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DETERMINATION OF RADIUM IN MINE WATER FROM THE NORTH OF TRANSYLVANIA, ROMANIA

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Abstract. Groundwater, once surfaced, may contain some radioactive elements that can increase the natural radioactivity levels of different environmental factors, having a varied impact on the ionizing radiation dose received by the human body. This paper presents radium in groundwater concentration measurements from various galleries of disused mines. The studied area is located in Gutâi, Rodna and Maramures Mountains, in the northern region of Transylvania, Romania. In order to determine the radium concentration in the collected water samples, two methods were used, both based on radium and its decay product, radon, secular equilibrium. After achieving the radioactive balance, the radon was measured using two types of scintillation Lucas cells. The obtained values of radium concentration range from 20 to 510 mBq/l for the 1 liter Lucas cell method and from 50 to 501mBq/l for the 145 ml Lucas cell method. The highest radium concentration values were found for samples from Nistru Gallery (510mBq/1, respectively, 501mBq/l) and Herja Gallery - Baia Mare (410 mBq/l, respectively, 463 mBq/ l). The lowest values radium was determined for samples from Boului Gallery - Baia Sprie (20 mBq/l and 50 mBq/l) and Băiuţ Gallery (45 mBq/l, respectively, 50 mBq/l). The average obtained radium concentrations values are of 138 mBq/l for 1l Lucas cell method and 171 mBq/l for 145 ml Lucas cell method.

Keywords: mine water, radium, radon, secular equilibrium, Lucas cell

1. INTRODUCTION

Mining activities, regardless of raw material extracted from the earth's crust (iron ore, coal, nonferrous ore, uranium, etc.) result in bringing to the surface large quantities of sterile. Besides raw material, both can have a strong effect on environment and a high probability to produce environmental damage. Although mining activity itself is closed, different materials can still reach the earth's surface, mine water in particular. In addition to various substances that reach the surface, under different chemical and physical forms, substances with radioactive containing can cause an increase in the natural radioactivity concentration of different environmental factors, with respect to the natural received radiation dose by the human body. (UNSCEAR, 2000).

Among the natural radioactive elements, uranium and thorium have an important role, especially their decay products, such as ^{226}Ra, ^{228}Ra, ^{222}Rn, ^{210}Pb and ^{210}Po, most of them being alpha emitting nuclides (Lehto & Hou, 2011).

^{226}Ra plays an important role in internal human body contamination, if it is ingested, due to: the alpha particle emission, its long half-life of 1602 years and also because its shot lived daughters. Once inside the body, from food and water, ^{226}Ra tends to accumulate in bones. Over 70% of radium from human body is contained in bones and teeth, the remaining fraction being distributed fairly evenly in soft tissues (Lloyd, 1961). Radium metabolic behavior inside the human body is similar to calcium. At the same time, ^{226}Ra act as parent for ^{222}Rn, which is a radioactive gas easily inhaled by humans, being the second leading cause, after smoking, for lung cancer occurrence (Baias et al., 2010; Cosma et al., 1997, Cosma et al., 2009).

The current paper presents ^{226}Ra concentration measurements in mine water samples, from different closed mine galleries.

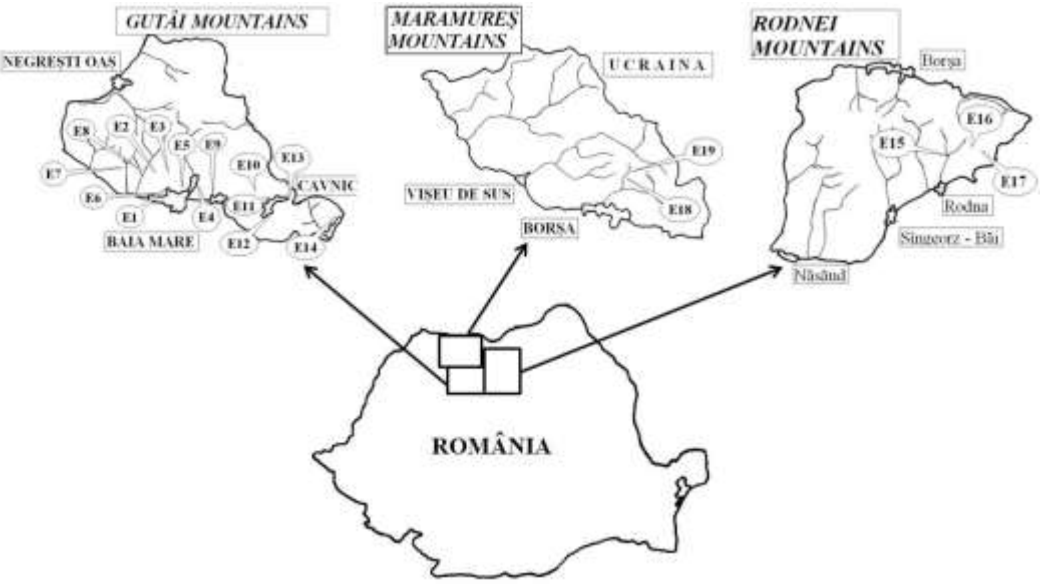

Figure 1. Study area – Gutai, Rodna and Maramures Mountains - and locations of groundwater sampling, E1 - E19.

2. AREA OF STUDY AND GEOLOGIC DATA

A mountainous region, from northern Romania was chosen as the study area. In this area, over several centuries, intense mining activities took place. From Gutâi Mountains, Rodna and Maramures Mountains, polymetallic ores with high content of lead, zinc, silver, gold, copper, antimony were exploited.

Baia Mare zone is the northwestern part of the Neogene volcanic chain median segment Vihorlat - Gutâi - Călimani - Gurghiu - Harghita inside the East Carpathian arc. Neogene magmatism in the Baia Mare zone has manifested itself by extensive volcanism with explosive character, effusive and intrusive. Over the pre-Neogene basement composed by crystalline rocks belonging to Median Dacide and sedimentary formations, Cretaceous-Paleocene of Transcarpathian flysch (Săndulescu 1984), were deposited the Neogene sedimentary formations divided on paleontological and stratigraphic criteria belonging to the Badenian – Sarmatian – Pannonian interval.

Pre-Neogene basement was deeply dislocated by latitudinal displacement of fractures (E-W), which controlled the development of volcanism and metallogenesis (Borcoş et al., 1994). These E-W type fractures had an important role in generating alignments tectonic-magmatic-metallogenetic, (Popescu 1986).

The magmatic activity has generated predominantly effusive volcanic sequence, subordinated explosive being accompanied by an intrusive magmatic phase. The volcanic activity took place in several stages from Badenian to Pontian - Pliocene (after Giuşcă et al., 1973, and Borcoş et al., 1973). After recent data regarding the age of the volcanic formations based on K-Ar dating (Edelstein et al., 1992), the volcanic products from the Gutâi Mountains were generated in a short period, from Badenian to Pannonian-Pontian. Magmatic sequences alternating with the sedimentary ones, investigated by different authors, contributed to the delimitation of volcanic phases during certain periods of time and to those of sedimentation. The predominant chemical nature of the magmatic activity is calc-alkaline.

The Volcanic activity from Gutâi Mountains started with an explosive acid phase, of Badenian age (first cycle of eruption, Giuşcă et al., 1973, Borcoş et al., 1973) whose products are represented by pyroclastic of fall and subordinated by welded tuffs indicating ignimbrite volcanic events (pyroclastic flow). The second eruption cycle corresponds to the andesite-dacite volcanism with products predominantly intermediate andesitic, between Sarmatian-Pontian, represented by three phases:

- Sarmatian Pyroxene andesites of Seini, with development in the South-Western part of the Gutâi Mountains;

- Pannonian quartziferous andesites of Piscuiatu, of Colbu, of Şuior and Highişa with development in the Northern part of Baia Mare and in the South-East of Cavnic. There are included in this cycle, on small areas dacite of Limpedea, of Şindileu, of Ulmoasa, and dacite of Dăneşti.

- to the Lower Pontian at the end of cycle, the pyroxene and amphibole andesites of Firiza, of

Jereapăn and Breze type, are assigned. These rocks outcrop from Firiza Valley to the east.

To the third eruption cycle corresponds the andesitic volcanic products being attributed to the Pliocene and are represented by pyroxene or biotite andesites.

In the Nistru zone Damian et al., (2009) described the igneous rocks with subvolcanic character, distinct by various morphological aspects: dykes, apophyses, irregular bodies appear on the surface in natural openings being in relationship with lavas and pyroclastic products.

In the sampling area Badenian pyroclastic rocks outcrop, as well as pyroxene adesites of Seine, quartziferous andesites of Piscuiatu, pyroxene andesites with amphibole of Jereapăn type. The andesites consist of minerals predominantly leucocrate (light) represented by feldspar, quartz, and subordinated by the melanocratic ones (dark) represented by pyroxene (augite and hyperstene), amphibole (green hornblende) and biotite, (Borcoș et al., 1973). Mostly the igneous rocks and some sedimentary rocks in the volcanic area are affected by postmagmatic volcanic changes (hydrothermal), namely: propylitization, sericitization, potassic alteration (adularia) silicification and argillization, (Stanciu 1972, Stanciu 1973). The sericitization and potassic alteration are characterized by the replacement of the primary minerals with secondary biotite, K-feldspar and with sericite, (Damian 2003). These secondary minerals introduce a large amount of potassium in rocks.

The metalogenetic activity would correspond to three distinct phases, Sarmatian-Pontian (Giuşcă et al., 1973) and occupies the southern border of the Gutâi Mountains. The first phase of copper, polymetallic and gold mineralization overlaps the Sarmatian pyroxene andesites in Ilba-Nistru sector. The second phase corresponds to the gold mineralization Săsar – Valea Roșie – Dealul Crucii associated with Pannonian quartziferous andesites. The third stage with polymetallic mineralization placed in the eastern of the Gutâi Mountains is associated with Pontian pyroxene andesites (Giuşcă et al., 1973, Borcoş et al., 1973). In Ilba – Nistru zone and in the eastern part of Baia Mare area, Herja-Baiut zone, there are polymetallic mineralizations (base-metal) and in Săsar zone the gold-silver mineralization prevail.

The mineralizations from Rodna zone are associated to the Neogene volcanism and mesometamorphic formations in Rebra series.

The mineralizations from Rodna Veche (Valea Vinului) are associated to the intrusive Neogene magmatism which manifested explosively leading to the formation of explosion and collapse breccia (Socolescu et al., 1977). The litho - petrographic environment represented by amphibolites and crystalline limestone of Rebra - Barnar series had an important role in the formation of different types of metal accumulations (Kräutner 1984). Iron sulphide mineralization (pyrite, pyrrhotite) were formed mainly from the contact of igneous rocks bodies with amphibolites of Rebra series. At the contact with crystalline limestone the sphalerite and galena mineralization were formed, particularly under the form of metasomatic bodies.

The mineralizations from Valea Blaznei are associated to the mesometamorphic rocks of Rebra type from South Eastern part of Rodna Mountains. The mineralization occurs as bands, lenses of various size consistently arranged in horizontal crystalline limestone (Udubaşa 1972). The mineralization consists mainly of galena, sphalerite and pyrite.

In the Maramureş Mountains there are mineralizations associated to metamorphic rocks of Tulgheş Series and metallogenesis associated to Neogene magmatic rocks of the Eastern Carpathians. In the series of Tulgheş three complexes can be separated. The upper complex consists of sericite-chlorite schist, green schist and intercalations of chlorite-sericite schists with albite porphyroblasts. According to Kräutner (1984) they have a basic-acid bimodal volcanic character. With this complex the massive pyrite mineralizations with sphalerite, galena are associated. The Neogene mineralization appear as veins predominantly of copper associated to Toroiaga subvolcanic massive. The subvolcanic massive consists of eruptive rocks of quartz-diorite affiliation, formed after a polyphasic magmatic activity, represented by different types of andesites and quartz diorite (Berza et al., 1981). The mineralization consists of pyrite, chalcopyrite, sphalerite, galena, and sulphosalts of Cu, Pb, Ag, (Steclaci 1962).

3. RADIUM IN GROUNDWATER

The emergence of radium in groundwater, as well as other chemical compounds, is due to the interaction of water with rocks with which it comes into contact, a process influenced by several factors, such as its solubility and the way of radioactive decay. The Radium, which arises by a radioactive alpha decay, is dislocated from the crystal lattice of minerals and accumulates itself in crevices of rocks, which falls more easily in direct contact with the groundwater. The radium concentration in groundwater depends on its concentration in soil, physical and chemical properties of water and soil, the character and the pore structure from the rocks system and last but not least, the physic - chemical properties of radium.

The measured values of ^{226}Ra concentrations in different types of rocks, in the Nordic Countries, are given in table 1, (The Radiation Protection Authorities în Denmark, Finland, Iceland, Norway and Sweden, 2000).

Currently the mining activities are stopped and the mine galleries are found in various closure stages. Once the flowing water is out of the mine galleries, it can reach directly the environment, or, it may pass, in advance, through a treatment plant and then is discharged into the environment. Note that the scope of the mine water treatment is not to decrease the natural radionuclides concentration, but to reduce the contained water with chemical elements that can reach the surface water.

Water samples were collected from 19 underground mine galleries from Gutâi, Rodna and Maramureș Mountains (Fig. 1). All the galleries from which the samples were taken are having the operating activities halted for several years.

4. MATERIALS AND METHODS

The water samples were collected from 19 mine galleries, brought to pH 1-2 with hydrochloric acid, and hermetically sealed into glass containers. The acid was added to prevent radium accession to walls of the container, to inhibit the biological activity during water storage containers and to avoid any non-homogenous distribution of the radium in the collected water sample. Water samples were taken only from those mine galleries where a strong and steady water flow from the inside gallery was, avoiding galleries with pool water, because of the possibility of water intake from outside the mines. In most mine galleries the water flows naturally, but in some, the water comes through mounted pipes, for cleaning, before discharging into the environment.

The collected water samples were measured using two methods, both based on the secular equilibrium principle between ^{226}Ra from water and its decay product, radon (^{222}Rn). Radon was measured by scintillation, using two types of Lucas cells: with a volume of 1000 ml and 150 ml, respectively.

The used principle underlying the determination of ^{226}Ra concentration in water samples is based on the secular equilibrium between ^{226}Ra and its decay product ^{222}Rn. The radioactive decay of ^{226}Ra to ^{222}Rn and ^{218}Po is expressed by (1), (Cosma & Jurcut, 1996).

$$ {}^{226}_{88}Ra \xrightarrow[1600 years]{} {}^{222}_{82}Rn \xrightarrow[3.82 days]{} {}^{218}_{84}Po \qquad (1)$$

For the radium in water activity determination two methods were used. Both methods are based on radon extraction, as a decay product of radium present in the water samples and its measurement by scintillation, using Lucas cells·

To achieve the secular equilibrium, between radium and radon, the collected water samples were kept sealed for longer period than 30 days.

After the secular equilibrium is achieved, by measuring the radon activity, the radium activity is practically determinate (Moldovan et al., 2009).

The first method (1000 ml Lucas cell)

For the first method, the measuring device is composed from a Lucas cell, with a volume of 1000 ml, attached to a photomultiplier connected to a single-channel analyzer (Fig. 2). The scintillation produced by an alpha particle and the ZnS from the Lucas cell, passes through a window located at the top of the Lucas cell, in a photomultiplier tube, closely related to it. The signal is generated by the photomultiplier and then recorded, using a single channel analyzer.

After achieving secular equilibrium, radon gas is extracted from the water sample, and it is transferred to a Lucas cell (scintillation camera), the air being evacuated in advance. Measurement time was set at 100 seconds. The used method is described in detail by Begy et al., (2012).

Table 1. Measured values of ^{226}Ra concentrations in different types of rocks, in the Nordic Countries

	Radium concentration (Bq/kg)
Granite	20 – 130
Diorite, gabbro, basic volcanic rocks	1 – 30
Sandstones and quartzites	5 – 60
Gneiss	25 – 130
Carbonate rocks	10 – 650
Limestone and dolomite	3 – 30
Shale	10 – 150
Alum shale	100 – 4300

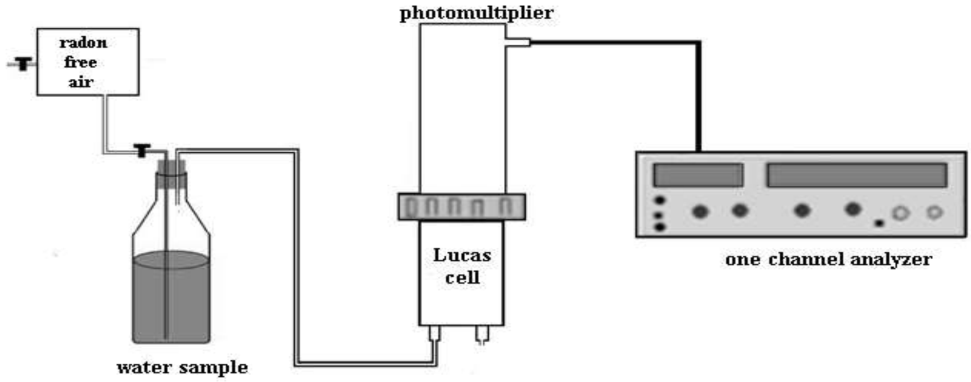

Figure 2: 1000ml Lucas cell method scheme.

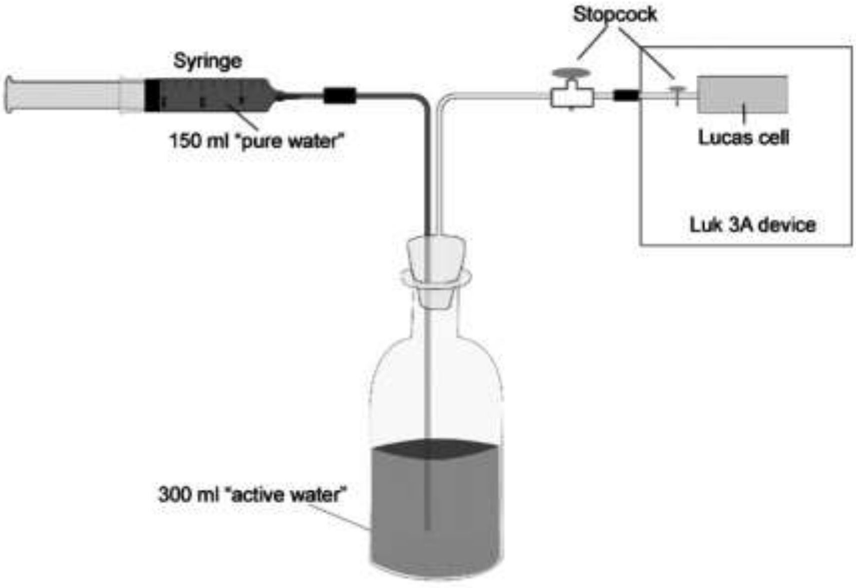

Figure 3. LUK-VR device for radon in water.

The radon concentration in water for the above described method is (Begy et al., 2012):

$$C_{Rn}(Bq/l) = C_{Luc}\left(k + \frac{v_A}{v_W}\right) \qquad (2)$$

where: C_{Rn} - radon concentration in water;

$$C_{Luc} = \frac{N_s - N_B}{\varepsilon \cdot t \cdot e^{-\lambda R}}$$

$$C_{Luc} = \frac{N_s - N_B}{\varepsilon \cdot t \cdot e^{-\lambda_{Rn} t}}$$

(3) – Lucas cell radon concentration;

k – solubility coefficient of radon in water (Ostwald coefficient);

V_A - volume of air in the measurement system ($V_A + V_D = V_S$),
where: - V_S - connecting tubes volume,
V_D - Lucas cell volume;

$V_W = 0.5$ l - water sample volume;

N_S - sample pulses number recorded by the analyzer

N_B - background pulses number recorded by the analyzer;

ε - Lucas cell efficiency;

$t = 100$ sec. - measurement time;

λ_{Rn} - radon decay constant.

The limit of quantification (determination) for the 1000 ml Lucas cells is 20mBq/l.

The second method (145 ml Lucas cell)

In the case of the second method, mine water samples were also obtained from the original samples and stored as stated above.

The radon from water was measured using Lucas cells type LUK-3C specially adapted for measuring radon in water. The measuring equipment, called LUK-VR, includes a 500 ml volume scrubber where a known amount of water (300 ml) is transferred.

Before the measurement, the water sample was brought to the room temperature and, with vigorous stirring, the balance between the radon in water and the air above it was achieved. The scrubber (Fig. 3) was connected on one side to the Lucas cell, evacuated previously of air through a valve, and on the other side to a Janet syringe containing a quantity of 150 ml of radon free water. By opening Lucas cell coupling valve and introducing the water from Janet syringe, the air contains radon from scrubber passes in the Lucas cell, which has a volume of 145 ml. The used protocol is described in detail by Cosma et al., (2008).

Because of the relatively low radium activity in water compared with the radon activity in fresh water, for which this method was initially adapted by Cosma et al., (2008), lower detection and quantification limits were needed. Thus, by increasing the measuring time to 2400 seconds the quantification limit was brought down to 50mBq/l and a new calibration constant of 5.3 were determined by experiments in our laboratory. The radium concentration in water (Bq/l) was determinate using the following equation:

$$A(Bq/l) = 5.3 \cdot N\,(c/s) \qquad (4)$$

where N (c/s) is the number of measured counts/second.

5. RESULTS AND DISCUSSION

Using both measurement protocols described above (1000 ml Lucas cell scintillation method and 145 ml Lucas cell scintillation method) the radium concentrations from 19 water samples were determined (Table 2).

Because of high statistical errors involved, for the second used method (145 ml Lucas cells) and also because of measurement errors for the first used method (1000 ml Lucas cells), relative large differences between the obtained values of each method for the same sample can be found.

The lowest value of the radium concentration was found in a sample from Baia Sprie –Boului Gallery in Maramures County. The value is under the quantification limit of both used methods.

The highest value of 510 ±50 mBq/l obtained with the 1000 ml Lucas cells method and 501±46 mBq/l obtained with the 145 ml Lucas cell method was found for a sample from Nistru – The Village Gallery in Maramures County The average radium concentration obtained values are 138mBq/l for the 1000 ml Lucas cell method and 171 mBq/l for the 145 ml Lucas cell method, see table 2.

Table 2. Radium concentration from mine water measured with two methods.

Water samples	**Radium concentration (mBq/l)**	
	Lucas cell (1000 ml)	Lucas cell (145 ml)
E1 - Baia Mare –SCHWEITZER Gallery	**150** ±15	**186** ±30
E2 - Baia Mare – Valea Rosie Gallery 1	**70** ±4.9	**75** ±7
E3 - Baia Mare – Valea Rosie Gallery 2	**98** ±7	**98** ±8
E4 - Baia Mare –Mina Herja Gallery	**410** ±40	**463** ±51
E5 - Baia Mare –Dealu Crucii Gallery	**51** ±5	<50
E6 - Baia Mare –Sasar Gallery	**112** ±10	**122** ±11
E7 - Nistru – The Village Gallery	**510** ±40	**501** ±46
E8 - Baita – Purge Station Gallery	**212** ±21	**254** ±31
E9 - Baia Sprie –Boului Gallery	<20	<50
E10 - Suior – Mining Gallery	**116** ± 12	**148** ±25
E11 - Cavnic –REINER (Hospital) Gallery	**141** ±15	**113** ±23
E12 - Cavnic – Flotation Station Gallery	**325** ±30	**286** ±27
E13 - Cavnic –ROATA Gallery	**72** ±6	**70** ±7
E14 - Baiut – The Village Gallery	**45**±4	**50** ±6
E15 - Valea Vinului –Izvorul Rosu Gallery	**74** ±6	**66** ±8
E16 - Valea Blaznei Gallery	**60** ±6	<50
E17 - Valea Mare –Faget Mine Gallery	**57** ±6	**69** ±7
E18 - Baia Borsa –EMERICH Gallery	**53** ±4	**59** ±6
E19 - Baia Borsa –23 August Gallery	**56** ±5	<50
Average	**138**	**171**

The average difference between the two used methods (see Table 2) is about 18% and it is connected with relative large statistical and measurement errors in these determinations.

Of the 19 water samples, four (21%) samples have values greater than the MCL (Maximum Contaminant Level - 185 mBq/l) (US Environmental Protection Agency, 2000) and (78%) had values below the MCL.

In southern New Jersey, USA, 33% from 170 water samples, radium total concentrations exceeded the MCL of 5 pCi/l (Szabo & De Paul, 1998).

The reported values of ^{226}Ra activity in the water samples of the present study are low and are comparable with those of the values reported elsewhere. In the Cauvery river ecosystem, India, the radium concentration range between 0.82 and 1.06 mBq/l . Most rivers have low ^{226}Ra values, like some rivers in Russia, the average being 155 mBq/l, (Kuptsov et al., 1969) and in India, parts of the Ganges are having 48.1 mBq/l (Banerjee & Chatterjee, 1966) and Periyar river 122 mBq/l (Paul et al., 1980).

6. CONCLUSIONS

According to the results of this study, it is evident that the radium activity of the measured water mine samples from the North of Transylvania presents no significant risk for the environment. The radium concentration of four water samples (E4, E7, E8, E12) is greater than 185 mBq/l MCL, the rest of the samples having radium concentrations below the MCL value.

For a single mine water sample (E7 - Nistru - The Village Gallery) a higher ^{226}Ra value than 500 mBq/l was obtained; the 500 mBq/l value representing the maximum ^{226}Ra allowed in drinking water value suggested by ICRP (International Commission on Radiological Protection) as the derived concentration for a group of individuals (Botezatu et al., 2001).

Thus, the low radium concentration in the mine water of the studied region was highlighted and no significant health risk, to the people who use contaminated surface water, with mine water, was found.

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