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RADON AND RADIUM CONCENTRATIONS IN WATER FROM TRANSYLVANIA AND THE ASSESSMENT OF THE RESULTING DOSE

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Abstract:
The results present here are from a survey carried out in the Transylvania region in Romania for radon and radium concentrations in natural waters (surface, springs, wells, mineral water). The radon concentration is equal to the radium concentration, this happens after a period of 30 days when radium can be considered in secular equilibrium with radon. The measurements were made using the LUK-VR system that is based on radon gas measurement with Lucas cell, specially adapted for radon in water measurement. The results show that the radon concentrations are within the range of 0,5 – 90,7Bq/l with an average value of 16,3Bq/l for all types of water covered within this survey. For the radium concentration ranged from 0,5mBq/l to 397mBq/l. The estimated committed effective dose of different age groups of radon and radium from drinking 1l day^{-1} different type of water, using the ingestion dose conversion coefficients from the IAEA (1995) and WHO (1993) ranged from 0,017mSvy^{-1} to 0,855mSvy^{-1} to radon and ranged from 0,019mSvy^{-1} to 0,242mSvy^{-1} to radium. The expected doses from consumption of water in Romania are insignificant compared with other natural sources of radiation (such as radon in indoor air, ^{40}K, etc.).
Keywords: well waters speings water, radon, radium,dose.

1. Introduction

Radium (^{226}Ra) and radon (^{222}Rn) are radioactive progeny in the decay chain of ^{238}U. ^{226}Ra is an alpha-emitting radionuclide with a half-life of 1600 years. Within the body it behaves as calcium and is deposited primarily in the bone. After entering the body it is retained with a physiologic half-life exceeding 20 years. Exposure to radon (generic term used commonly to refer to the isotope ^{222}Rn) occurs primarily through inhalation, resulting in radiation dose to the lung and to a lesser degree also other organs through radon gas dissolved in blood. Yet, in conditions with very high concentrations in drinking water, ingestion can be the primary source of exposure to organs other than the lung [1].

In the last decade, the radon issue has become one of the major problems of radiation protection. Radon is odorless, colorless, water-soluble, radioactive and the heaviest noble gas. Radon-222 (and its decay products) is the only radon isotope sufficiently abundant and longlived to cause a health concern. Since rocks and soil contain radium that decays to form radon, both underground water and surface water should contain the dissolved ^{222}Rn gas. The contribution to the mean effective dose equivalent from inhalation of ^{222}Rn and its short-lived decay products is estimated to be about 50% (1.1mSv/year) of the total effective dose equivalent from natural radiation sources. [2]

The concentration of ^{222}Rn in ground water depends on the concentration of its parent ^{226}Ra, in the underlying rock. The short-life of 222Rn (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 granits and in high-grade metamorphic rocks, whereas less metamorphosed rocks have somewhat less ^{226}Ra. The highest ^{226}Ra/^{222}Rn levels in ground water are found in granites, silimanite or silimanite/orthoclase grade rocks; the lowest levels are found in chlorite to staurolite grade rocks. Radon is an inert gas whose concentrations in ground water are reportedly controlled by and related to a number of factors including emission of radon from surrounding rocks, temperature, pressure, rainfall, and earthquake activities. Various investigators have reported conflicting findings on the temporal stability of groundwater radon concentrations. [3, 4].

This paper presents radon and radium concentrations in water from Transylvania; and the effective dose assessments were provided by the activity of the annually ingested multiplied by the proper dose conversion factor.

2. Method and samples

2.1. Sampling

Measurements were carried out in 12 counties from Transylvania and two counties from surrounding areas (Figure 1). From the geological point of view, Transylvania mainly corresponds to a post-tectonic depression, surrounded by the Alpine chain of the Carpathians. In the Eastern Carpathians, three main units can be separated: “the Flysch zone” to the East, “the Mesozoic and metamorphic zone” in the central part, and “the volcanic zone” to the west. In the eastern border of the depression, metamorphic rocks dominate the lithology of the Southern Carpathians. The western part of the chain (Apuseni Mountains) composed of a

mosaic of magmatic, metamorphic and sedimentary rocks. The depression is filled with tertiary, mainly Neogene deposits [5].

In all, 1200 samples from surface, well and springs waters were collected during the springs and summers in the years from 2002 to 2009.

FIGURE 1. Study area.

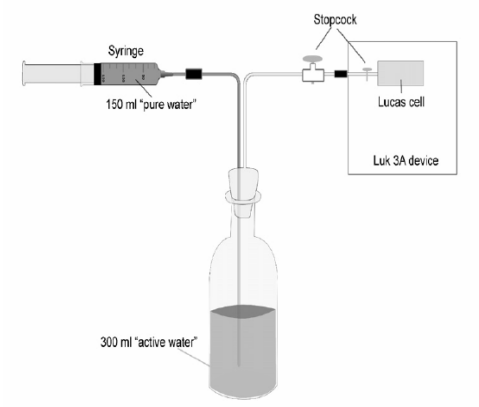

FIGURE 2. LUK-VR device.

2. 2. Measurement of ^{226}Ra and ^{222}Rn concentrations in the water samples

Generally, ^{226}Ra, the direct forerunner of radon, is in secular equilibrium with the last one. We note with λ_{Ra} and λ_{Rn} the decay constant of radium and radon, respectively, and with N_{Ra} and N_{Rn} the number of atoms of radium and radon, respectively. In the case of secular equilibrium, for a time $t \ll T_{1/2}(Ra)$, where $T_{1/2}(Ra) = 1620$ years, the parent radium disintegration speed is actually constant, therefore, we can approximate $e^{-\lambda_{Ra} t} \approx 1$, meaning $N_{Ra} = N_{Ra}(0)$ and the number of radon atoms is given by:

$$N_{Rn} \approx N_{Ra} \frac{\lambda_{Ra}}{\lambda_{Rn}} \left(1 - e^{-\lambda_{Rn} t}\right)$$

Moreover, if $t \geq T_{1/2}(Rn)$, where $T_{1/2}(Rn) = 3.82$ days, then $e^{-\lambda_{Rn} t} \approx 0$, which leads to:

$$N_{Rn} = N_{Ra} \frac{\lambda_{Ra}}{\lambda_{Rn}}$$

or $\lambda_{Rn} N_{Rn} = \lambda_{Ra} N_{Ra}$, which means that the activities of the parent (^{226}Ra) and the daughter (^{222}Rn) become equal. The radon concentration is equal to the radium concentration, this happens after a period of 30 days when radium can be considered in secular equilibrium with radon.

The radon in water was measured using a LUK-3A device specially adapted for radon in water measurement. [6]

The LUK-VR system consists of a LUK-3A device and a scrubber dedicated for radon measurements in water samples, [7]. The scrubber consists of a glass vessel of 500cm^3

volume where a known quantity of water sample (i.e. 300cm^3) is introduced (Figure 2). The principle of operation is that the concentration of the radon dissolved in the water sample is mixed with the air that is on the top of the water level within the scrubber volume. Following this, air is then transferred from the scrubber to be measured for radon using, for example, a Lucas cell method. With this system, the water sample (with radon concentration A and volume V_w) is carefully transferred from the water container into the scrubber. For a certain volume (V_a) of air that is kept above the water level in the scrubber, radon equilibrium is established after a certain period of time between the concentration of radon in the water sample (A_w) and that in the air above the water level (A_a), so that the total radon concentration is divided between the two. The relationship between Aw and Aa is given by the equation derived by Genrim [8]:

$$\alpha = \frac{A_w}{A}\frac{V_a}{V} \tag{1}$$

where (α) is the coefficient of solubility of ^{222}Rn in water. By substituting $A - A_a$ for A_w and rearranging Eq. (1), the radon concentration value A_a that passes from the water sample into the air within the scrubber volume, may be described by the following relationship:

$$A_a = \frac{A}{\alpha}\frac{V_a / V_W}{1 - V_a(\alpha V_W)} \tag{2}$$

2. 3. Extracting radon from the water samples for measurement

The preparation for each water sample is described in the following lines. Before the start of each measurement, the water samples were brought to room temperature. The sample of 300 cm^3 was introduced into the scrubber, and then the scrubber is closed and shaken very well for 1 min. It is then expected that, the equilibrium, between the radon dissolved in the water sample and the air above it, is established as was indicated by producer and verified in our laboratory [9].

The air occupying the upper part of the scrubber (that has become mixed with radon) is then transferred for measurement with the Lucas cell. The transfer process is simply based on using a syringe for adding a volume of 150cm^3 of distilled water (radon-free water) into the water sample in the scrubber, so that the level of water inside the scrubber increases and a similar volume of air is pushed out through the valve (which is to be open during this process) to fill the Lucas cell (of size 145cm^3). The Lucas cell is previously evacuated in order to be filled with the air being forced as a result of the extra water being injected to the scrubber. A

radon progeny filter is introduced between the scrubber and the Lucas cell. The Lucas cell is then connected to a photomultiplier tube for readout.

The radon concentration in the water sample "A" (Bq/l) and the number of collected counts "N" (counts/s) are related as follows:

$$A(Bq/l) = 9.85N(c/s) \quad (3)$$

The efficiency detection of the Lucas cell was determined using a control source manufactured and certified by The State Metrological Institute of Czech Republic.

3. Results and Discussions

3.1 Radon and radium in springs waters

The radon concentration was determined in 123 samples of water collected from springs. The lowest value of the radon concentration of 1,12Bq/l was found in a spring in Mogosa in Maramures County, and the highest value of 72,9Bq/l was found in a spring in Somesul Rece in Cluj county. The average radon concentration value is 20,24Bq/l. The spring waters with relatively high radon concentrations are in springs in Santaimbru (56,9Bq/l), Sansimion (48,8Bq/l) in the Harghita County, and some other springs in Baile Balvanyos (44,6Bq/l), from the Covasna County. These regions are characterized by post-volcanic emanations, present in the form of mofettes.

For radium concentration measured 62 samples of water only 65% have a higher value than the limit of detection device used to determine radium from water (50mBq/l). The maximum value was obtained in a spring of Covasna city with a concentration of 397mBq/l.

3. 2. Radon and radium in wells waters

The radon concentration in water from wells have values between 0,5 – 90,7Bq/l and very few cases were with high radon concentrations. The average value is around 14,8Bq/l. High radon concentration values, as compared to the average value, were found with water samples from wells in Bihor County in Rosia locality (72,7Bq/l), and in Beius zone (62,6Bq/l). Also similar radon concentration levels were found in Hunedoara County in Hateg zone of 70,8Bq/l, in Cluj County in Baisoara village of 55,4Bq/l and Somesul Rece village of 52,4Bq/l. All these localities are in regions where the soil is made up generally of granitic formations. Within the Bihor County, higher radon concentration levels were measured to be in excess of 90Bq/l in regions situated on the limestone plateau of Vascau (Codru–Moma Mountains) in Bihor County.

The radium concentration was determined in 300 water samples collected from wells in different parts of the study area. Of the total samples measured more than 200 samples have a higher radium concentration limit of detection device. The maximum value was determined in a sample of water from Hateg from Hunedoara county (230 mBq/l).

3. 3. Dose due to radon and radium from water

Effective dose due to is calculated by the following formula:

$E = K \times G \times C \times t$

where:

E - effective dose due to radon (radium) ingestion (Sv)

K - conversion factor of radon ingested dose (Sv Bq-1)

G - average consumption of water (it is usually between 1 and 2 liters per day)

C - concentration of radon (radium) in water

t - time drinking water (365 days)

Water considered in this study as drinking water is the water samples collected from springs and wells.

To determine the dose received from ingestion of water to the population using the recommended conversion factor of three international institutions. In any case the effective dose does not exceed 1mSv year^{-1}, contributing to an annual dose of only 0,183mSv year^{-1} in children, if we use factor conversion recommended UNSCEAR (2×10^{-8}SvBq^{-1}), or 0,024mSv year^{-1} if we use conversion factor recommended IRCP ($0{,}07 \times 10^{-6}$SvBq^{-1}). In adults the use of a liter of water per day to contribute to the annual dose value 0,069mSv year^{-1} - if the dose calculation using the UNSCEAR recommended conversion factor (10^{-8}SvBq^{-1}) [10], or 0,482mSv year^{-1}, if we use the conversion factor recommended IRCP (0,01 x 10^{-6}SvBq^{-1}) [11].

The mean concentration of radium in the water is 155,7mBql^{-1}. Annual dose resulting from consumption of such water is $6{,}7 \times 10^{-2}$mSv year^{-1} if we use conversion factor recommended W.H.O. [12](2,2 x 10^{-7}Sv Bq^{-1}) or $6{,}9 \times 10^{-2}$mSv year^{-1} if the dose calculation using the I.A.E.A. ($2{,}8 \times 10^{-7}$Sv Bq^{-1})[13] which is only 2,8% of the average annual dose due to natural background (2,4mSv).

Conclusions

From the results of these measurements, a correlation may be observed between the radon concentration and the geological structure. In most of the cases, the measured values

are not very high and do not exceed the radioprotection standards recommended by national and international institutions (US EPA, 1991). In most of the cases, the measured values are not very high and do not exceed the radioprotection standards recommended by national and international institutions (US EPA, 1991).

According to the results of this study, it is evident that the activity concentration of radon and radium measured in water in the assumed ratio presents no significant risk for the population from study area. The concentrations and the effective doses due to intake of radon and radium from drinking water are compared to the findings of similar works in other parts of the world. The expected doses from consumption of water in Romania are insignificant compared with other natural sources of radiation (such as radon in indoor air, ^{40}K, etc.).

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