

MEASUREMENTS OF TERRESTRIAL GAMMA DOSE RATES AND RADON CONCENTRATIONS FROM INDOOR AIR AND WATER IN TRANSYLVANIA REGION

M. DOLHA^{1,2*}, A. TIMAR-GABOR^{1,2}, T. DICU^{1,2}, C. COSMA^{1,2}

¹“Babeş-Bolyai” University, Faculty of Environmental Science and Engineering, 30 Fântânele,
Cluj-Napoca, 400294, Romania

²Interdisciplinary Research Institute on Bio-Nano Sciences, “Babeş-Boyal” University,
42 Treboniu Laurian, Cluj-Napoca, 400271, Romania

*Corresponding author E-mail: monica.zeciu@ubbcluj.ro

Received December 16, 2015

Abstract. Terrestrial gamma dose rates have been measured using thermoluminescence method in Alba county. More than 150 dosimeters were placed in 52 locations. The obtained values ranged from 41 ± 2 to 91 ± 4 nGy/h, being in agreement with UNSCEAR Report. Radon concentrations for indoor air were performed in 12 locations from Alba county using CR39 detectors accompanied by radon groundwater measurements, being in accordance with WHO and EURATOM Treaty. Annual effective doses due to radon exposure were also calculated. Alongside with previous results, this work represents an important stage in the development of the high resolution environmental radioactivity map for Transylvania region.

Key words: thermoluminescence, gamma dose, radon, map.

1. INTRODUCTION

There are many areas on the Earth which exhibit elevated natural gamma dose rates caused by the geological setting or geochemical structure of different rocks. Also, the concentrations of natural radionuclides in the environment can be changed deliberately or accidentally by humans, and thus the situation should be managed in an active manner [1, 2]. As a consequence of these variations of gamma dose around the world, it is mandatory to have accurate estimations of gamma dose rates and carrying such systematic surveys is an obligation of our country as an European Union member state according to EC Treaty, Article 174.

Previous studies [3, 4, 5, 6] have demonstrated that thermoluminescence method along with a dosimetric system consisting of a Harshaw 3500 Reader and LiF:Mg,Cu,P detectors is capable of measuring doses of a magnitude from a few μ Gy to tens Gy.

^{222}Rn is a natural occurring radionuclide in a gaseous phase (formed from ^{226}Ra , which is a decay product of ^{238}U) with a half life of 3.8 days (a sufficient period for an independent migration in the environment). This radionuclide tends to concentrate in enclosed spaces like underground mines or houses. In spaces such as homes, basements and office buildings, higher levels of radon were observed and is considered an important contributor in the long-term dose exposure [6, 7]. Many European countries have undertaken different studies in order to estimate the risk of lung cancer associated with residential radon exposure [9]. In Romania similar studies have been conducted especially in radon prone areas such as Băița-Ștei (Bihar county) for indoor radon concentration [8]. More earlier results concerning radon concentration in air, water and soil have been reported [10, 11].

This paper aims to present the results obtained for Alba county with respect to gamma dose rates using thermoluminescence detectors. An additional purpose was to estimate the annual effective doses due to radon exposure. Since Romanian National Commission for Nuclear Activities Control in accordance with European requirements has limited public dose at 1 mSv/year above natural background, more specific studies are required [12]. Consequently, the present study brings an important contribution to the natural radioactivity database in Transylvania region.

2. MATERIALS AND METHODS

GAMMA DOSE RATES MEASUREMENTS

In order to obtain the gamma dose rates, LiF:Mg,Cu,P detectors have been used. This material is extremely sensitive at very low doses but also at high doses (1 μGy –10 Gy) [13, 14]. A number of 156 detectors (based on LiF doped with Mg, Cu and P, coded MCP-7, produced by TLD Poland) were used in the form of pellets of 4.5 mm diameter and 0.9 mm thickness. They were placed in 52 different locations from Alba County between March and July 2014, covering about 5200 km². Alba county relief is characterised by the presence of high hills and mountains.

In order to eliminate any residual signal from the memory of the detector, before placing the dosimeters into the environment, a standard annealing procedure was performed consisting of a heating of 10 minutes at 240°C. Each set, containing 3 pellets was placed at a distance of 1 meter above ground, as standard procedures require [15]. The time of exposure ranged from 21 to 125 days. A Harshaw 3500 TLD Reader was used for reading the detectors. For the calibration procedure, reference is made to a previous study of the authors [4].

RADON MEASUREMENTS IN AIR AND WATER

In the case of radon concentration measurements, CR-39 track etched detectors were used. They were placed in 12 locations (about 60 dwellings) for at least three months between March and July 2014. Subsequently to the exposure, alpha tracks have been counted and converted into indoor radon concentration using a Radosys system [16]. The results are shown in Table 2.

In the case of water measurements, the samples were taken from wells in bottles of 0.5 l and totally filled. ²²²Rn concentration was measured using the LUK-VR system consisting of a LUK-3A device and a VR-scrubber [17, 18]. The radon concentration dissolved in the water sample (0.3 l) is mixed with the air that is on the top of the water level within the scrubber volume. Further, air is transferred from the scrubber in order to be measured for radon using a Lucas cell method. A more detailed description of this method is extensively discussed by Cosma *et al.* [18].

3. RESULTS AND DISCUSSIONS

The results obtained for the 52 considered locations are given in Table 1. The mean gamma dose rate was found to be 62 nGy/h which is a normal value according to UNSCEAR 2008 (column no. 8 from Table 6, Annex B) [19]. Also, it can be noted that the dose rates were in the normal range of variation given by the report (total outdoor range gamma dose rate 52–163 nGy/h).

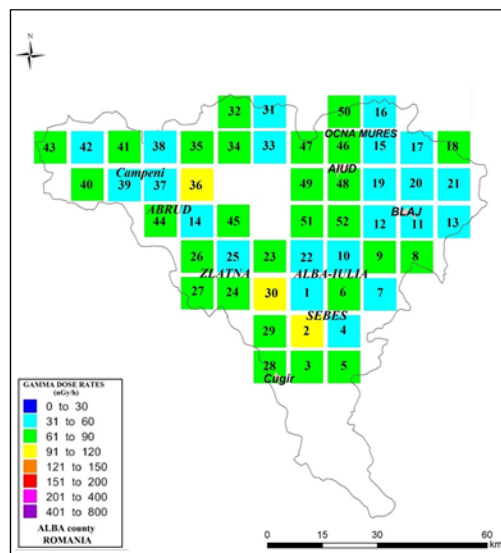


Fig. 1 – The high resolution map of gamma dose rates [nGy/h] for Alba County, Romania (the codes within every cell are given in Table 1).

Table 1

Gamma dose rates and external annual effective gamma doses

Location	Codes	Days of exp.	Gamma dose rate (nGy/h)	Ext. ann. eff. gamma dose (mSv)	Location	Codes	Days of exp.	Gamma dose rate (nGy/h)	Ext. ann. eff. gamma dose (mSv)
Lancram	1	125	55±3	0.07±0.01	Nadastia	27	21	61±9	0.07±0.01
Petresti	2	125	91±4	0.11±0.01	Vinerea	28	21	75±6	0.09±0.02
Rachita	3	125	78±6	0.09±0.01	Sibot	29	21	79±5	0.09±0.02
Sasciori	4	125	56±3	0.07±0.01	Mereteu	30	21	87±7	0.10±0.02
Capalna	5	125	76±6	0.09±0.01	Rimetea	31	21	53±7	0.06±0.02
Daia-Romana	6	125	68±6	0.08±0.01	Posaga de Jos 1	32	21	64±5	0.08±0.02
Spring	7	125	56±4	0.07±0.01	Posaga de Jos 2	33	21	51±3	0.06±0.01
Rosia de Secas	8	125	76±4	0.09±0.01	Brazesti	34	21	72±4	0.09±0.02
Ohaba	9	125	67±3	0.08±0.01	Baia de Aries	35	21	62±5	0.07±0.02
Seusa	10	125	57±3	0.07±0.01	Lupsa	36	21	84±7	0.10±0.02
Blaj	11	125	46±2	0.06±0.01	Bistra	37	21	56±3	0.07±0.01
Tiur	12	125	46±3	0.05±0.01	Garde	38	21	46±3	0.05±0.01
Valea Lunga	13	125	59±3	0.07±0.01	Vadu Motilor	39	21	56±3	0.07±0.01
Bucium	14	125	51±3	0.06±0.01	Vidra	40	21	63±4	0.07±0.01
VamaSeaca	15	118	57±7	0.07±0.02	Albac	41	21	73±5	0.09±0.02
Uioara de Sus	16	21	51±4	0.06±0.02	Garda de Sus	42	21	41±3	0.05±0.01
Farau	17	21	47±2	0.06±0.01	Arieseni	43	21	78±4	0.09±0.02
Silea	18	21	59±3	0.07±0.01	Abrud	44	21	65±5	0.08±0.02
LopadeaNoua	19	21	48±3	0.06±0.01	Mogos	45	21	66±6	0.08±0.02
Sancel	20	21	58±5	0.07±0.02	Aiud	46	21	68±5	0.08±0.02
Jidvei	21	21	41±2	0.05±0.01	Magina	47	21	65±4	0.08±0.02
Sard	22	21	50±4	0.06±0.01	Tifra	48	21	60±3	0.07±0.01
Metes	23	21	64±3	0.08±0.01	Geoagiu de Sus	49	21	63±3	0.08±0.01
Patrangeni	24	21	63±4	0.08±0.01	Unirea	50	21	72±6	0.09±0.02
Zlatna	25	21	55±3	0.07±0.01	Stremt	51	21	65±4	0.08±0.01
Izv. Ampoiului	26	21	62±8	0.07±0.01	Teius	52	21	65±4	0.08±0.01

The higher dose rate (91 ± 4 nGy/h) was observed in Petrești village. Surprising, this mean that value for Alba county is smaller as initially expected for this region from Transylvania, being significant smaller as average in Cluj and Bihor counties [20].

A high-resolution map of Alba County, Romania for gamma dose rates was plotted on the reference grid 10×10 km developed by Joint Research Centre of the European Commission [10] (Fig. 1). Radon measurements in water are presented in Table 2.

According to EURATOM Treaty 59/2013, all European countries must develop action plans regarding radon exposure until 2018. Many European countries have already established remedial action levels for indoor radon. The recommended level given by WHO (World Health Organization) in the case of indoor air should not exceed 100 Bq/m^3 . However, a limit of 300 Bq/m^3 is given. As can be observed from Table 2, the higher value has been measured in Căpâlna

village (476 Bq/m^3) that is above the remedial action level mentioned before. This can be explained by a poor ventilation, taking into account that during the monitoring period the investigated room was not used. For water samples, all measurements were under the limit (1000 Bq/l) given by [21]. It can be noted that the arithmetic mean obtained (93 Bq/m^3) is less than the one obtained by [10] by considering 158 dwellings from Alba County. This is due to the limited number of chosen houses in this study. To remark that a very recent study find a mean value of 115 Bq/m^3 (412 dwellings) and an average water value of 9.7 Bq/l (112 measurements) [22].

Table 2

Radon concentrations for indoor air and water samples in Alba County

Codes of locations	Indoor radon conc. (Bq/m^3)			Radon conc. in water (Bq/l)		
	A.M.*	S.D.*	Max.	A.M.	S.D.	Max.
1	94.3	22.4	118	3.9	3.2	7.6
3	44.7	22.1	70	13.7	9.6	24.4
4	40	–	40	10.3	0.6	10.8
5	255	153.8	476	–	–	10.5
6	95.5	79.7	208	7.8	4.1	11.6
7	65.3	41.2	113	3.1	1.3	4.4
8	91.5	47	133	6.7	0.5	7.1
9	89.7	43.4	127	7.2	4.5	10.2
10	83.8	18.5	107	–	–	2.6
13	90.7	42.4	162	3.4	13	4.3
40	36.3	20.6	58	–	–	3.4
44	50.7	47.3	104	–	–	2
Total	93	78	476	6.8	0.9	24.4

*A.M. – Arithmetic mean, S.D. – Standard deviation.

We have also carried out estimations of the annual effective doses for ingestion and inhalation of radon using the conversion factors given by UNSCEAR 2006 Report [23] in order to assess the effective exposure of humans to radon (See Table 3):

$$D_{inh} = C_{Rn} (\epsilon_r + \epsilon_d f) O, \quad (1)$$

where D_{inh} is annual effective dose for inhalation; C_{Rn} represent the mean annual radon activity concentration (Bq/m^3); ϵ_r and ϵ_d are dose conversion factors for radon gas and its short-lived progeny respectively, $\epsilon_r = 0.17(\text{nSv/h})/(\text{Bq/m}^3)$ and $\epsilon_d = 9(\text{nSv/h})/(\text{Bq/m}^3)$; f is the equilibrium factor between radon and its short-lived progeny, $f = 0.4$ for indoor environment and O is the occupational factor, $O = 7000 \text{ h/y}$.

$$D_{ing} = C_{Rn} \times F_{Rn} \times C_w, \quad (2)$$

where D_{ing} is annual effective dose for ingestion; C_{Rn} is the mean annual radon activity concentration (Bq/l); F_{Rn} is the committed effective dose per unit intake of radon in water for adults (10^{-8} Sv/Bq) and C_w is the water consumption rate (L/y) $C_w = 1$ l/day [23].

Table 3

Annual effective doses due to radon inhalation and ingestion

Codes of location	Ann. eff. dose (inhalation) (mSv)	Ann. eff. dose (ingestion) (mSv)
1	2.49	0.01
3	1.18	0.05
4	1.06	0.04
5	6.73	0.04
6	2.52	0.03
7	1.72	0.01
8	2.41	0.02
9	2.37	0.03
10	2.21	0.01
13	2.39	0.01
40	0.96	0.01
44	1.34	0.01
Total	2.28	0.02

Taking into account the reference level within the range of 3 mSv/year to 10 mSv/year proposed by ICRP 1993 [24] it can be observed from Table 3 that our results concerning the annual effective doses due to radon inhalation are normal and varies from 0.96 mSv/year to 6.73 mSv/year with a standard deviation of 1.52 mSv. In a previous study regarding radon measurements were reported doses due to radon inhalation ranging from 0.07 to 1.2 mSv which are lower than the results presented within this work [25]. Also, regarding annual effective doses due to radon ingestion, the results obtained ranged between 0.01 and 0.05 mSv/year (having a standard deviation of 0.01 mSv) being in perfect accordance with WHO [21] which gives a dose limit of 0.1 mSv/year.

4. CONCLUSIONS

Thermoluminescence detectors based on LiF:Mg, Cu, P were used for the assessment of gamma dose rates in Alba County, Romania. A high resolution map for this area concerning the gamma dose rates is presented.

Radon concentrations measurements were made within 60 dwellings from Alba County. Our results show that for people living in the investigated areas, radon concentrations from indoor air as well as for water are normal, being in accordance with WHO and EURATOM Treaty. Estimations of annual effective dose due to radon inhalation/ingestion were also performed. The results were in the normal range of variation according to ICRP 1993 Report and WHO.

Alongside with the measurements performed in a previously study from Cluj county, the present work represents a new stage in the development of the high resolution map of Transylvania region.

Acknowledgements. This work was supported by a grant of the Romanian National Authority for Scientific Research and Innovation, CNCS-UEFISCDI, project number PN-II-RU-TE-2014-4-1009. M.D., T.D. and C.C are also grateful for the financial support of CNCS-UEFISCDI, PN-II-PT-PCCA2011-3.2-1064 nr. 73/2012.

REFERENCES

1. B. Michalik, *It is necessary to raise awareness about technologically enhanced naturally occurring radioactive materials?*, Journal of Environ Monit **11**, 1825-1833 (2009).
2. Michalik B., Brown J. and Krajewski P., *The fate and behavior of enhanced natural radioactivity with respect to environmental protection*, Environ. Impact. Assess. Review **38**, 163-171 (2013).
3. M. Dolha-Zeciu, A. Timar-Gabor, A. Cameniță, D. Costin and C. Cosma, *Gamma background measurements by TL method: applications in locations with varied geological context*, Carpathian Journal of Earth and Environmental Sciences **8** (4), 109-114 (2013).
4. M. Dolha, A. Timar-Gabor, T. Dicu, R. Begy, M. Anton and C. Cosma, *A high-resolution map of gamma dose rates in Cluj county, Romania using LiF :Mg,Cu,P detectors*, Radiation Protection Dosimetry **162** (1-2), 14-19 (2014).
5. V. Benea, D. Vandenberghe, A. Timar, P. Van Den Haute, C. Cosma, M. Gligor and C. Florescu, *Luminescence dating of neolithic ceramics from lumea nouă, Romania*, Geochronometria **28** (1), 9-16 (2007).
6. Van Deynse, A. Cosma, C. Poffijn, A., *A passive radon dosimeter based on the combination of a track etch detector and activated charcoal*, Radiation Measurements **31** (1-6), 325-330 (1999).
7. C. Cosma, K. Szacsvai, A. Dinu, D. Ciorba, T. Dicu and L. Suci, *Preliminary integrated indoor radon measurements in Transylvania (Romania)*, Isotopes in Environmental and Health Studies **45** (3), 259-268 (2009b).
8. S. Darby, D. Hill, A. Auvinen, J.M. Barros-Dios, H. Baysson, F. Bochicchio, H. Deo, R. Falk, F. Forastiere, M. Hakama, I. Heid, L. Kreienbrock, M. Kreuzer, F. Lagarde, I. Mäkeläinen, C. Muirhead, W. Oberaigner, G. Pershagen, A. Ruano-Ravina, E. Ruosteenoja, A. Schaffrath Rosario, M. Tirmarche, L. Tomáček, E. Whitley, H.E. Wichmann and R. Doll, *Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies*, BMJ **330** (7485), 223 (2004).
9. C. Sainz, A. Dinu, T. Dicu, K. Szacsvai, C. Cosma and L.S. Quindos, *Comparative risk assessment of residential radon exposures in two radon-prone areas, Stei (Romania) and Torreldones (Spain)*, Science of the Total Environment **407** (15), 4452-4460 (2009).
10. C. Cosma, A. Dinu and T. Dicu, *Preliminary results regarding the first map of residential radon in some regions in Romania*, Radiation Protection and Dosimetry **155** (3), 343-350 (2013a).
11. C. Cosma, A. Dinu, B. Papp, R. Begy and C. Sainz, *Soil and building material as main sources of*

- indoor radon in Băița-Steii radon prone area (Romania)*, Journal of Environmental Radioactivity **116**, 174-179 (2013b).
12. Official Gazette of Romania, *Basic rules for radiological safety, Romania*, Dictate no.14, pp. 1-131 (2000).
 13. P. Olko, M. Budzanowski, P. Bilski, S. Milosevic, B. Obryk, E. Ochab, M. Simic, P. Stegnar, M.P.R. Waligorski, Z.S. Zunic, *Application of MCP-N (LiF:Mg,Cu,P) TL detectors in monitoring environmental radiation*, Nuclear Technology and Radiation Protection **1**, 20-25 (2004).
 14. P. Bilski, T. Cybulski, M. Puchalska and M. Ptazkiewicz, *Sensitivity loss and recovery for individual TL peaks in LiF:Mg,Ti and LiF:Mg,Cu,P after high-dose irradiation*, Radiation Measurements **43**, 357-360 (2008).
 15. P. Olko, *Advantages and disadvantages of luminescence dosimetry*, Radiation Measurements **45**, 506-511 (2010).
 16. A. Cucuș (Dinu), C. Cosma, T. Dicu, R. Begy, M. Moldovan, B. Papp, D. Niță, B. Burgehele and C. Sainz, *Thorough investigations on indoor radon in Băița radon-prone area (Romania)*, Sci. Total Environ **431**, 78-83 (2012).
 17. C.A. Roba, D. Niță, C. Cosma, V. Codrea, and Ș. Olah, *Correlations between radium and radon occurrence and hydrogeochemical features for various geothermal aquifers in Northwestern Romania*, Geothermics **42**, 32-46 (2012).
 18. C. Cosma, Moldovan M., T. Dicu and T. Kovacs, *Radon in water from Transylvania (Romania)*, Radiation Measurements, **43**, 1423-1428 (2008).
 19. United Nations Scientific Committee on the Effects of Atomic Radiation Report, *Sources and effects of ionizing radiation*, **Annex B – 1** (2008).
 20. M. Dolha, *Thermoluminescence dosimetry applied in environmental radioactivity monitoring*, PhD Thesis, 2016.
 21. World Health Organization, *Guidelines for Drinking-water Quality*, 4th Edition, 2011.
 22. C. Cosma, A. Cucos, T. Dicu and B. Papp, *Residential, soil and water radon maps in north-western part of Romania*, International Workshop on the *European Atlas of Natural Radiation*, 2015.
 23. United Nations Scientific Committee on the Effects of Atomic Radiation Report, **Annex E** (2006).
 24. ICRP, *Protection against Radon-222 at Home and at Work*, ICRP Publication **65**. Ann. ICRP **23** (1993).
 25. D.C. Nita, M. Moldovan, T. Sferle, V.D. Ona, B.D. Burgehele, *Radon concentrations in water and indoor air in north-west regions of Romania*, Romanian Journal of Physics **58**, S196-S201 (2013).