Carpathian Journal of Earth and Environmental Sciences, August 2013, Vol. 8, No. 3, p. 81 - 86

RADON CONCENTRATION IN GROUND WATER FROM MĂGURI RĂCĂTAU AREA, CLUJ COUNTY

Mircea MOLDOVAN[1], Dan Constantin NIȚĂ[2*], Dan COSTIN[1] & Constantin COSMA[2]
[1]Babes-Bolyai University, Faculty of Environmental Science and Engineering, Cluj-Napoca, Romania
[2] Environmental Radioactivity and Nuclear Dating Research Group, Babes-Bolyai University, Cluj Napoca Romania
**Corresponding author - dan.nita@ubbcluj.ro*

Abstract: Măguri Răcătău area is characterrised by granitic rock presence at surface, favorable to radon emission and accumulation in soil and underground water. The radon measurements were made using the LUK-VR system which is based on radon gas measurement with Lucas scintillation cell. In 55 water samples taken from different points of interest from Măguri Răcătău area, Cluj County, radon concentration was measured. The results show that the radon concentrations are within the range of 3.9 Bq/L and 361.9 Bq/L with an average value of 68.4 Bq/L for all types of water. From the results of these measurements, a correlation may be observed between the radon concentration and the granite geological formations. In most of the cases, high values for radon in drinking water were found**.** The average effective dose equivalent from radon in well water and spring water has been calculated, the obtained values being 0.49 mSv·y^{-1} and 0.55 mSv·y^{-1}, respectively**.**

Keywords: radon, groundwater, effective dose, double lognormal, granite

1. INTRODUCTION

Radon-222 is an inert gas, whose concentrations in ground water are usually controlled and related to a number of factors, including ^{222}Rn emission from surrounding rocks, temperature, pressure, rainfall, and earthquake activities (Hess et al., 1985, Loomis et al., 1988). Various investigators have reported conflicting findings on the temporal stability of groundwater ^{222}Rn concentrations.

The concentration of ^{222}Rn in ground water depends on the concentration of its parent ^{226}Ra, in the underlying rock. The short half-life of ^{222}Rn (3.82 days) together with the slow rate of migration of ground water allows the ^{222}Rn in solution to be in approximate secular equilibrium with the ^{226}Ra in the local rock. Radon concentrations in water have been known to be high in most granites and in high-grade metamorphic rocks, whereas less metamorphosed rocks have somewhat less ^{226}Ra. High concentrations of radon in ground water from the bedrock are frequent in the areas where the bedrocks consist of granites, (UNSCEAR, 2006).

High radon concentrations sometimes also occur in association with pegmatites, as well as, gneisses and vulcanites rich in quartz and feldspar (Žunica et al., 2006).

The radon concentration in groundwater from bedrock is low in the area where uranium concentration in rocks is low (Song et al., 2012).

Radon and its progenies are recognized as the most significant natural source of human radiation exposure and the most important cause of lung cancer incidence, after smoking (I.A.R.C., 1988). After several studies on the effect of radon on human health is accepted that there is a significant correlation between exposure to radon and lung cancer (B.E.I.R. VI, 1999).

Due to its negative impact on human health radon in water has attracted considerable attention in recent years (Adrovic et al., 2009; Kozlowska et al., 2009; Yigitoglu et al., 2010).

Thus, the importance of radon concentration measurements was highlighted, especially in water. The water transports radon from underground to surface and radon can diffuse in indoor air if the water is used in household (Maged, 2009).

If the water is used as drinking water, the human radon exposure is by ingestion. The radon in water ingestion can add an additional exposure dose to the stomach and also to the whole body (Hopke et al., 2000).

This has generated global interests in water monitoring and some national authorities have established water standards that stipulate maximum levels for radon and other radionuclides (Health Canada, 2009).

The first aim of this paper is to describe ^{222}Rn concentrations in different drinking water supplies of Măguri Răcătau area, around the Muntele Mare granitoid, the largest intrusive body in the Apuseni Mountains. The main reason of this study is related with the mentioned area location

The basement of this area is mostly granitic and these results can make an important data contribution to the literature, being also valuable for local population. The second aim of this study was to see if there is a correlation between geological structure and radon in water. The results are also compared with other studies.

2. MATERIALS AND METHODS

The analyzed water from different sources are both used as drinking water and for other domestic uses by the population from Măguri-Răcătau area.

2.1. Characteristic of geological background in area Măguri-Răcătău

The Măguri-Răcătău village, located at about 40 km west from Cluj-Napoca city, covers a total area of 27,000 ha in the geographical unit named Muntele Mare Mountain (North-Eastern part of Apuseni Mountains). The specific topography is due to the presence of medium height mountains (500 to 1800 m altitude) with near flat, large top zones and steep valleys (e.g. Someşul Rece Valley). The study area is underlain by Someş metamorphic sequence and magmatic rocks (Balintoni, 1997).

Gneiss, amphibolites, muscovite and chlorite schists, quartzite are the main metamorphic rocks. These rocks are intruded by Muntele Mare granitoid, an extremely complex magmatic body. The granitoid emplacement generated various contact metamorphic rocks (Ghergari & Mariş, 2007).

Muntele Mare granitoid, the largest intrusive body in the Apuseni Mountains, has an North-South elongated shape covering approximately 300 km^2. The emplacement age of this pluton composed mainly by biotite granodiorite is late Variscan (297-291 Ma) (Balintoni et al., 2009).

Various textural rock types have been described: porphyritic, pegmatitic, microgranular and gneissic. The granitoid mineralogical composition consists of plagioclase, quartz, K-feldspar, biotite, muscovite, tourmaline, apatite, zircon, monazite and allanite (figure 1.).

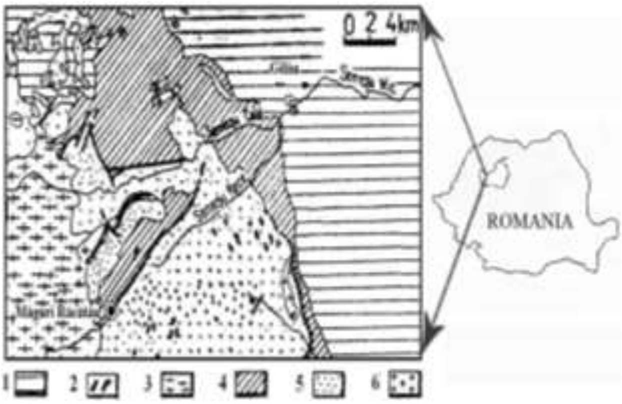

Figure 1. Simplified geological map of north-eastern area of Muntele Mare Mountain (Mârza, 1980).
Legend: 1 – sedimentary rocks; 2 – pegmatites; 3 – granitic rocks; 4 – low metamorphic rocks; 5 – medium metamorphic rocks; 6 – Laramic and Neogen magmatic rocks.

2.2. Sampling procedure

The samples were collected from places randomly selected. Water samples (drinking water) originating from different wells, springs and surface water located in Cluj County (Măguri Răcătău, Mărişel, Muntele Rece, Măguri) were collected in 2012. The water samples were collected during spring and summer. The sampling procedure was carried out using 500 ml polyethylene bottles that had been previously washed with pure water.

2.3. Analysis of water samples

Radon analyses were conducted using a LUK-VR system equipped with Lucas cells and a special device for dissolved gas extraction (Plch, 2002).

For a temperature of 20°C, between the radon content in water A_w and the number of counts/second N (c/s) recorded by the device, the following relationship was established:

$$A_w = 7.7 \times N \text{ (c/s)} \quad (1)$$

Statistical errors associated with the radon analysis were 5-6% (Cosma et al., 2008; Moldovan et al., 2009).

2.4. Effective doses

The annual effective doses for ingestion were estimated according to parameters introduced by UNSCEAR report (UNSCEAR, 2006).

The annual effective dose due to intake of radon was calculated on the basis of the mean activity concentration using the relation:

$$D_{water} = F_{Rn} * C_{w} * A_w \quad (2)$$

where F_{Rn} is the committed effective dose per unit radon in water intake for adults taken as 10^{-8} Sv/Bq (UNSCEAR, 2006), C_w is the water consumption rate (L/y) taken to be 1 L per day and A_w is the activity of radon in water.

3. RESULTS AND DISCUSSION

3.1. Radon in water and connection with geological background

The results presented and discussed in this paper include 55 water samples, 24 water samples were collected from wells, 27 water samples collected from springs and 4 from surface waters.

Radon concentration in water samples collected from this area has values between 3.9 and 361.9 Bq/L, with an geometric mean (GM) value of 31.47 Bq/L and median (MV) of 22.7 Bq/L. This big diference between GM and MV says that the distribution is not lognormal.

The average of radon concentration in water (55 samples) obtained in Măguri Răcătău area is 68.4 Bq/L, this value being for times higher than the average obtained in water from Transylvania (Romania), 15.4 Bq/L (Cosma et al., 2008). Thus, we conclude that this area is a high radon area in Romania.

The summary results of radon content in water from different sources from the study area are shown in table 1. Elevated levels of radon concentration were found in samples collected from Măguri Răcătău area where the geology is characterized by basement complex with granite as the most extensive unit. The existence of high radon concentrations and granite geological formations confirms the link between them (Varley & Flowers, 1992).

The maximum radon concentration in spring water (27) was found in a spring from Măguri Răcătău village, 278.1 Bq/L. 9 water samples from spring water (33.3%) are values more than 100 Bq/L and 18 (55.5%) values less 100 Bq/L. The results show that the highest value was measured in a well water sample from Marisel village, 361 Bq/L. The average of radon in well water (24) is 79.2 Bq/L with 14 values (58.3%) higher than 100 Bq/L, and 10 values (41.6%) less than 100 Bq/L, value recommended by the World Health Organization (WHO, 2008) in drinking water for public water supplies.

Of the 55 water samples analyzed, 51 are used in household water and drinking water (except surface waters), 12 (23.5%) drinking water samples having less than the maximum contaminant level (MCL), 39 water samples (76.4%) having values higher than MCL (MCL=11.1 Bq/L) proposed by United States Environmental Protection Agency for radon level in drinking water (US.EPA, 1991).

For the drinking water with concentration higher than the recommended values, it is recommended to remove the radon before consumption, by shaking, bubbling or by using active charcoal filters for water.

Water distribution (excluding the surfaces water) is a double lognormal distribution (Fig. 1). The first distribution of 33 water samples is a lognormal distribution with arithmetic mean, geometric mean and median to 17.58 Bq/L, 15.4 Bq/L and 15.8 Bq/L respectively.

As can see in figure 2, in the second group of 18 water samples was determined average of 175.4 Bq/L, the geometric mean of 166.7 Bq/L and median is 154.7 Bq/L, the distribution of these waters is also one lognormal. This highlights the existence of additional sources of radon pronounced, in this case the granite from geological substrate. Similar distribution with two peaks was obtained in waters from South of Belgium by Hallez (Hallez & Flemal, 1994).

3.2. Estimated dose

The average effective doses (ingestion due to radon from water) were calculated using committed effective dose of radon in water for adults from UNSCEAR 10^{-8} Sv/Bq (UNSCEAR, 2006).

From the present data, two liters per day of drinking water from the same source, delivers to adults a average effective dose equivalents 0.49 mSv/y from spring water and 0.55 mSv/y. The estimate dose varies between 0.067 mSv/y and 2.212 mSv/y in wells water

Table 1. Descriptive statistics of water samples from Măguri Răcătău area.

Water sample	Frequency	A.M. ± S.D. (Bq/ L)	G.M. ± G.S.D. (Bq/L)	Range (Bq/L)
Well waters	24	79.2 ± 22.5	42.5± 18.5	9.6-316
Spring water	27	70.6 ± 3.5	30.5±16.4	3.9-278.7
Surface waters	4	6.4 ± 1.2	3.2 ± 1.4	5.4 – 7.8

and between 0.027 mSv/y and 1.95 mSv/y in spring waters.

The contribution of drinking water to total exposure is typically very small and is largely due to naturally occurring radionuclides in the uranium and thorium decay series (WHO, 2004).

In Măguri Răcătau area it was observed that the estimated dose, due to ingestion has 18 (35%) values above and 33 (65%) below the recommended value of 0.3mSv/y by WHO and UNSCEAR (WHO 2008; UNSCEAR 2000).

A comparison of the average effective doses estimated from the present study with the results from other studies carried out in the world is given in table 2.

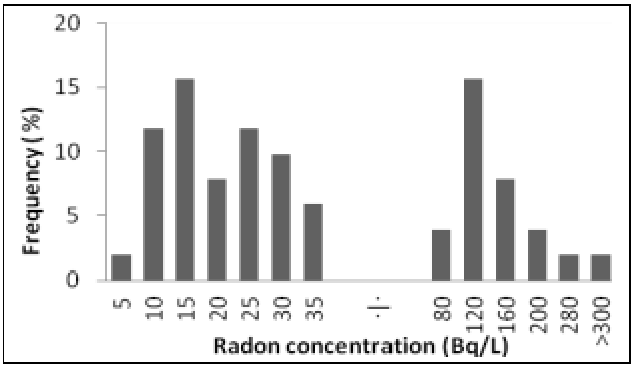

Figure 2. Histogram of radon concentration in drinking water measurements in Măguri Răcătău area.

Dose estimates are similar to those determined in Pakistan where geological substrate in most part is composed by the limestone and sandstone formations of the Ligocene and Miocene age and the rocks with sedimentary origin in age from Jurassic to Paleocene.

In most cases the estimated dose is on average lower than the average obtained by us, this is due to the fact that the geological structure is different from that found in Măguri Răcătau area.

This confirms once again that the presence of granitic rocks has an effect on groundwater, enriching them with radon.

4. CONCLUSIONS

The high concentration of radon in some water samples can be explained by the fact that the region is dominated by granite intrusions.

The results of this study indicate that the radon concentrations in drinking water samples in Măguri Răcătau area are majority below the proposed reference level of the EU Commission Recommendation (European Commission 2001) with important exceptions (18 water samples from 55).

The annual effective dose due to radon ingestion is consistent with WHO recommended dose level with the exception of 35% (i.e. 18 samples). The results of this study indicate that Măguri Răcătău area is a region with an average of four times higher radon in water concentration than the average determined in Romania.

Another important result is the confirmation of a double lognormal distribution for the radon concentration in the underground waters from the studied area.

The radon concentrations found in different water sources suggests the radon concentrations in soil and thus, also the indoor radon concentration. No data regarding lung cancer incidence in the studied area exists, but future studies on indoor radon will be carried out in Măguri Răcătau area.

REFERENCES

Adrovic, F., Kasic, A., Kasumovic, A. & Tresnjo, Z., 2009. *Investigation of radon in drinking of the North-Eastern region of Bosnia and Herzegovina*. Technics Technologies Education Management, 4, 201 - 207.

Table 2. The comparison of equivalent doses (ingestion) with previous results from different countries

Country	Annual effective dose (mSv/year)	References
Serbia	0,01	(Todorovic et al., 2012)
Pakistan	0.73-2.03	(Ali et al., 2010)
India	0.009-1.586	(Mahesh et al., 2001)
China	0.03-0.14	(Xinwei 2006)
Turkey	0.03-0.05	(Yigitoglu et al, 2010)
Sudan	0.001-0.440(0.11)	(Hajo et al.,2011)
Iran	0.1-0.5	(Binesh et al., 2011)
Italia	0.02	(Desideri et al., 2005)
Finland	0.29	(Vesterbacka et al .,2005)
Austria	0.01-0.86	(Wallner & Steinger, 2007)
Romania	**0.027-2.212**	**This study**

Ali N., Khan E. U., Akhter, P., Khan, F. & Waheed A., 2010. *Estimation of mean annual effective dose through radon concentration in the water and indoor air of Islamabad and Murree.* Radiation Protection Dosimetry, 141, 2, 183–191.

B.E.I.R. VI (Board on Radiation Effects Research VI)**,** 1999. *Health Effects of Exposure to Radon*. Washington, D.C. National Academy Press.

Balintoni, I., 1997. *The geotectonics of the metamorphic terrains from Romania*. Ed. Carpatica, Cluj Napoca, p.176 (in Romanian).

Balintoni, I., Balica C., Cliveți M., Li L.Q., Hann H.P., Chen, F. & Schuller, V., 2009. *The emplacement age of the Muntele Mare Variscan granite (Apuseni Mountains, Romania).* Studia Universitatis Babeş-Bolyai, Geologica Carpathica, 60, 495-504.

Binesh, A. & Arabshahi, H., 2011. *Radon and Radium Concentrations in 120 Samples of Drinking, Springs and Rivers Water Sources of North West Regions of Mashhad*. Environmental Research Journal, 15, 3, 117-120.

Commission of the European Communities. Commission recommendation of 20 December 2001 on the protection of the public against exposure to radon in drinking water supplies: *Official Journal of the European Communities*

Cosma, C., Moldovan M., Dicu T. & Kovacs T., 2008. *Radon in water from Transylvania (Romania)* Radiation Measurements, 43, 1423-1428.

Desideri, D., Roselli, C., Rongoni, A. & Saetta, D., 2005. *^{222}Rn determination in drinkable waters of a central eastern Italian area: Comparison between liquid scintillation and gamma-spectrometry*. Journal of Radioanalytical and Nuclear Chemistry, 266, 191-197.

Ghergari, L. & Mariş C., 2007. *Petrology and geochemistry of the Muntele Mare granitoid (Northern Apuseni Mountains), Romania*. Studia Universitatis Babeş-Bolyai, Geologia, 52, 2, 11 - 18.

Hajo, I., Salih, I. & Sam, A.K., 2011. *Study of radon in ground water and physicochemical parameters in Khartoum state* Journal of Radioanalytical and Nuclear Chemistry, 290, 333-338.

Hallez, S. & Flemal, J.M., 1994. *Le Radon, mythe ou realite: La situation en Belgique*. Annales de L'Association Belge de Radioprotection, 19, 227.

Health Canada, 2009. *Guidelines for Canadian Drinking Water Quality: Guideline Technical Document - Radiological Parameters.* Radiation Protection Bureau, Healthy Environments and Consumer Safety Branch, Health Canada, Ottawa, Ontario.

Hess, C.T., Michel, J., Horton, T.R., Prichard, H.M. & Conglio, W.A., 1985. *The occurrence of radioactivity in public water supplies in the United States*. Health Physics, 48, 5, 553-586.

Hopke, P.K., Borak, T.B., Doull, J., Cleaver, J.E., Eckerman, K.F., Gundersen, L.S., Harley, N.H., Hess, C.T., Kinner, N.E., Kopecky, K.J., Mckone, T.E., Sextro, R.G. & Simon, S.L., 2000. *Health risks due to radon in drinking water,* Environmental Science & Technology, 34, 921–926.

I.A.R.C. (International Agency for Research on Cancer).1988. *Manmade mineral fibres and radon. Monographs on the Evaluation of Carcinogenic Risks to Humans*. IARC Scientific Publications 43 (Lyon, France: IARC), 173–259

Kozlowska, B., Morelli, D., Walencik, A., Dorda, J., Altamore, I., Chieffalo, V., Giammanco, S., Imme, G. & Zipper, W., 2009. *Radioactivity in waters of Mt. Etna (Italy)*. Radiation Measurements, 44, 384–389.

Loomis, D.P., Watson, J.E. & Crawford-Brown, D.J., 1988. *Predicting the occurrence of Rn-222 in North Carolia groundwater*, Environmental Geochemistry and Health, 10, 41-45.

Maged, F., 2009. *Estimating the radon concentration in water and indoor air,* Enviro. Monit. Assess. 152, 195-201.

Mahesh, H.M., Avadhani, D.N., Karunakara, N., Somashekarappa, H.M., Narayana, Y. & Siddappa, K., 2001. *^{222}Rn Concentration in ground waters of coastal Karnataka and Kaiga of south west coast of India*, Health Physics, 81, 6, 724-728.

Mârza I., 1980. *Considérations génétiques sur les pegmatites du cristalin du Gilău - Apuseni. La province pegmatitique Carpatique*. Ann. Inst. Geol. and Geophys., LVII, 423-431.

Moldovan, M., Cosma, C., Encian, I. & Dicu, T., 2009. *Radium-226 concentration in Romanian bottled mineral waters*, Journal of Radioanalytical and Nuclear Chemistry, 279, 487–491.

Plch, J., 2002. *Radon Detector LUK 3A. Manual for Operating LUK 3A*. Jiri Plch M Eng SMM, Prague.

Song, G., Chen, D., Tang, Z., Zhang, Z. & Xie, W., 2012. *Natural radioactivity levels in topsoil from the Pearl River Delta Zone, Guangdong, China*. Journal of Environmental Radioactivity, 103, 1, 48–53.

Todorovic, N., Nikolov, J., Forkapic, S., Bikit, I., Mrdja, D., Krmar, M. & Veskovic, M., 2012. *Public exposure to radon in drinking water in Serbia*, Applied Radiation and Isotopes, 70, 543 - 549.

UNSCEAR, 2000. *Sources, effects and risks of ionizing radiation*. UNSCEAR report to the General Assembly. New York, NY, United Nations Scientific Committee on the Effects of Atomic Radiation.

UNSCEAR, 2006 United Nations Scientific Committee on the Effects of Atomic Radiation. Annex E: *Sources-to-Effects Assessment for Radon in Homes and Workplaces.* United Nations, New York.

US.EPA, 1991. *National primary drinking water regulation, radionuclides* (proposed rules), vol

56. US Environmental Protection Agency, Federal Register, p 138

Varley, N.R. & Flowers, A.G., 1992. *Radon and its Correlation with Some Geological Features of the South-West of England*. Radiation Protection Dosimetry, 45 ,1-4, 245-248.

Vesterbacka, P., Makelainen I. & Arvela H., 2005. *Natural radioactivity in drinking water in private wells in Finland. Radiat Prot Dosimetry*, 113, 2, 223-232.

Wallner, G. & Steinger, G., 2007. *Radium isotopes and ^{222}Rn in Austrian deinking waters* Journal of Radioanalytical and Nuclear Chemistry, 274, 3, 511-516.

World Health Organisation (WHO), 2004. *Guidelines for Drinking Water Quality*, (third edition) Vol. 1. World Health Organisation, Geneva.

World Health Organization, (WHO) 2008 *Guidelines for Drinking-water Quality*, Third Edition incorporating the first and second addenda, Volume 1, Recommendations , Geneva.

Xinwei, L., 2006. *Analysis of radon concentration in drinking water in Baoji (China) and the associated health effects*. Radiation Protection Dosimetry, 121, 4, 452–455.

Yigitoglu, I., Oner, F., Yalim, H.A., Akkurt, A., Okur, A. & Ozkan, A., 2010. *Radon concentrations in water in the regionof Tokat city in Turkey*, Radiation Protection Dosimetry, 142, 2-4, 358-362.

Žunića, Z.S., Kobalb, I., Vaupotičb, J,. Kozakc, K,. Mazurc, J., Birovljevd, A., Janikc, M., Čelikovića, I., Ujića, P., Demajoa, A., Krstiće, G., Jakupie, B., Quartof, M. & Bochicchio, F., 2006. *High natural radiation exposure in radon spa areas: a detailed field investigation in Niška Banja (Balkan region)*, Journal of Environmental Radioactivity, 89, 3, 249 – 260.

Received at: 11. 01. 2013
Revised at: 16. 05. 2013
Accepted for publication at: 25. 06. 2013
Published online at: 27. 06. 2013