/y and 3.13 mSv/y, while radon contribution ranged between 0.53 mSv/y and 13.99 mSv/y. Consequently, it is obvious that in some situations the worldwide average annual exposure to natural radiation sources (2.4 mSv/y suggested by UNSCEAR 2000) is exceeded by thoron itself, before adding radon contribution.

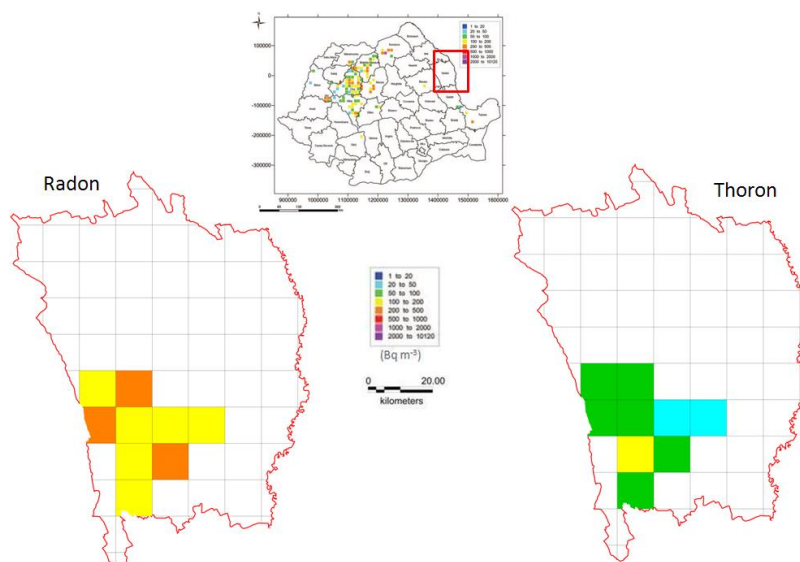


Figure 13. Arithmetic mean of indoor radon and thoron concentration for the surveyed cells of Vaslui County.

5.6. Influence of CO₂ concentration on radon measuring devices

The experiment performed at the Institute of Radiochemistry and Radioecology, Veszprém (Hungary) in collaboration with DurrIDGE Company Inc., USA and the University of Wyoming, USA (Lane-Smith and Sims, 2013) was divided into two stages: first was focused on comparing the results obtained with different radon devices in a chamber presenting environmental CO₂ concentration (0.04%) and a second stage, when the above mentioned chamber was filled between 0% and 100% with CO₂.

Twelve samples were taken from the radon chamber at random CO₂ concentrations within 50% - 100%.

The conclusion of the experiment was that the radon concentration measured by grab-sampling technique, using scintillation cell and Pylon AB-5 was overestimated when the CO₂ level in the analysed air was situated between 50% and 70% and underestimated above 70% CO₂.

5.7. Radon mitigation techniques applied to 3 houses

Three houses were chosen from a village located in the vicinity of the former uranium mine Băița-Ștei where long-term indoor measurements performed during two years (2010-2011) pointed out at least one room with radon activity concentration higher than 300 Bq m⁻³, the reference level recommended by ICRP 115.

Averages of 15 samples of air per house were collected from cracks found in the flooring or at the contact of it with walls, locations representing potential radon leakages into the house. Values greater than 1 kBqm⁻³ confirm the presence of leakages; such values were found in all three houses.

Soil-gas radon concentration was also investigated, by collecting averages of 15 soil-gas samples from the surrounding propriety of the house together with soil permeability measurements in order to determine the radon potential and index of the soil.

Continual measurements of indoor radon concentration were performed using several radon devices, such as: Sarad, Radim, AlphaGuard, Rad7 and Ramon.

Beside radon gamma anomalies were also investigated using Gamma Scouts and gamma spectrometry.

All diagnostic investigations had as main purpose the identification of radon sources. The general conclusion points towards the house foundation as the prime source of radon indoor, which is in all cases filled with stones from the rock spoils of the uranium mine, as the prime source of radon indoor. In order to decrease the radon level identified, several previously tested (Cucos et al., 2011; Cosma et al., 2013-b) remediation techniques were applied (Burghele et al., 2012).

Testing of remediation techniques was based on efficiency accountancy by comparing annual activity concentrations (integrated measurements) found before and after implementation.

Table 11. Data reported before and after applying remedially techniques obtained by integrated measurements and efficiencies for each case.

Code	Annual concentration (Bqm ⁻³)		Efficiency (%)
	before	after	
A1	1139	133	88,3
A2	1062	100	90,6
B1	1543*	315	79,6
B2	1205	456	62,2
C1	761	137	82,0
C2	711*	98	86,2
C3	595	116	80,5
C4	944	83	91,2

* data recorded with continual measurements.

Consequently, with efficiencies ranging from 79% till 91% and indoor radon activity concentration after remediation situated mainly below 200 Bqm⁻³ the three houses submitted to experiment could be considered radon 'free'.

6. General conclusions

As laid out in this thesis the measured concentration of thoron progeny in Romanian dwellings increased from 1.07 Bq m⁻³ (grab sampling) in 1996 to 1.6 in 2013 (solid state nuclear track detectors). The Romanian average thoron activity concentration of 58.7 Bqm⁻³ calculated from the total of 145 existing indoor measurements, which are described here, is 2.1 times lower than the Romanian radon mean of 126 Bqm⁻³. Nevertheless, data regarding thoron, radon and their progeny activity concentration are still rather scarce for a national statistic, which is why Romania is still debating the necessity of legislation in force regarding reference or action levels for these carcinogenic gasses. In order to overcome this deficiency a larger scale survey that would provide a national statistic about not only thoron and its progeny but also radon and its progeny indoor activity concentrations is to be encouraged and very much needed.

Feasibility studies within three different counties (Sălaj, Satu Mare and Bihor) pointed out thoron concentrations ranging between 41 Bqm⁻³ and 128 Bqm⁻³ with a mean concentration of 75 Bqm⁻³ which represent a lower activity compared to those reported by other countries and a low contribution to the effective dose, ranging between 0.003 mSv/y and 0.07 mSv/y. Further investigations focused on a particular target that is schools, came to confirm that thoron level is commonly below 100 Bqm⁻³ while radon activity is mostly above 100 Bqm⁻³ but without exceeding 500 Bqm⁻³. The seasonal effective dose calculated from thoron exposures, overlapping all three counties, ranged between 0.004 mSv and 0.05 mSv.

In order to assess the reliability of data our measuring systems were firstly tested within international exercises of intercomparison. The relatively low percentage distribution for thoron intercomparison showed that the quality for worldwide thoron measurements is still under development; our laboratory obtained generally lower results than the reference with a -41 PD for Low exposure and -38 PD for High exposure. Results of this intercomparison reveal as a first monitoring problem the detectors calibration errors. On the other hand, in situ intercomparison of two types of the most commonly used track detectors pointed out one important issue of radon monitoring which is represented by the aging process of the CR-39 chip that can cause a decrease in recorded concentration.

Indoor thoron investigations indicated a slightly higher activity of thoron in the vicinity of the walls and low activity towards the centre of the room, while radon concentration follows a relatively constant pattern indifferent to the distance from the wall. Surface emission measurements pointed out that the walls (building material) represent the source of thoron and the floor (filling) of radon.

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Diurnal investigations performed in a 50 years old adobe building provided a rather constant behaviour for Tn activity throughout the whole investigated interval. Unlike in the red brick building the radon activity was found to be constant during day time but decreasing consistently after midnight. Neither gas appears to be influenced in any way by the temperature or humidity.

Further research described the seasonal variations of indoor ^{220}Rn and ^{222}Rn concentration measured during summer (drought season) versus winter season while the second survey presents the seasonality between winter and summer (rainy) season. The survey comes to confirm the previous study conducted in Transylvania regarding radon seasonal variability indoor according to which radon presents high activities during winter season and low activities during the summer season. In case of thoron, the second set of data confirms the international database through the fact that indoor thoron activity concentration decreases during rainy season while the first step of the survey brought to attention a novelty in the field, the fact that indoor thoron activity concentration appears to present an increasing tendency at high atmospheric temperature.

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However, since knowledge is never enough, after two years of radon investigations three houses were chosen amongst those with indoor level higher than the action level suggested by authorities and submitted to radon mitigation. Testing of remediation techniques was based on efficiency accountancy by comparing annual activity concentrations (integrated measurements) found before and after implementation. Radon activity concentration significantly decreased after mitigation in all three locations, confirming that implementation of drainage piping is one of the most effective method for radon control.

List of publications

This thesis is based on the work contained in the following papers and submitted manuscripts:

1. Armencea (Mutoiu) E. S., Armencea A., **Burghele B.**, Cucuș (Dinu) A., Maloș C., Dicu T., Indoor Radon Measurements in Bacău County. Romanian Journal of Physics, Vol. 58, Supplement, pp.: S189–S195, 2013. **(IF: 0,414)**
2. **Burghele B.D.**, Cosma C., Intercomparison between radon passive measurements and active measurements and problems related to thoron measurements, Romanian Journal of Physics, Vol. 58, Supplement, pp.: S56-S61, 2013. **(IF: 0,414)**
3. **Burghele B.D.**, Cosma C., Măsurarea simultană a radonului și toronului cu detectori de urme – Situația actuală în lume. Ecoterra Journal of Environmental Research and Protection. 01/2011; 28:43-48. **(BDI)**
4. **Burghele B.D.**, Cosma C., Thoron and radon measurements in Romanian schools, Radiation Protection Dosimetry; Nov2012, Vol. 152 Issue 1-3, p.:38-41. **(IF: 0,909)**
5. **Burghele B.D.** and Moldovan M.C., Intercomparison of radon in soil instruments at reference site in Czech Republic. Annals of the West University of Timisoara – Physics Series (in press).
6. **Burghele B.D.**, Moldovan M., Papp B., Niță D.C., Rusu (Dumitru) O., Cucuș A., Cosma C., Sainz C. Neznal Matej, Neznal Martin. Applied techniques to diminish radon concentration in three dwellings of Băița-Ștei, România. Ecoterra Journal of Environmental Research and Protection, no. 33, pp. 7-12, 2012. **(BDI)**
7. **Burghele B.D.**, O privire de ansamblu asupra măsurătorilor de toron în România, Tendințe și cerințe de interdisciplinaritate în cercetare, Ed. Politehniun, Iași, pp.: UBB (1-8), 2013.
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11. Cucuș (Dinu) A., Cosma C., Dicu T., Begy R., Moldovan M., Papp B., Niță D., **Burghelle B.**, Sainz C. Thorough investigations on indoor radon in Băița radon-prone area (Romania) *Science of The Total Environment* 01/2012; 431:78-83. **(IF: 3,258)**
12. Cucuș (Dinu) A., Cosma C., Dicu T., Papp B., Begy R., Moldovan M., Niță D.C., **Burghelle B.**, Cîndea C., Fulea D., Sainz C., Neznal Martin, Neznal Matej. Radon diagnostic measurements in a pilot house from Baita Region, Romania. *STUDIA UBB AMBIENTUM*, LVI, 1, 2011, pp. 31-41. **(BDI)**
13. Frunzeti N., Moldovan M., **Burghelle B.D.**, Cosma C., Papp B., Popita G.E., Stoian L.C., Flux measurements of ^{222}Rn , CH_4 and CO_2 along with soil gas concentrations (^{222}Rn , CO , NO_2 and SO_2) over a methane reservoir in Transylvania (Romania). *Carpathian Journal of Earth and Environmental Sciences*, August 2013, Vol. 8, No. 3, p. 75 – 80. **(IF: 1,450)**
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1. VIth Hungarian Radon Forum and Radon in Environment Satellite Workshop, Veszprém, Hungary, 2011.
2. Ediția a II-a: Contribuții științifice în tehnologii și echipamente pentru evaluarea și protecția mediului, Arcalia, România, 2011.
3. Environment & Progress. Environment-Research, Protection and Management, Cluj-Napoca, România, 2011.
4. International Symposium on Natural Exposure and Low Dose Radiation. Epidemiological Studies. Hirosaki, Japan, 2012.
5. Terrestrial Radionuclides in Environment. International Conference on Environmental Protection, Veszprém, Hungary, 2012.
6. First East European Radon Symposium (FERAS), Cluj-Napoca, Romania, 2012.
7. The XIth International Workshop on Geological Aspects of Radon Risk Mapping, Prague, Czech Republic, 2012.
8. Environmental Legislation Safety Engineering and Disaster Management (ELSEDIMA), Cluj-Napoca, Romania, 2012
9. Workshop Internațional pe tema: Tehnici de remediere a radonului din locuințe. Harta de radon pentru regiunile de centru, vest și nord-vest ale României, Arcalia, România, 2012.
10. Physics Conference, TIM12, Timișoara, România, 2012.
11. Workshop: Tendințe și cerințe de interdisciplinaritate în cercetare. Prezentarea rezultatelor obținute de doctoranzi. Iași, România, 2012.
12. VIIth Hungarian Radon Forum and Radon in Environment Satellite Workshop, Veszprém, Hungary, 2013.

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