Babeș-Bolyai University
Faculty of Environmental Science and Engineering

THERMOLUMINESCENCE DOSIMETRY APPLIED IN ENVIRONMENTAL RADIOACTIVITY MONITORING

SUMMARY

MONICA ZECIU(DOLHA)

Promoter:
Prof. Dr. Constantin Cosma

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INVITAŢIE

Cu respect vă invităm în data de 15. 01. 2016, ora 12:00, în Amfiteatrul Iustinian Petrescu din cadrul Facultăţii de Ştiinţa şi Ingineria Mediului, Universitatea Babeş-Bolyai, Cluj-Napoca, str. Fântânele nr. 30, să participaţi la şedinţa publică de susţinere a tezei de doctorat intitulată "Dozimetrie prin termoluminescenţă aplicată în monitorizarea radioactivităţii ambientale", elaborată de Monica Zeciu (Dolha), conducător de doctorat Prof. dr. Constantin Cosma.

Cluj-Napoca, 2016

The results presented in this thesis are focused on developing a reliable TL dosimetric system for implementing environmental gamma dose monitoring in Romania.

The research discussed in the present thesis has been carried out at the Laboratory of Luminescence Dating and Dosimetry, Center of Environmental Radioactivity and Nuclear Dating, Interdisciplinary Institute on Bio-Nano-Sciences, Babes-Bolyai University, Cluj-Napoca

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CONTENTS

1. INTRODUCTION

Thermoluminescence (TL) dosimetry technique applied in studies of environmental radioactivity using passive termoluminescent detectors has shown significant development in the last decade (**e.g**. **Vandecasteele, 2004; Olko et al., 2004; Karunakara et al., 2014, Warnery et al., 2015**). Since natural gamma dose rates presents important variations from one location to another it is mandatory to make accurate estimations in order to correctly quantify a potential radioactive contamination is case of an unwanted nuclear event (**UNSCEAR 2008 Report**).

The main purpose of this thesis was to establish a reliable dosimetric system in order to be able to provide accurate results for environmental effective gamma dose rates with the goal of developing high resolution maps of gamma dose rates according to the European Environmental Policies. Environmental gamma dose measurements according to Romania's obligations as an EU Member State (EC Treaty, art. 174, section 1) were carried out. Assessing the environmental gamma dose rates represents an action of preserving the quality of the environment. Also, by knowing how environmental gamma dose varies from one location to another, in case of an unexpected nuclear event, the database obtained would be an important prerequisite in order to establish human exposure and the necessary measures.

Aspects on thermoluminescence phenomena and natural radioactivity are discussed. A description of the dosimetric system with respect to TL detectors, the reader, calibration procedure and dose determination is presented. The applications of thermoluminescence dosimetry in environmental monitoring are further detailed. These studies were carried out in the framework of this thesis and published in two peer-reviewed articles as well as work which is under preparation.

Within the first study seven locations from Romania with different geological context in order to establish a connection between ambiental gamma dose rate in air and geological setting were investigated. Our results indicated the correlation between the geological substrate and the gamma dose rate as in an

investigated granitic area, the measured dose rates were about 70% higher than the national average.

A detailed database for natural gamma dose rates in Cluj County was established. The obtained results were used to create a high resolution map for this area according to European Environmental Policies.

In a subsequent study, the area of investigation was extended to Alba County where external annual effective gamma dose were computed as well. Alongside with these measurements, radon concentrations for indoor air and water were determined and annual effective doses for ingestion and inhalation of radon were calculated.

The presented studies have prooven the potential of thermoluminescence dosimetry as a tool for environmental monitoring studies and final conclusions are discussed.

2. LUMINESCENCE PHENOMENA

2.1. Introduction

Luminescence phenomena is the emission of light from solids called phosphors. This emission does not include black body radiation. Themoluminescence is the emission of light from certain insulators or semiconductors when heated. The emitted radiation represents the release of an amount of energy stored in solid by different ways of stimulation of its electronic system. Luminescence centres and considered to be atoms, ions or groups of ions situated in the vicinity of a lattice defect. These impurities are created by the insertion of an activator (an impurity atom) or by creating a vacancy. The transition of an excited luminescence centre into its ground state is accompanied by light emission. This is called luminescence. Thermoluminescence is a form of luminescence and is the thermally stimulated emission of light following the previous absorption of energy from nuclear radiation (**Furetta and Weng, 1998**).

2.2. The mechanism of luminescence

The processes that occur during luminescence production are not yet fully understood. However, the following simplified representation does allow understanding how luminescence is produced. Taking into account that luminescence is related only to materials that have more or less order in their structure the best suitable materials would be crystals and some vitreous materials. These kinds of materials are either semiconductors or insulators. This implies that the main delocalized energy states or bands are separated by an energy gap. However, natural crystals contain defects such as impurities or missing atoms or ions which give rise to localized states or intermediary energy levels inside the energy gap.

In luminescence phenomena three important steps can be distinguished (**Figure 2.1**):

i) **Irradiation**. After the interaction of ionizing radiation with a crystal, electrons from the valence band get enough energy to leave this band and move to the conduction band. For every electron that is transferred into conduction band a hole is assigned in the valence band. However, it is not possible for electrons to remain accumulated in the conduction band. Thus, these electrons have two possibilities of migrating: to return into the valence band or to get trapped by a defect. The same process applies for holes.

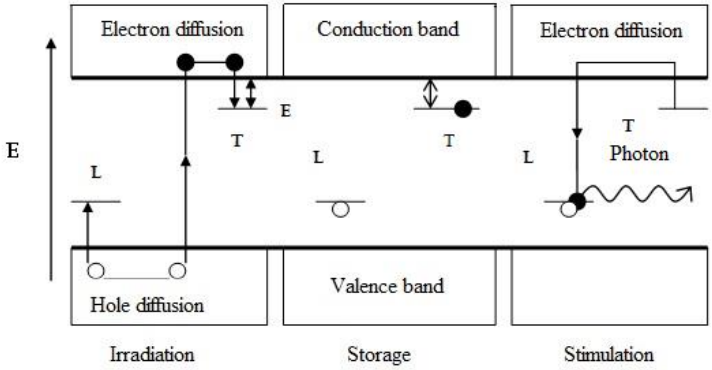

Figure 2.1. *Schematic representation of luminescence processes (adapted from* ***Aitken, 1998****) – T represents electron traps while L represents luminescence centres.*

ii) **Storage**. The energy of the nuclear radiation is now partially stored in the crystal lattice (trapped electrons). Energy (E) is usually referred to as the trap depth.

iii) **Eviction**. If the crystal is now exposed to heat or light the electrons would absorb enough energy to jump into the conduction band again. They can be trapped again or they can recombine with the holes in the recombination centres. If this transition will result in the emission of a photon, luminescence is present. In the case of thermoluminescent detectors the amount of this emmited light emission is proportional with the accumulated dose.

Given thermal stimulation, as this is the case of thermoluminescence, the probability that electrons are released from the trap (T) is governed by Boltzmann's statistics:

$$p = s \times \exp(\frac{-\Delta E}{kT}) \tag{2.1}$$

where s is frequency factor, ΔE – energetic difference between levels, T – temperature. It can be observed that the higher the temperature the deeper traps can be accessed.

The present model above is referred to as the general one trap model (GOT). There are more than one type of traps so this representation is being simplified. Also, exposure to radiation can produce more defects, but is not the case since we work with low radiation doses from the environment.

3. NATURAL RADIOACTIVITY

3.1. Naturally occurring radioactive materials

Natural radioactivity originates from extra-terrestrial sources as well as from radioactive elements in the earth's crust. About 340 nuclides have been found in nature, of which about 70 are radioactive and are found mainly among the heavy elements. All elements having an atomic number greater than 80 possess radioactive isotopes, and all isotopes of elements heavier than number 83 are radioactive (**Eisenbud and Gesell, 1997**).

The natural radioactivity has three major sources:

- primordial radionuclides – their half-life was sufficiently long for them to remain since their creation
- secondary radionuclides – are derived from the radioactive decay of the primordial nuclides, such as the radioactive nuclides produced in the radioactive series of ^{238}U, ^{233}Th and ^{235}U.
- cosmogenic radionuclides – are continuously produced by nuclear reactions produced by the bombardment of stable nuclides by cosmic rays mainly in atmosphere.

These natural radionuclides are known as Naturally Occuring Radioactive Materials –

NORM).

Ionising radiation sources for humans is illustrated in **Figure 3.1**. It can be observed that natural sources that contributes to annual effective gamma dose at global level is about 65% from the total ionizing radiation sources.

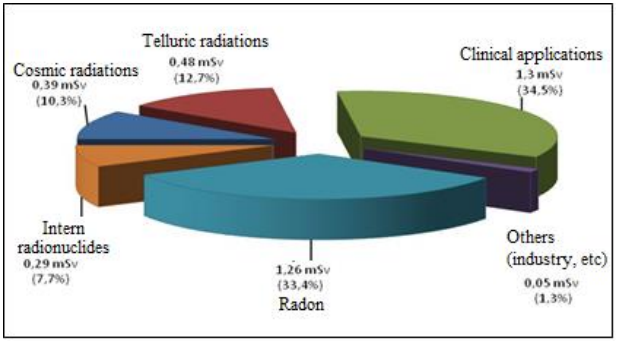

Figure 3.1. *Ionising radiation sources contribution to annual effective gamma dose at global level (Adapted from* ***UNSCEAR 2000****)*

3.2. High Background Radiation Areas (HBRAs)

A high background radiation area is defined as a dwellings complex where the effective dose received by population (from cosmic radiation, soil natural radioactivity, indoor and outdoor air, water and food products) exceeds a certain threshold (Eisenbud and Gesell, 1997). Annual effective dose are classified as: low: ≤ 5 mSv/year; intermediate: 5-20 mSv/year; high: 20-50 mSv/year and very high: > 50 mSv/year (**Hendry et al., 2009**).

Figure 3.2 presents the high background radiation areas along with average annual effective dose and maximum.

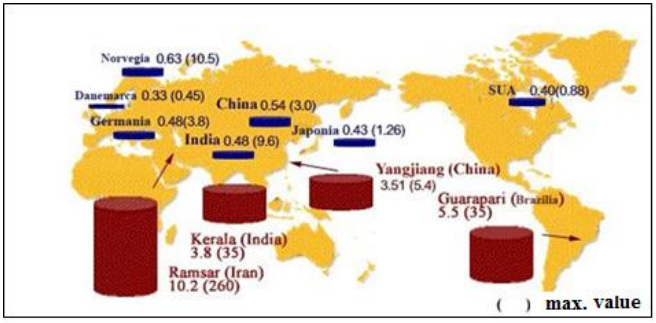

Figure 3.2. *Areas with high background radiation (mSv/year; mean value and ( ) - maximum value)(adapted from* ***UNSCEAR 2000****)*

3.3. Radon-prone areas

Radon and thoron are continuously generated into earth's crust by the decay of U and Th frm various types of rocksRadon concentration in water varies from one location to another. Surface water contain less than 2000 Bq/m^3, meanwhile groundwater can contain a concentration of radon between 20-44.000 Bq/m^3 (**Kronfeld, 2004**). On the other hand, the UNSCEAR Report from 2008 gives a reference value of 1000 Bq/m^3. Radon easily penetrates into dwellings throughout walls fissures, concrete floor or drainage pipes contributing to the increment of Radon concentration for indoor air.

The magnitude of the radon problem largely depends on geology, specifically on the ^{226}Ra content and the permeability of the rock formation. Other factors that play an important role include the construction techniques of buildings, and the climate. A strategy to control radon exposure should address the whole population, both at home and in the workplace, while devoting special efforts and resources to the individuals most at risk (**Garcia-Talavera et al., 2013**).

A very high radon level can be found in a building regardless of its location, but certain areas are much more prone to give rise to high concentrations in dwellings. Consequently, the identification of such areas is a key issue in an effective regulatory control of radon exposure (**Garcia-Talavera et al.,2013**).

There is no consensus definition of a radon-prone area. Generically, the 2007 Recommendations of the ICRP define it as an area in which the concentration of radon in buildings is likely to be higher than the national average. In some countries, this definition is based on a given fraction of dwellings exceeding a regulatory reference level. The most straightforward method to delineate radon-prone areas is based on actual in-house radon measurements (**Miles 1997, Andersen et al 2001, Friedmann 2012**). Because it requires a relatively large number of indoor radon measurements and a sampling density that is representative of the building stock, this method is only achievable nationwide when a significant fraction of all dwellings across the country has been monitored for radon (**Garcia-Talavera et al., 2013**).

Examples of radon-prone areas around the world were identified in different countries such as Canada (**Ford et al., 2000**), Sweden (**Akerblom 1986**), Germany (**Kemski et al., 2001**), France (**Ielsch et al., 2010**), Northern Ireland (**Appleton et al., 2011**), Norway (**Smethurst et al., 2008**), Băița-Ștei, Romania (**Cucoș (Dinu) et al., 2012, Cosma et al, 2013b, 2013c**).

4. THE THERMOLUMINESCENCE DOSIMETRIC SYSTEM

4.1. Introduction

Thermoluminescence dosimetry is a solid state dosimetric method, therefore, requiring calibration. The successful application of this method is dependent of the characteristics of a whole dosimetric system which comprises of: TL dosimeters, TL reader, calibration sources and mathematical evaluation of the results.

The main requirements for a dosimetric system according to **ISO/IEC 61066/2006** are presented in **Table 4.1.** Reproducibility is the ability of the system to give similar results when measurements are made according to the same conditions. Reproducibility is a component of precision, alongside with repeatability and is usually reported as standard deviation. Linearity is a measure of how the output of the system changes in response when changing the input. Detection limit is the lowest dose that can be detected by the system considered and is usually reported as three times standard deviation. The last requirement discussed is stability. It is the property that describes how the system is behaving under various conditions (climatic, handling, etc).

ISO/IEC 61066 requirement	ISO/IEC 61066 values
Reproductibility	max 7.5%
Linearity	max 10%
Detection limit	max 0.1 mSv
Stability	5% for 30 days, in normal conditions; 10% for 80 days in normal conditions

Table 4.1.*.Main requirements for a dosimetric system (Adapted from* ***Stochioiu et al., 2008)***

4.2. TL detectors (MCP-7 and MCP-N) and equipment (HARSHAW Reader)

In the case of TL materials, the most used phosphors are represented by lithium fluoride (LiF), lithium borate ($Li_2B_4O_7$), calcium sulphate ($CaSO_4$) and

calcium fluoride (CaF_2). Lithium fluoride along with different dopants is one of the most common materials used for TL synthesized detectors.

The detectors considered in this thesis are LiF doped with Mg,Cu and P (MCP) (**Figure 4.1**). There are two MCP types: MCP-N (with natural abundance of ^{6}Li 7.5% and ^{7}Li 92.5%) and MCP-7 (where ^{7}Li is enriched to 99.9%). This dosimeter, was found to be, according to UNSCEAR Report from 2008, the most suitable detector capable to be used *in vivo* dosimetry for clinical applications but not only, being as well, extremely adequate for environmental dosimetry. LiF:Mg,Cu,P shows a very high sensitivity, low susceptibility at humidity, good linearity (1μGy to 10 Gy) and a negligible fading (the loss of information).

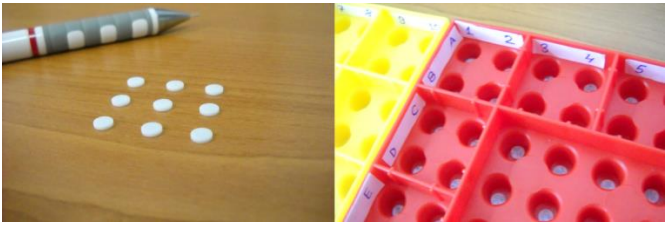

Figure 4.1. *LiF:Mg,Cu,P detectors*

In order to read the information stored in the lattice of the detectors, TL reading equipment is required. For this purpose, a Harshaw TLD Model 3500 Reader was used (**Figure 4.2**). It includes a simple drawer for a single element TL detector, a linear, programmable heating system, and a cooled photomultiplier tube with associated electronics to measure the TL light output. The operational software, Thermo Scientific^{TM}WinREMS^{TM}, which runs on a separate computer, provides the user interface, the reader control and the applications software (**http://www.thermoscientific.com**)

Figure 4.2. *Harshaw 3500 TLD Reader*

4.3. Calibration procedure

The calibration procedure is carried out with the purpose to determine the sensitivity of the detectors in order to calculate the gamma dose acquired. ^{60}Co ($E_{\gamma 1}$=1.17MeV, $E_{\gamma 2}$=1.33MeV and a half life of 5.3 years) is one of the most used radioactive sources for calibration purposes and was also used within the studies presented in this thesis.

4.4. Dosimetric quantities and units for dose determination

4.4.1. Absorbed dose

Absorbed dose, *D*, is a non-stochastic quantity applicable to both indirectly and directly ionizing radiations. This quantity is defined as the mean energy dε imparted by ionizing radiation to matter of mass dm in a finite volume V by:

$$D = \frac{d\mathcal{E}}{dm} \qquad (4.1)$$

In the international system of units this quantity is expressed in terms of Joule per Kilogram named Gray (Gy). Absorbed dose rate is given by the ratio between dose and time:

$$\dot{D} = \frac{dD}{dt} \tag{4.2}$$

4.4.2. External annual effective gamma dose (E)

The external annual effective gamma dose (E) can be calculated according to UNSCEAR Report from 2008 as follows:

$$E = D \times 0.7 \times 0.2 \tag{4.3}$$

where D is gamma dose rate; 0.7 is the conversion factor from absorbed dose in air in external effective annual gamma dose in adults; 0.2 reflect the outdoor occupancy component. This quantity is expressed in Sieverts (Sv).

4.4.3. Effective dose for radon inhalation and ingestion

The health risk due to exposure to ^{222}Rn (radon) and ^{220}Rn (thoron) comes from the inhalation of the short-lived decay products and the resulting alpha particle irradiation of the bronchial airways. The radiation dose delivered to the respiratory system and the resulting potential health detriment are a complex function of the radon decay product aerosol characteristics and the physiological parameters of the exposed individual.

In dosimetric approach, the effective doses for ingestion and inhalation can be calculated with the formulas and the conversion factors given by UNSCEAR 2008:

$$D_{inh} = C_{Rn} (\varepsilon_r + \varepsilon_d f) O, \tag{4.4}$$

where D_{inh} is annual effective dose for inhalation; C_{Rn} represent the mean annual radon activity concentration (Bq/m^3); $\varepsilon_r$and ε_d are dose conversion factors for radon gas and its short-lived progeny respectively, ε_r=0.17(nSv/h)/(Bq/m^3) and ε_d=9(nSv/h)/(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 h/y.

$$D_{ing}=C_{Rn}\times F_{Rn}\times C_w, \quad (4.5)$$

where D_{ing} in 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.

5. STUDIES CONCERNING NATURAL RADIOACTIVITY IN ROMANIA

Chapter based on:

Dolha M., Timar-Gabor A., Dicu T. and Cosma C., 2015. Measurements of terrestrial gamma dose rates and radon concentrations from indoor air and water in Transylvania region Article submitted. Romanian Reports in Physics

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(1-2), 14-19.

Zeciu-Dolha M., Timar-Gabor A., Cameniţă A., Costin D., 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.

5.1. Gamma background measurements by TL (thermoluminescence) method: Applications in locations with varied geological context

INTRODUCTION

The aim of this study was to establish that our thermoluminescence laboratory is suitable for natural gamma dose monitoring with the ultimate goal to apply this method for "Radon map (residential, geogenic, water) for Center, West and Northwest regions from Romania" research project. The importance of these measurements is two–fold:

(i) outdoor gamma dose rates obtained will allow establishing a very high resolution database for natural gamma dose rates in the investigated region that can be integrated into the seasonal maps of natural terrestrial dose rates at the European level (**Szegvary et al., 2007**)

(ii) (ii) the corroboration of indoor radon concentration levels and indoor as well as outdoor gamma dose rates can serve for realistic input data for the effective dose received by the population in the investigated area to be computed (**UNSCEAR 2000**).

Seven locations with varied geological context have been selected (**Figure 5.1**) (Arad, Cluj-Napoca, Bologa, Miercurea-Ciuc, Predeal, Belis, Sinaia). Ultrasensitive TL dosimeters, LiF:Mg,Cu,P were used in order to quantify the gamma doses.

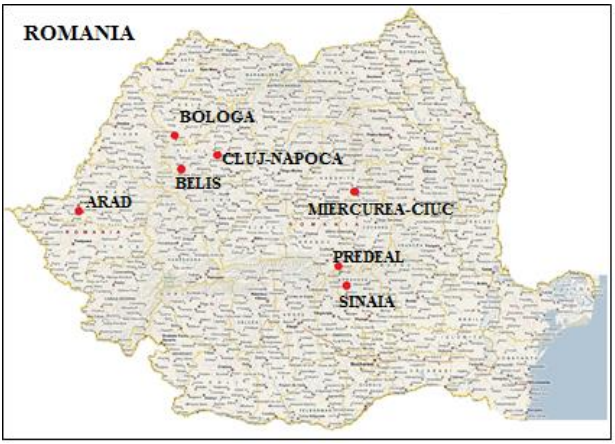

Figure 5.1. *The seven selected locations-Romania*

SELECTED LOCATIONS

Arad is a city situated on the right bank of the Mures river in the Banat region of western Romania. The geology of the area is characterized by up to 100 m thick Holocene and Pleistocene sediments (sands, gravels, clays, and loess) underlying Neogene sedimentary units composed by sands and clays having up to 3000 m thick (**Mutihac, 1990**). Located in the Somesul Mic river valley, Cluj-Napoca metropolitan area lies at the confluence of the Apuseni mountains, the Someş plateau and the Transylvanian plain. Substrate geology of this region is represented by holocene alluvial deposits (Transylvanian Basin) formed by sands, gravels underlying Upper Eocene-Lower Oligocene marine and continental deposits (limestones, marls, sandstones, clays, and gypsum) (**Baciu and Filipescu, 2002**). Bologa village is situated at the confluence of the Crişul Repede and Henţ (Sebeş or Săcuieu) rivers (the confluence is known as "guraapelor" - "the mouth of the waters" - in the local toponymy) and at the foot of the Vlădeasa mountains (1863 m), part of the Apuseni Carpathians. In this region there is evidence of the presence of Paleocene dacites and andesites (North Apuseni Mountains) locally covered by thin alluvial deposits (sands and gravels) along the streams (**Ianovici et al., 1976**).

Miercurea Ciuc city is situated at an altitude of 662 m, on the river Olt, in Ciuc depression. The geology of the area is characterized by up to 40 m thick Holocene and Pleistocene sediments (clays, sands, gravels, and loess) deposited in an intramontane basin underlying Lower-Middle Pleistocene pyroxene and amphiboles andesites (eastern Carpathians) (Mutihac, 1990). Predeal is situated in the Prahova Valley, in the southern part of Brașov County. Neighboring towns include Azuga to the south, Bușteni to the southwest, Râșnov to the northwest and Brașov to the north. The town is located in a mountainous area, with the Piatra Mare mountains to the north, the Bucegi mountains to the southwest and the Postăvarul massif to the northwest. Another town considered in this study is Sinaia which is also a mountain resort in Prahova County, Romania. It is located at about 60 km northwest of Ploiești and 50 km south of Brașov, in a mountainous area on the Prahova River valley, just east of the Bucegi Mountains. The altitude of the town varies between 767 m and 860 m. From a geological viewpoint, the areas are characterized by the presence of the lower Cretaceous flysch deposits (Eastern Carpathians) made by sandstones, shales, and limestones (Mutihac, 1990). Belis village is extended north-west slopes of the mountains Gilău and south-southeast of Magura Călăţele Apuseni mountains, on the banks of the anthropogenic Fântânele Lake of the upper Warm Somes river. The village is situated at an altitude of 1,050m. The geology of the area is characterized by the existence of upper Paleozoic granitoid rocks of Muntele Mare magmatic body intruded in metamorphic rocks (North Apuseni Mountains) (**Balintoni, 1997**).

MATERIALS AND METHODS

In order to measure gamma dose rates, ultra-sensitive LiF:Mg,Cu,P dosimeters (code MCP-N and MCP-7 produced by TLD Poland) were used, in the form of 4.5 mm diameter and 0.9 mm thickness pellets. The standard annealing procedure by heating the dosimeters at 240°C for 10 minutes was performed before placing the dosimeters in the environment. Seven sets of dosimeters (at least 2 MCP-N pellets and at least 3 MCP-7 pellets for each location) were packed in closed

plastic bags and coded. Every detector was wrapped in aluminum foil in order to have humidity and light protection and placed at a distance of 1 meter above ground, according to standard procedures (**Olko, 2004**). The time exposure was set to at least 3 weeks. After this period, TLDs were collected and brought into the laboratory and read with a Harshaw 3500 TLD Reader. The time temperature profile (TTP) for dosimeters reading was selected by having four regions of interest in the glow curve (ROI 1: first 50 channels, ROI 2: channel 50 to channel 120, ROI 3: channel 120 to channel 155 and ROI 4 channel 155 to channel 200). The heating rate was 5°C/s with the maximum temperature of 220°C (for 20 seconds). The third region of interest has been used for integration, as in this region the main peak at 210°C of LiF:Mg,Cu,P is registered. Calibration was performed using the ^{60}Co radioactive source from Physics Faculty, Babes-Bolyai University. Background subtraction was performed using the response of a batch of freshly annealed dosimeters.

RESULTS AND DISCUSSION

Background signal was determined for both types of detectors and was found to be equal to 0.29 nC for MPC-N type (**Figure 5.2.a**) and 0.32 nC for MCP-7 type (**Figure 5.2.b**). The lowest limit of detection for our system can be estimated as a ratio between there times the standard deviation of background signal (nC) and the sensitivity of the dosimeters (μC/mGy) and was found to be equal to 304 nGy for MCP-N detectors and 175 nGy for MCP-7 detectors. This is a very realistic value taking into account the lowest detectable dose for these detectors is around 60 nGy (**Olko, 2004**). **Figure 5.3.a** and **5.3.b** presents calibration procedure. It can be observed that the response of MCP is highly linear.

Figure 5.4 presents a typical glow curve for a MCP-N detector exposed at Belis. The integral TL response in the region of interest is 57 nC. It can be noticed that this value is 2 orders of magnitude higher than the background recorded for this type of detector. This result confirms that the TL method using MCP detectors is ultra-sensitive and can be applied for monitoring environments with low dose rates (tens of nGy/h) for exposure times of less than a few weeks.

The determined gamma dose rates for the investigated locations are presented in **Table 5.1** and plotted for the sake of clarity in **Figure 5.5**, where these values are compared to the gamma dose rate information available at the moment for Romania in the **UNSCEAR 2008 Report.**

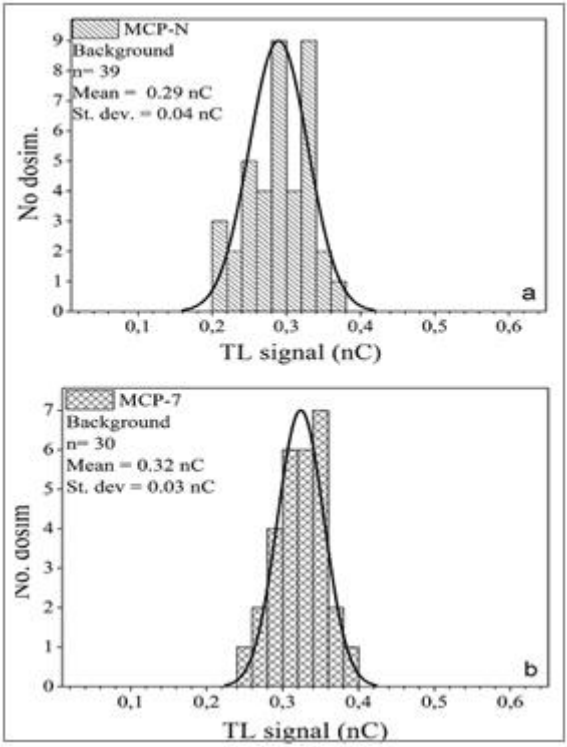

Figure 5.2. *Histogram of background signal for MCP-N (a), respectively MCP-7(b) detectors. It can be observed that the distribution for the batch of detectors is normal and background signals are very small (nC).*

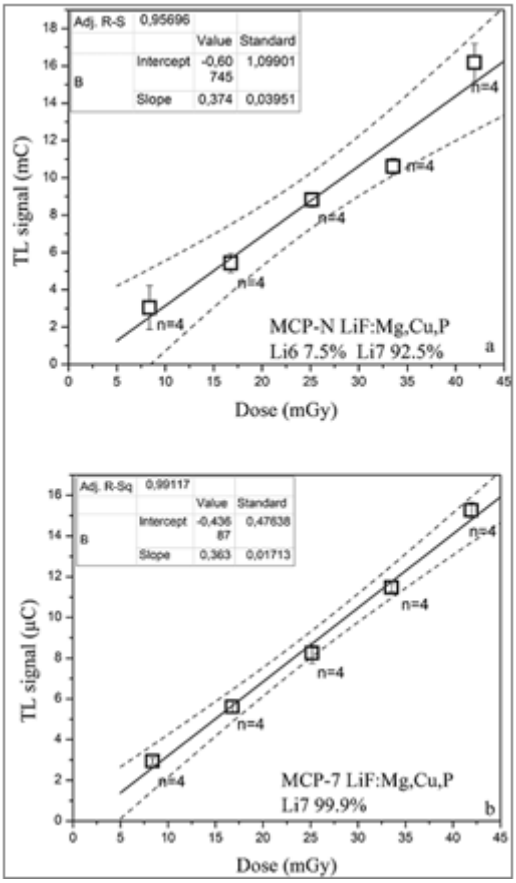

Figure 5.3. *Calibration of MCP-N and MCP-7 TL detectors respectively. The number of detectors used for each dose point is denoted in the graph. It can be noted that the dose response to ^{60}Co is highly linear (solid line).Dashed lines represent the 95% confidence limits.*

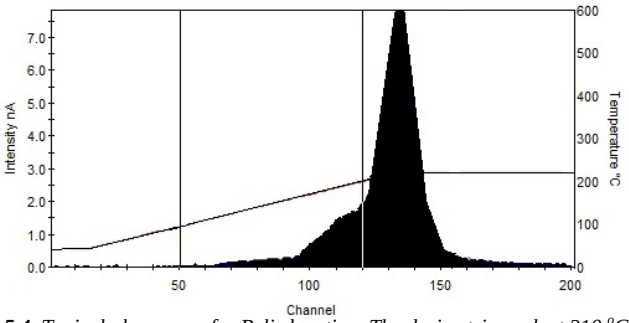

Figure 5.4. *Typical glow curve for Belis location. The dosimetric peak at 210 ^{o}C can be observed in the third selected region of interest.*

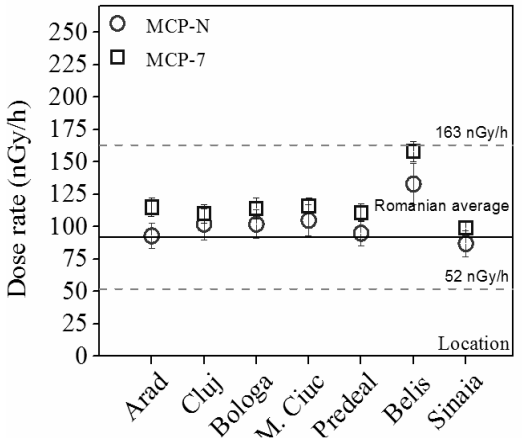

Figure 5.5. *Gamma dose rates obtained for the 7 locations considered. The continous line represents the Romanian average for gamma dose rates – 92 nGy/h (UNSCEAR Report 2008). The two dashed lines represent the interval of variation for gamma dose rates in Romania – 52-163 nGy/h (**UNSCEAR Report 2008**).*

The gamma dose (μGy) was obtained as a ratio between the signal light (nC) and the sensitivity of our detectors (μC/mGy) (previously described throughout calibration procedure):

$$D = \frac{sign}{Sens} \tag{5.1}$$

Where D is the gamma dose, $Sign.$ is the signal light obtained and $Sens.$ is the sensitivity of MCP-7 detectors obtained.

Gamma dose rate was obtained with:

$$Dr = \frac{D}{t} \tag{5.2}$$

where Dr is the gamma dose rate, D is the gamma dose and t is time.

Except Belis location, for all other locations, results were very close to the Romanian average which is 92 nGy/h. It can be noted that the dose rates were in the normal range of variation given by **UNSCEAR Report 2008**, which is 52-163 nGy/h. The higher dose rate observed for Belis location (133±16 nGy/h for MCP-N detectors and 158±8 nGy/h for MCP-7 type) is explained by the existent geological substrate in this area, a granitic one, with rocks rich in uranium and potassium that increase the existing natural background (**Moldovan et al., 2013**). The good agreement between the dose rates determined using MCP-N and MCP-7 detectors further confirms the robustness of the applied method.

Table 5.1. *TL signal, gamma dose and gamma dose rate obtained for each investigated location*

Location	Days of exp.	Type of dosim.	Mean signal (nC)	Bck. corrected signal (nC)	Signal /day (nC)	Gamma dose/day (μGy)	Gamma dose rate (nGy/h)	Geological setting
Arad	24	MCP-N	20.34	20.05	0.83	2.23±0.24	93±10	*Sediments*
		MCP-7	24.42	24.10	1	2.77±0.17	115±7	
Cluj	40	MCP-N	36.96	36.67	0.92	2.44±0.30	102±12	*Alluvial deposits*
		MCP-7	38.80	38.48	0.97	2.65±0.16	110±7	
Bologa	40	MCP-N	36.96	36.67	0.92	2.44±0.26	102±11	*Volcanic rocks*
		MCP-7	40.09	39.77	0.99	2.74±0.18	114±8	
M.-Ciuc	20	MCP-N	19.12	18.83	0.94	2.51±0.28	105±12	*Sediments*
		MCP-7	20.51	20.19	1	2.78±0.15	116±6	
Predeal	23	MCP-N	19.96	19.67	0.85	2.28±0.24	95±10	*Sedimentary rocks*
		MCP-7	22.60	22.28	0.97	2.67±0.16	111±7	
Belis	40	MCP-N	48.23	47.94	1.2	3.20±0.39	133±16	*Magmatic rocks*
		MCP-7	55.48	55.16	1.4	3.80±0.20	158±8	
Sinaia	23	MCP-N	18.20	17.91	0.78	2.08±0.24	87±10	*Sedimentary rocks*
		MCP-7	20.20	19.88	0.87	2.38±0.13	99±5	

CONCLUSIONS

Highly sensitive MCP's (LiF:Mg,Cu,P) thermoluminescent detectors were applied for environmental monitoring of radiation doses. Due to the high sensitivity of MCP detectors, our dosimetric system allows for short term (a couple of weeks) integrated measurements to be performed, as well as long term (months and up to years) surveys of environmental doses to be carried out. The lowest gamma dose rates have been recorded in a location characterized by sedimentary rocks (Sinaia, Predeal) and thick sediments units (Arad).Alluvial deposits (Cluj), thin sediments layers (Miercurea Ciuc) and volcanic rocks (Bologa) show lower to medium values. One single location (Beliş) indicates a very strong correlation between geological setting and gamma dose rates: the highest values are due to the presence of granitic rocks composed by radioisotopes bearing minerals (feldspars and other silicates). Our relatively short term experiments, conducted for an exposure period of about three weeks have shown that this exposure time is sufficient for a clear identification of locations where the environmental dose rates are above the average value due to the natural geological substrate. Because of the low cost of the individual dosimeters, our system will allow measuring environmental gamma dose rates simultaneously at a large number of locations. Thus, it can be concluded that the successful testing of the investigated method will be a basis for obtaining realistic input data for the calculation of the effective dose received by the population in Romania. This is of paramount importance as carrying systematic surveys at a national level regarding natural exposure of the population to ionising radiation is one of the obligations of our country as a European Union member state.

5.2. A high resolution map of gamma dose rates in Cluj county, Romania using LIF:Mg,Cu,P detectors

INTRODUCTION

This study aimed to develop the first high resolution map of gamma dose rate in Cluj County, Romania using passive thermoluminescentdosemeters. The external annual effective dose received by the population in the investigated area was also computed. Additionally, the specific activities of ^{238}U, ^{232}Th, ^{40}K from 5 soil samples was determined using high resolution gamma spectrometry in order to re-estimate the annual effective gamma dose by converting data from Bq/kg in nGy/h according to **UNSCEAR 2008 Report**.

Outdoor gamma radiation measurements in Cluj County, Romania have been performed using solid state thermoluminescent detectors in order to develop a high resolution database for natural gamma dose rates. Integrated measurements have been carried out for an exposure time of minimum 3 weeks. According to EU requirements, the territory has been divided in 69 grids of 10X10 km. The cells were monitored using LiF:Mg,Cu,P detectors.

MATERIALS AND METHODS

In order to obtain a map of gamma dose rates for Cluj County, more than 240 detectors based on LiF:Mg, Cu, P (coded MCP-7 produced by TLD Poland) in the form of 4.5 mm diameter and 0.9 mm thickness pellets have been used. The dosemeters have been placed in 69 different locations from Cluj County between April and August 2013 (by dividing the territory in cells of 10 x 10 km as European Union requires) thus covering about 7000 km^2 (around 3% of Romania's territory). Cluj County relief is characterized by the presence of plateau and mountain areas. About a quarter of the territory surface is mountainous (Apuseni Mountains) with heights of up to 1800 m.

Before placing the dosemeters in the environment, a standard annealing procedure by heating the dosemeters at 240°C for 10 minutes was performed. A number of 3 up to 5 MCP-7 pellets for each location were packed in closed plastic bags and coded. Every detector was wrapped in aluminum foil in order to have humidity and light protection and placed at a distance of 1 meter above ground, according to standard procedures (**Olko et al., 2004**). The time exposure ranged from 24 to 51 days. After this period, TLDs were collected and brought into the laboratory and read with a Harshaw 3500 TLD Reader. The time temperature profile (TTP) for reading procedure was set by selecting four regions of interest in the glow curve (ROI 1: first 50 channels, ROI 2: channel 50 to channel 120, ROI 3: channel 120 to channel 155 and ROI 4 channel 155 to channel 200). The heating rate was 5°C/s with the maximum temperature of 220°C (for 20 seconds). Integration of the main peak at 210°C was made within the third region of interest. Calibration was performed using the ^{60}Co radioactive source from Physics Faculty, Babes-Bolyai University (described at the beginning of this chapter). Background subtraction was performed using the response of a batch of freshly annealed dosemeters.

High resolution gamma ray spectrometry analysis was carried out on a ORTEC hiperpure germanium detector having an active volume of 181 cm^3 and 34.2% relative efficiency at 1332.5 KeV. Relative calibration has been performed using IAEA 312 soil standard. The lines that were investigated for ^{232}Th series are: 238 KeV- ^{212}Pb, 338.3 keV- ^{228}Ac, 583.2 keV- ^{208}Tl and 911keV- ^{228}Ac. For ^{238}U the emissions at 295 KeV- ^{214}Pb, 352 KeV- ^{214}Pb and 609 KeV- ^{214}Bi have been used, while ^{40}K has been determined considering the gamma line at 1461 KeV. An example of a gamma spectrum obtained is depicted in **Figure 5.6**.

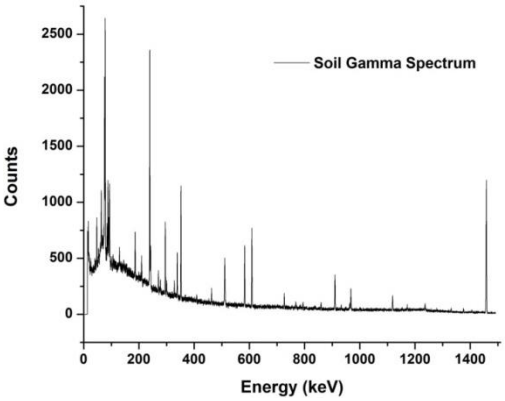

Figure 5.6. *Gamma spectrum obtained for a soil sample in Cluj County*

RESULTS AND DISCUSSION

The average background signal for 30 pellets has been determined. It was found to be equal to 0.32 nC. In the case of our dosimetric system, the lowest limit of detection can be estimated as a ratio between three times the standard deviation of background signal (nC) and the sensitivity of the dosemeters (μC/mGy):

$$LLD = \frac{3 \times stdev(bck.Sign)}{s} \quad (5.3)$$

where LLD is the lowest limit of detection and s is the sensitivity of our detectors.

A value of 247 nGy was obtained. As was already mentioned, this result is a very realistic one.

A typical glow curve for a detector exposed at Măguri-Răcătău is presented in **Figure 5.7**. The integral TL response in the third region of interest is 48 nC, being 2 orders of magnitude higher than the average background signal registered. The obtained result confirms that TL method using MCP-7 detectors is ultra-sensitive and can be applied for monitoring low dose rates for relative short periods. The sensitivity of our detectors was found to be equal to 0.363 μC/mGy.

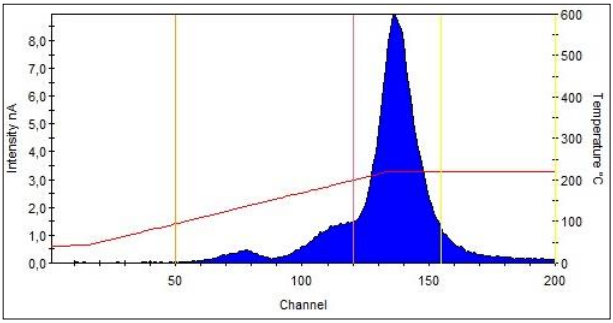

Figure 5.7. *Typical glow curve for Măguri-Răcătău location. The dosimetric peak is recorded in the 3rd region of interest at 210°C.*

Table 5.2. *TL signals, gamma dose rates and external annual effective gamma doses obtained in Cluj County (1)*

Location	Codes	Days of exp.	No. of dosim.	TL corr. signal /day (nC)	St. dev.TL corr. signal /day (nC)	Gamma dose rate (nGy/h)	External annual effective gamma dose (mSv)
Cluj-Napoca I	32	48	5	0.93	0.05	107±8	0.13±0.01
Apahida	33	48	4	1.01	0.01	117±6	0.14±0.01
Gheorgheni	44	48	5	0.75	0.02	87±4	0.11±0.01
Cluj-Napoca II	43	40	3	0.80	0.02	93±5	0.11±0.01
Baciu	31	48	2	0.91	0.04	106±7	0.13±0.01
Floreşti	42	42	3	0.97	0.02	112±6	0.14±0.01
Tureni	54	45	5	0.92	0.02	107±5	0.13±0.01
Turda	55	45	5	0.87	0.02	101±5	0.12±0.01
CâmpiaTurzii	65	45	5	0.91	0.02	106±6	0.13±0.01
Mihai-Viteazu	64	45	2	1.02	0.03	119±6	0.15±0.01
Cornești I	63	45	5	0.76	0.01	88±4	0.11±0.01
Iara	62	46	4	0.89	0.01	103±5	0.13±0.01
Băişoara	61	46	5	1.06	0.03	123±7	0.15±0.01
Lita	52	45	5	0.94	0.03	109±6	0.13±0.01
Gilău	41	36	5	0.97	0.02	113±6	0.14±0.01
Gârbău	30	36	5	0.86	0.02	99±5	0.12±0.01
Aghireşu	29	36	5	0.82	0.02	95±5	0.12±0.01
Păniceni	40	36	5	0.90	0.01	105±5	0.13±0.01
Bedeci	39	36	5	0.87	0.02	100±5	0.12±0.01
Huedin	28	36	5	0.88	0.01	101±5	0.12±0.01
Someşu-Rece	51	36	5	1.17	0.03	136±7	0.17±0.01
Măguri-Răcătău	60	36	5	1.29	0.02	150±7	0.18±0.01
Sălicea	53	32	5	0.98	0.02	114±6	0.14±0.01
Cojocna I	45	51	3	0.94	0.08	109±11	0.13±0.01
Cojocna II	34	51	3	0.70	0.05	81±7	0.10±0.01
Ceanu-Mare	46	51	3	0.85	0.06	99±8	0.12±0.01
Bolduţ	56	51	3	0.75	0.01	87±4	0.11±0.01
Clapa	57	51	3	0.82	0.07	95±10	0.12±0.01
Frata	47	51	3	0.93	0.07	108±10	0.13±0.01
Mociu	35	51	3	0.83	0.03	96±6	0.12±0.01
Cămăraşu	36	51	3	0.89	0.04	103±7	0.13±0.01
Buza	19	51	3	0.92	0.01	106±5	0.13±0.01
Geaca	25	51	3	0.67	0.08	77±10	0.09±0.01
Ţaga	18	51	3	0.56	0.03	64±4	0.08±0.01
Sânmarin	13	51	3	0.82	0.02	94±5	0.12±0.01
Fizeşu-Gherlei	12	51	3	0.54	0.05	62±7	0.08±0.01
Bonţ	17	51	3	0.71	0.02	82±5	0.10±0.01
Sic	24	51	3	0.72	0.05	84±7	0.10±0.01
Răscruci	23	51	3	0.72	0.03	83±5	0.10±0.01
Iclod	16	51	3	0.61	0.02	71±4	0.09±0.01
Mihăieşti	20	50	3	0.65	0.04	75±6	0.09±0.01
Aşchileu-Mare	14	50	3	0.68	0.04	79±6	0.10±0.01
Vultureni	21	50	3	0.66	0.03	77±5	0.09±0.01
Bobâlna	5	50	3	0.61	0.01	71±3	0.09±0.01
Pruni	4	50	3	0.70	0.02	81±5	0.10±0.01
Câţcău	2	50	3	0.81	0.03	93±5	0.11±0.01

Table 5.2. *TL signals, gamma dose rates and external annual effective gamma doses obtained in Cluj County (2)*

Location	Codes	Days of exp.	No. of dosim.	TL corr. signal /day (nC)	St. dev.TL corr. signal /day (nC)	Gamma dose rate (nGy/h)	External annual effective gamma dose (mSv)
Rugăseşti	3	50	3	0.74	0.02	85±5	0.10±0.01
Chiuieşti	1	50	3	0.64	0.03	74±5	0.09±0.01
Dej	6	50	3	0.76	0.03	88±5	0.10±0.01
Mica	7	50	3	0.79	0.03	92±5	0.11±0.01
Unguraş	8	50	3	0.57	0.03	66±5	0.08±0.01
Nima	11	50	3	0.68	0.08	79±10	0.10±0.01
Cornești II	10	50	3	0.70	0.02	81±4	0.10±0.01
Lujerdiu	15	50	3	0.72	0.03	84±5	0.10±0.01
Borşa	22	50	3	0.65	0.05	76±6	0.09±0.01
Recea-Cristur	9	50	3	0.65	0.02	75±4	0.09±0.01
Negreni	26	24	3	0.57	0.02	66±4	0.08±0.01
V. Drăganului	27	24	3	0.74	0.07	85±9	0.10±0.01
Săcuieu I	37	24	3	0.57	0.01	66±3	0.08±0.01
Săcuieu II	38	24	3	0.61	0.005	71±3	0.09±0.01
Margău	48	24	3	0.54	0.03	62±5	0.08±0.01
Călăţele	49	24	3	0.77	0.01	89±4	0.11±0.01
Beliş	59	24	3	0.64	0.02	74±4	0.09±0.01
Poiana-Horea I	58	24	3	0.67	0.02	78±4	0.10±0.01
Poiana-Horea II	66	24	3	0.68	0.07	79±9	0.10±0.01
Mărişel	50	24	3	1.00	0.03	116±6	0.14±0.01
Lungeşti	67	24	3	0.90	0.05	104±8	0.13±0.01
Moldoveneşti	68	24	3	0.48	0.03	56±4	0.07±0.01
Bădeni	69	24	3	0.57	0.03	66±5	0.08±0.01

Table 5.2 (1),(2) presents the results obtained for the 69 locations considered. The gamma dose (μGy) was obtained as a ratio between the signal light (nC) and the sensitivity of our detectors (μC/mGy) (previously calculated throughout calibration procedure).

$$D = \frac{S}{s} \tag{5.4}$$

where D is the gamma dose, S is the signal light obtained in TL method and s is the sensitivity of MCP-7 detectors obtained.

Gamma dose rate was obtained with:

$$Dr = \frac{D}{t} \tag{5.5}$$

where Dr is the gamma dose rate, D is the gamma dose and t is time.

The effective external gamma dose (mSv) was calculated using a conversion factor of 0.7 (see UNSCEAR Report 2008, Annex B, Section I) in order to convert the absorbed dose in air in external effective annual gamma dose in adults. A factor of 0.2 reflecting the outdoor component of the annual effective dose was also used.

$$Eff = 0.7 \times Dr \times 0.2 \quad (5.6)$$

where Eff is the effective external gamma dose and Dr is the gamma dose rate.

The mean gamma dose rate was found to be equal to 91 nGy/h which is very close to the Romanian average stated by **UNSCEAR 2008 Report** (92 nGy/h). Also, it can be noted that the dose rates were in the normal range of variation given by **UNSCEAR 2008** from the total outdoor range gamma dose rate (see column no. 8 from Table 6, Annex B) which is 52-163 nGy/h. The higher dose rate (150±7 nGy/h) was observed in Măguri-Răcătău village. This result is given by the existent geological substrate in this area, a granitic one that increase the existing natural background.

In order to compare the results obtained by TLD method, we have carried out estimations of the external effective annual outdoor gamma dose using the specific activities of five soil samples (the samples were taken from the same location where TLD were placed) determined by high resolution gamma spectrometry using the conversion factors stated by Table 1 Annex B of **UNSCEAR 2008 Report**.

$$Eff = A^{226}_{Ra} * DCF^{226}_{Ra} + A^{40}_{K} * DCF^{40}_{K} + A^{232}_{Th} * DCF^{232}_{Th} \quad (5.7)$$

where Eff is the external effective annual gamma dose, A is the activity of radionuclides and DCF is the conversion factor for natural radionuclides.

Cosmic dose rates have been computed for the investigated locations using the conversion factors given by **Prescott and Hutton, 1994**. The obtained results are given in **Table 5.3.** It can be noted that the results obtained by gamma spectrometry are consistent within error with the results obtained by TLD method considering a 95% confidence level.

Table 5.3. *Gamma spectrometric information for soil samples collected from five locations. Annual external effective outdoor gamma doses calculated based on the specific activities are compared to the values obtained by TLD.*

Location	Specific activity (Bq/kg)			High above sea level (m)	Cosmic Dose rate (mGy /year)	External annual effective gamma dose (mSv) Gamma Spec	External annual effective gamma dose (mSv) TLD
	^{238}U	^{232}Th	^{40}K				
Somesu-Rece	44.7±1.6	43.7±2.5	729±18	592	0.324±0.005	0.14±0.01	0.17±0.01
Bobalna	32.3±1.1	32.2±1.8	552±13	305	0.307±0.005	0.11±0.01	0.09±0.01
Cluj-Napoca	31.3±1.1	31.4±1.9	669±16	351	0.310±0.005	0.11±0.01	0.11±0.01
Bedeci	49.7±1.7	35.5±1.5	715±12	637	0.327±0.005	0.13±0.01	0.12±0.01
V. Dragan	22.5±0.8	46±2.6	705±18	574	0.323±0.005	0.13±0.01	0.10±0.01

A high resolution map of Cluj County, Romania for gamma dose rates is presented (see **Figure 5.8 and 5.9**).

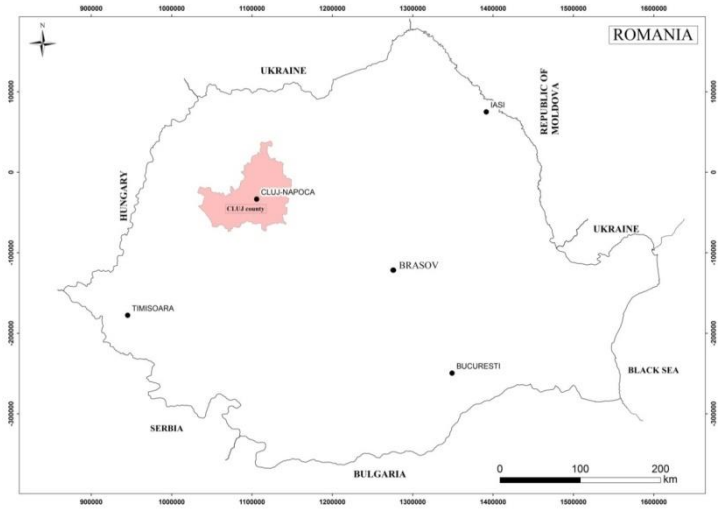

Figure 5.8. *Map of Romania with the localization of Cluj County*

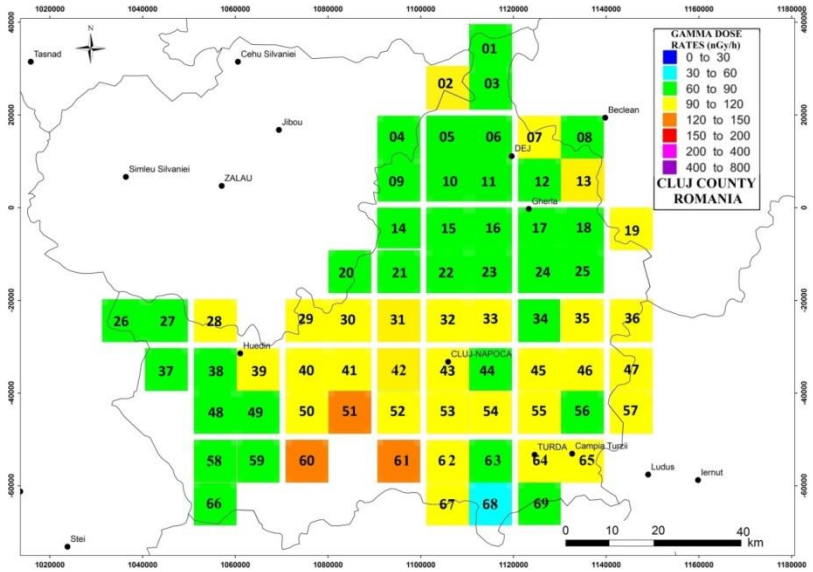

Figure 5.9. *The high resolution map of gamma dose rates for Cluj County, Romania*

The map was developed according to Romania's obligations as an EU Member State (by dividing the territory in cells of 10X10 km) and also with regard to the environmental policy stated in the EC Treaty, Art. 174. Present data will be used in order to settle a correlation between the gamma dose rates obtained and radon concentrations measured within RAMARO research project (Radon map – residential, geogenic, water – for Center, West and Northwest regions from Romania"), (**RAMARO - http://radon.com.ro**).

CONCLUSIONS

Ultra sensitive MCP-7 (LiF:Mg,Cu,P) TL detectors were used in order to assess the gamma doses for Cluj County, Romania with the ultimate goal to develop a high resolution map of gamma dose rates. Results were in perfect accordance with UNSCEAR 2008 Report regarding gamma dose rates for Romania. A high resolution map of Cluj County for gamma dose rates is presented according to the

European Environmental Policies. This work is an important step in developing high resolution maps of gamma dose rates at national level.

As an outlook to future work on the subject, it is planned to expand the investigated area of interest, considering more counties, in order to improve the existent databases and create new high resolution maps of gamma dose rates for Romania's territory. Furthermore, the results of the present study will represent a valuable database in order to establish a correlation between gamma dose rates and radon concentration within „Radon map (residential, geogenic, water) for Center, West and Northwest regions from Romania" research project.

5.3. Measurements of terrestrial gamma dose rates and radon concentrations from indoor air and water in Transylvania region.

INTRODUCTION

Previous studies (**Dolha-Zeciu, 2013; Dolha, 2014**) have demonstrated that TL 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 of a few μGy.

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 (**Dictate no.14, 2000**). Consequently, the present study brings an important contribution to the natural radioactivity database in Transylvania region.

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 (1μGy-10 Gy) (**Olko et al., 2004; Bilski et al., 2008**). 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 km^2. 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 (**Olko et al., 2010**). 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 (**Dolha-Zeciu et al., 2013**).

Radon measurements in air and water

In the case of radon concentration measurements, CR-39 track etched detectors were used. In the case of Cluj County, about 73 detectors were placed in 19 grids and a number of 46 water samples were collected between April-August 2013. In Alba County, the detectors were placed in 12 locations (43 dwellings) and 27 water samples were considered. The period of exposure was 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 (**Cucos-Dinu et al., 2012**).

In the case of water measurements, the samples were taken from wells in bottles of 0.5 l and totally filled. ^{222}Rn concentration was measured using the LUK-VR system consisting of a LUK-3A device and a VR-scrubber (**Plch, 1977, Cosma et al., 2008b**). The radon concentration dissolved in the water sample 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.,2008b**).

RESULTS AND DISCUSSIONS

The results obtained for the 52 considered locations are given in **Table 5.4.** The mean gamma dose rate was found to be 62 nGy/h which is a normal value according to **UNSCEAR 2008 Report**. 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 -see column no. 8 from Table 6, Annex B). The higher dose rate (91±4 nGy/h) was observed in Petresti village.

Table 5.4. *Gamma dose rates and external annual effective gamma doses*

Location	Lab. Codes	Days of exp.	No. of dosim.	TL corr. signal/day (nC)	St. err. TL signal (nC)	Gamma dose rate (nGy/h)	Ext. ann. eff. gamma dose (mSv)
Lancram	1	125	3	0.47	0.01	55±3	0.07±0.01
Petresti	2	125	3	0.79	0.01	91±4	0.11±0.01
Rachita	3	125	3	0.68	0.05	78±6	0.09±0.01
Sasciori	4	125	3	0.49	0.01	56±3	0.07±0.01
Capalna	5	125	3	0.66	0.05	76±6	0.09±0.01
Daia-Romana	6	125	3	0.59	0.04	68±6	0.08±0.01
Spring	7	125	3	0.49	0.02	56±4	0.07±0.01
Rosia de Secas	8	125	3	0.65	0.01	76±4	0.09±0.01
Ohaba	9	125	3	0.58	0.01	67±3	0.08±0.01
Seusa	10	125	3	0.49	0.01	57±3	0.07±0.01
Blaj	11	125	3	0.40	0.05	46±2	0.06±0.01
Tiur	12	125	3	0.40	0.02	46±3	0.05±0.01
ValeaLunga	13	125	3	0.51	0.01	59±3	0.07±0.01
Bucium	14	125	3	0.44	0.01	51±3	0.06±0.01
VamaSeaca	15	118	3	0.49	0.06	57±7	0.07±0.02
Uioara de Sus	16	21	3	0.44	0.03	51±4	0.06±0.02
Farau	17	21	3	0.41	0.01	47±2	0.06±0.01
Silea	18	21	3	0.51	0.01	59±3	0.07±0.01
LopadeaNoua	19	21	3	0.41	0.01	48±3	0.06±0.01
Sancel	20	21	3	0.50	0.03	58±5	0.07±0.02
Jidvei	21	21	3	0.36	0.01	41±2	0.05±0.01
Sard	22	21	3	0.43	0.02	50±4	0.06±0.01
Metes	23	21	3	0.55	0.01	64±3	0.08±0.01
Patrangeni	24	21	3	0.55	0.02	63±4	0.08±0.01
Zlatna	25	21	3	0.48	0.01	55±3	0.07±0.01
Izv. Ampoiului	26	21	3	0.54	0.06	62±8	0.07±0.01
Nadastia	27	21	3	0.52	0.07	61±9	0.07±0.01
Vinerea	28	21	3	0.65	0.03	75±6	0.09±0.02
Sibot	29	21	3	0.68	0.02	79±5	0.09±0.02
Mereteu	30	21	3	0.75	0.04	87±7	0.10±0.02
Rimetea	31	21	3	0.46	0.05	53±7	0.06±0.02
Posaga de Jos 1	32	21	3	0.56	0.03	64±5	0.08±0.02
Posaga de Jos 2	33	21	3	0.44	0.01	51±3	0.06±0.01
Brazesti	34	21	3	0.63	0.02	72±4	0.09±0.02
Baia de Aries	35	21	3	0.53	0.03	62±5	0.07±0.02
Lupsa	36	21	3	0.72	0.05	84±7	0.10±0.02
Bistra	37	21	3	0.48	0.02	56±3	0.07±0.01
Garde	38	21	3	0.39	0.02	46±3	0.05±0.01
VaduMotilor	39	21	3	0.49	0.01	56±3	0.07±0.01
Vidra	40	21	3	0.54	0.01	63±4	0.07±0.01
Albac	41	21	3	0.63	0.03	73±5	0.09±0.02
Garda de Sus	42	21	3	0.35	0.02	41±3	0.05±0.01
Arieseni	43	21	3	0.67	0.02	78±4	0.09±0.02
Abrud	44	21	3	0.57	0.03	65±5	0.08±0.02
Mogos	45	21	3	0.57	0.04	66±6	0.08±0.02
Aiud	46	21	3	0.59	0.03	68±5	0.08±0.02
Magina	47	21	3	0.57	0.02	65±4	0.08±0.02
Tifra	48	21	3	0.52	0.01	60±3	0.07±0.01
Geoagiu de Sus	49	21	3	0.55	0.01	63±3	0.08±0.01
Unirea	50	21	3	0.62	0.04	72±6	0.09±0.02
Stremt	51	21	3	0.56	0.01	65±4	0.08±0.01
Teius	52	21	3	0.56	0.01	65±4	0.08±0.01

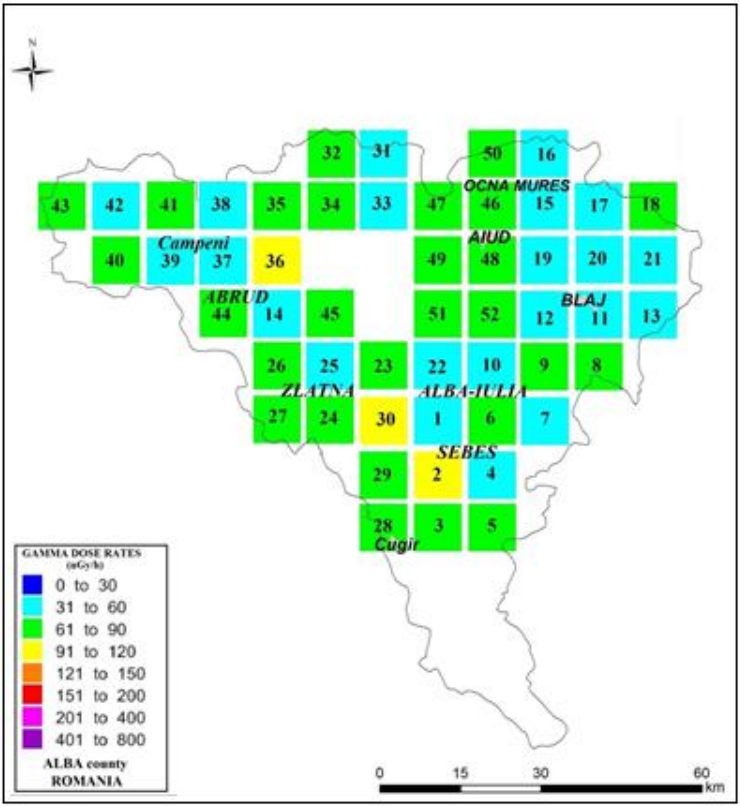

Figure 5.10. *The high resolution map of gamma dose rates [nGy/h] for Alba County, Romania (the codes within every cell are given in* ***Table 5.4****)*

A high-resolution map of Alba County, Romania for gamma dose rates is presented by dividing the territory into cells of 10X10 km (**Figure 5.10**).

Estimations of the external effective annual outdoor gamma dose using the specific activities obtained within high resolution gamma spectrometry were also carried out in order to compare the doses obtained by TLD method. Were used the conversion factors stated by Table 1 Annex B of **UNSCEAR 2008 Report**.

$$Eff = A^{226}_{Ra} * DCF^{226}_{Ra} + A^{40}_{K} * DCF^{40}_{K} + A^{232}_{Th} * DCF^{232}_{Th} \quad (5.8)$$

where Eff is the external effective annual gamma dose, A is the activity of radionuclides and DCF is the conversion factor for natural radionuclides.

Cosmic dose rates have been computed using the conversion factors given by Prescott and Hutton. The obtained results are given in **Table 5.5**. It can be noted that the results obtained by gamma spectrometry are consistent within error with the results obtained by TLD method considering a 95% confidence level.

Table 5.5. *Gamma spectrometric information for soil samples collected from eight locations from Alba County. Annual external effective outdoor gamma doses calculated based on the specific activities are compared to the values obtained by TL method.*

Location	Specific activity (Bq/kg)			High above sea level (m)	Cosmic Dose rate (nGy/h)	External annual effective gamma dose (mSv) Gamma Spec.	External annual effective gamma dose (mSv) TLD
	^{238}U	^{232}Th	^{40}K				
Abrud	35.2±0.3	47.7±0.5	855.0±1.2	600	37±6	0.08±0.01	0.10±0.01
Capalna	27.3±0.2	41.5±0.3	995.4±7.7	365	35±5	0.09±0.01	0.10±0.01
D. Romana	22.2±0.7	27.8±2.3	556.0±6.4	342	35±5	0.08±0.01	0.06±0.01
Vidra	24.0±1.4	29.7±0.7	464.7±6.9	626	37±6	0.08±0.01	0.06±0.01
Lancram	27.8±0.1	37.4±0.3	705.0±6.4	238	35±5	0.07±0.01	0.08±0.01
Petresti	32.7±0.2	38.9±0.4	789.4±7.5	278	35±5	0.11±0.01	0.09±0.01
Sasciori	26.2±1.6	38.2±0.3	757.2±8.3	673	38±6	0.07±0.01	0.08±0.01
Spring	18.3±1.1	26.4±1.8	593.3±6.9	313	35±5	0.07±0.01	0.06±0.01

Regarding radon measurements, for Alba County were monitored 43 dwellings and 27 water samples. In the case of Cluj County, 73 dwellings were considered while a number of 46 water samples were analysed. The obtained results are presented within **Table 5.6** and **Table 5.7.**

Table 5.6. *Radon concentrations for indoor air and water samples in Alba County*

Locations	Indoor radon conc. (Bq/m^3)			Radon conc. in water (Bq/l)		
	A.M.*	S.D.*	Max.	A.M.	S.D.	Max.
Lancram	94.3	22.4	118	3.9	3.2	7.6
Rachita	44.7	22.1	70	13.7	9.6	24.4
Sasciori	40	-	40	10.3	0.6	10.8
Capalna	255	153.8	476	-	-	10.5
DaiaRomana	95.5	79.7	208	7.8	4.1	11.6
Spring	65.3	41.2	113	3.1	1.3	4.4
Rosia de Secas	91.5	47	133	6.7	0.5	7.1
Ohaba	89.7	43.4	127	7.2	4.5	10.2
Seusa	83.8	18.5	107	-	-	2.6
ValeaLunga	90.7	42.4	162	3.4	13	4.3
Goiesti	36.3	20.6	58	-	-	3.4
Abrud	50.7	47.3	104	-	-	2

Table 5.7. *Radon concentrations for indoor air and water samples in Cluj County*

Locations	Indoor radon conc. (Bq/m^3)			Radon conc. in water (Bq/l)		
	A.M.*	S.D.*	Max.	A.M.	S.D.	Max.
Gadalin	489	407	936	1.9	1.2	3.2
Orman	240	165	456	2	1.3	3.3
Capalna	264	169	485	1.2	0.7	1.9
Livada	183	77	266	3.6	0.6	4
Bobalna	249	109	385	2.1	1.4	3.1
Pruni	582	208	705	3	0.6	3
Chiuiesti	168	66	212	7.5	7.4	3.9
V.Draganului	275	286	758	21.8	16.2	39.4
Negreni	170	117	313	1.2	0.1	1.3
Tranis	362	311	689	8.8	3.6	6.2
Bedeci	303	96	457	6.3	5.1	11.3
Paniceni	247	168	456	4.2	5.4	8
Salicea	114	64	182	10.9	10.3	17
Deleni	529	725	1606	15.4	9.8	22.3
Filea de Jos	194	158	356	8.5	6.4	13
Baisoara	105	43	144	2.2	1.6	3.9
Hasdate	140	103	287	17.6	17.5	35.7
Belis	97	60	158	45.9	21.6	61.1
Marisel	342	283	613	178.7	94.9	352.2

Table 5.8 *Water and Indoor Rn concentrations vs gamma dose rates obtained by TL method for Cluj and Alba Counties (* ***Cosma et al., 2015****)*

County	Average soil Rn conc. (kBq/m^3)	Average water Rn conc. (Bq/l)	Average indoor Rn conc. (Bq/m^3) A.M.	Average gamma dose rate (nGy/h) by TL A.M.
Alba	29.1±18.5	9.7±0.9	115±160	62±12
Cluj	37.9±21.3	20.6±3.9	140±154	91±18

According to **EURATOM Treaty 59/2013**, all European countries must develop action plans regarding radon exposure starting with 2018. Many European countries have already established remedial action levels for indoor radon as well as for water. 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 400 Bq/m^3 is given. As can be observed from **Table 5.6** regarding Alba County, the higher value has been measured in Capalna village (476 Bq/m^3) that is above the remedial action level mentioned before. For water samples, all measurements were under the limit (100 Bq/l) given by **WHO**. In the case of Cluj County, in 11 dwellings the indoor radon concentration exceeded the limit of 400 Bq/m^3 (**Table 5.7**). The higher concentration was observed in Deleni village (1606 Bq/m^3). Regarding water measurements, the concentrations obtained were clearly higher compared with the results observed in Alba County. The highest concentration was registred in Marisel village (352.2 Bq/l). The 11 results that exceded 400 Bq/m^3 can be correlated with the geological context (granitic substrate - Marisel village), poor ventilation of the rooms (the detectors were placed from September 2013 until March 2014) or the possibility that the dwellings/rooms were not used during the monitoring period. According to **Table 5.8** it can be observed that the results obtained for Alba County were always smaller than those obtained for Cluj County both for Radon concentration (considering for Alba County a number of 168 soil measurements, 112 water measurements and 412 indoor measurements and for Cluj County a number of 71 soil measurements,166 water measurements and 544 indoor measurements) (**Cosma et al., 2015**) as well as for gamma dose rates (156

measurements for Alba County and 241 measurements for Cluj County). This results can be explained by the differences on the geological settings from the two Counties. In the case of Cluj County, it can be also observed that Someșu-Mic river which begins from Măguri-Răcătău area (which presents a granitic context) influences the ambiental gamma dose rate in his flow by a continuous decreasing of the gamma dose rates as can be observed in **Figure 5.9** (**Cosma et al., 2016**).

We have also carried out estimations of the annual effective doses for ingestion and inhalation of radon using the conversion factors given by **UNSCEAR 2008 Report** in order to assess the effective exposure of humans to radon (**Table 5.9** and **Table 5.10):**

$$D_{inh}=C_{Rn}\,(\varepsilon_r+\varepsilon_d f)\,O, \quad (5.9)$$

where D_{inh} is annual effective dose for inhalation; C_{Rn}represent the mean annual radon activity concentration (Bq/m^3); $\varepsilon_r$and ε_d are dose conversion factors for radon gas and its short-lived progeny respectively, ε_r=0.17(nSv/h)/(Bq/m^3) and ε_d=9(nSv/h)/(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 h/y.

$$D_{ing}=C_{Rn}\times F_{Rn}\times C_w, \quad (5.10)$$

where D_{ing} in 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.

Table 5.9. *Annual effective doses due to radon inhalation and ingestion in Alba County*

Location	Ann. eff. dose (inhalation) (mSv)	Ann. eff. dose (ingestion) (mSv)
Lancram	2.49	0.01
Rachita	1.18	0.05
Sasciori	1.06	0.04
Capalna	6.73	0.04
DaiaRomana	2.52	0.03
Spring	1.72	0.01
Rosia de Secas	2.41	0.02
Ohaba	2.37	0.03
Seusa	2.21	0.01
ValeaLunga	2.39	0.01
Goiesti	0.96	0.01
Abrud	1.34	0.01

Table 5.10. *Annual effective doses due to radon inhalation and ingestion in Cluj County*

Location	Ann. eff. dose (inhalation) (mSv)	Ann. eff. dose (ingestion) (mSv)
Gadalin	12.9	0.01
Orman	6.3	0.01
Capalna	7	0.01
Livada	4.8	0.01
Bobalna	6.6	0.01
Pruni	15.4	0.01
Chiuiesti	4.4	0.03
V.Draganului	7.2	0.08
Negreni	4.5	0.01
Tranis	9.6	0.03
Bedeci	8	0.02
Paniceni	6.5	0.02
Salicea	3	0.04
Deleni	13.9	0.06
Filea de Jos	5.1	0.03
Baisoara	2.8	0.01
Hasdate	3.7	0.06
Belis	2.6	0.17
Marisel	9	0.65

Taking into account the reference level within the range of 3 mSv/year to 10 mSv/year proposed by **ICRP 1993** it can be observed that for Alba County (**Table 5.9**) 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. 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** which gives a dose limit of 0.1 mSv/year. In the case of Cluj County (**Table 5.10**), the doses vary from 2.6 mSv to15.4 mSv with a standard deviation of 4.9 mSv. For three grids the annual effective doses due to radon inhalation exceeded the limit of 10 mSv/year (Gadalin, Pruni and Deleni). The higher dose observed in Deleni village was estimated at 15.4 mSv/year. As was already mentioned, the most credible reason is the poor ventilation of the dwelling considered. With respect to annual effective dose due to ingestion, almost all results were found to be normal except two locations (Belis and Marisel) were the doses exceeded the limit given by **ICRP 1993**. (0.17 mSv/year and 0.65 mSv/year respectively having a standard deviation of 0.14 mSv). This can be explained by the geological context existent within the area considered (a granitic one, with rocks rich in uranium).

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 43 dwellings from Alba County and 73 dwellings for Cluj County as well. For 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**. It was also observed that all results including radon concentrations for soil, water and indoor air as well as for gamma dose rates were clearly higher in Cluj County compared with the results obtained for Alba County. These discrepancies are explained by the differences between the geological substrates existent within the two Counties. 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**. Regarding Cluj County, in 11 dwellings the indoor radon concentrations exceeded the limit given by

WHO and **EURATOM Treaty** and was explained either by the geological setting existent or by the poor ventilation within the dwellings investigated. Also, the annual effective doses due to radon inhalation/ingestion exceeded in some cases the limit given by **ICRP 1993 Report.**

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.

6. FINAL CONCLUSIONS

Applications of thermoluminescence dosimetry in environmental monitoring over an extended area (about 13 000 km^2) have been carried out for the first time in Romania. This was achieved following the implementation of a dosimetric system based on LiF:Mg,Cu,P detectors and a Harshaw 3500 TLD reader. The method was applied for measuring the gamma dose rates in Transylvania region. High resolution maps of gamma dose rates are presented for the first time for Cluj and Alba County.

In an initial study the existence of a correlation between environmental gamma dose rates and geological substrate was shown, based on the investigations of seven different locations in Romania. The results obtained showed a clear dependence between gamma dose rates and geological setting. Dose rates of 158±8 nGy/h have been measured in Belis village, a location characterized by a granitic substrate, a value which is about 70% higher than the average value obtain on the rest of the investigated locations. These results strengthen the correlation between gamma dose rate and geological substrate.

A high resolution map of gamma dose rates in Cluj and Alba Counties was first developed on the reference grid 10 x 10 km as recommended by Joint Research Centre of the European Commission.

More than 240 detectors based on LiF:Mg,Cu,P have been used for this purpose in Cluj County covering about 7000 km^2. The gamma dose rates obtained were in the normal range of variation according to **UNSCEAR Report from 2008** (52-163 nGy/h). Additionally, external effective annual outdoor gamma dose have been estimated. The accuracy of the values obtained using the TLD method have been further confirmed by calculating the effective annual doses bases on factors stated by **UNSCEAR 2008 Report** and the specific activities of U, Th and K measured through high resolution gamma spectrometry. In addition, radon concentrations for indoor air (73 dwellings) and water (43 samples) have measured and associated effective doses due to radon inhalation and ingestion have been computed.

The same investigations as in Cluj have been carried out over Alba County. In order to estimate the gamma dose rates, more than 150 dosimeters based on LiF:Mg,Cu,P, have been placed, covering about 5200 km^2. The results obtained were in the normal range of variation with **UNSCEAR Report from 2008**. High resolution gamma spectrometry was also applied for eight soil samples in order to estimate the external effective annual outdoor gamma dose. The results obtained were compared with the doses obtained by thermoluminescence method and were found to be consistent within errors considering a 95% confidence interval. For radon monitoring 43 dwellings were considered along with 27 water samples.

For Cluj County, in 11 dwellings the indoor radon concentration exceeded the limit of 400 Bq/m^3. The higher concentration was observed in Deleni village (1606 Bq/m^3). The higher concentration due to radon ingestion was registered in Marisel village (352.2 Bq/l) that was under the limit of 1000 Bq/l stated by WHO. The results can be correlated with geological context (granitic substrate - Marisel village) and/or poor ventilation of the rooms (the detectors were placed in from September 2013 until March 2014). Regarding Alba County, the higher value has been measured in Capalna village (476 Bq/m^3) that is above the remedial action level. For water samples, all measurements were under the limit given by **WHO**.

The obtain results clearly demonstrate the potential of themoluminescence dosimetry in environmental radioactivity monitoring. Continuing these investigations for the whole region of Transylvania is desirable in order for a realistic database for this region to be developed. Such surveys are extremely important since are an obligation of our country as a European Union member state.

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