AQUAKIT METHOD AND ESTIMATION OF INGESTION RADON DOSE IN THE ALL AGE GROUPS HUMANS

AQUAKIT METODA I PROCJENA INGESTIONE RADON DOZE ZA LJUDE SVIH DOBNIH SKUPINA

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ABSTRACT
Radon is a radioactive, colorless, odorless, tasteless noble gas, occurring naturally as the decay product of radium. It contributes about half the radiation dose received by the general population. The measurements where being done by AlphaGUARD radon portable measuring system. The estimated annual effective dose ranged from 2.26 to 3.5 μSvy^{-1}, 1.73 to 2.6 μSvy^{-1} and 4.7 to 7.2 μSvy^{-1} for adults, children and infants, for drilled and dug water samples collected, respectively. World Health Organisation (WHO) recommended limit for annual effective ingestion dose of 0.1 mSv y^{-1} for public exposure for all ages.

Keywords: radon activity concentration, annual ingestion dose, age groups.

REZIME
Radon je radioaktivni gas, bez boje, mirisa i okusa, koji nastaje radioaktivnim raspadanjem radija. Radon doprinosi polovini od ukupne doze koju prima ljudska populacija. Mjerenja su vršena pomoću AlphaGUARD radon sistema. Procijenjena godišnja efektivna doza za ispitivane uzorke je bila u intervalu (2.26-3.5) μSvg^{-1}, (1.73-2.6) μSvg^{-1} i (4.7-7.2) μSvg^{-1} za odrasle, djecu i dojenčad, respektivno. Svjetska zdravstvena organizacija preporučuje godišnji limit za ingestionu radon dozu od 0.1 mSv, za sve dobne skupine.

Ključne riječi: koncentracija aktivnosti radona, godišnja ingestiona doza, dobne skupine.

1. INTRODUCTION
The noble gas isotope ^{222}Rn (radon) is formed by decay of ^{226}Ra [1]. The major contribution of human exposure to natural radiation comes from radon. Radon is a natural inert radioactive tasteless and odourless gas [2]. As ^{226}Ra is one of the nuclides formed in the disintegration series from ^{238}U, the amount of radon formed in rocks and soils depends on their uranium content. Uranium occurs in contact with basic rocks, in skarn in iron ores and in clay gouge. Uranium is easily leached even at a normal pH of around 5-7, and many of the large uranium ore deposits are formed by ground water that has transported uranium which has been precipitated in a reducing environment [1].
Radon concentrations in soil, air or ground water are determined by the radium concentration in surrounding soil and bedrock, how many of the radon atoms emanate to the pore space

from the mineral grain in which they are formed, and the porosity and water content of the soil. Since the rock porosity is very low, the conditions exist for the radon concentration to be 10-100 times greater than the rock radon concentration. High radon concentrations sometimes also occur in association with pegmatites, as well as gneisses and vulcanites rich in quartz and feldspar. The concentration in ground water from bedrock is low in areas where concentrations in the rocks are low. In terms of transportation it should be noted that radon moves by two basic means, diffusion and forced flow. Diffusion inevitably occurs, even though its extent may be limited. Hence diffusive migration sets a lower limit on the transport of radon. Forced flow depends on pressure gradients, which may or may not be present in a given situation. Radon gas finds its way into indoor air mainly by migrating from bedrock, through the soil and into the home via cracks or other openings in the foundation. Radon from bedrock wells is released into indoor air during showering, dishwashing and doing laundry. The amount of radon released from stone building materials such as a granite block foundations, fireplace materials, counter tops and floor or wall tiles is usually insignificant [1]. Radon is a known human carcinogen and is estimated to be the second leading cause of a lung cancer. Only smoking causes more lung cancer deaths [3]. Radon in water can get into the human body via two distinct pathways-ingestion and inhalation. When someone drinks water that contains radon, a radiation dose is delivered to internal organs. Additionally, radon, as a gas, moves from water to air when the water is being used. When radon gas is inhaled, the radon daughters bind to the lung tissue and deliver a radiation dose to the surrounding tissue. Radon that is ingested in water is thought to create a cancer risk, primarily in the stomach. The dominant health concern with radon, however, is the lung cancer risk created when radon gas escapes water and is inhaled. Ingested radon diffuses into the tissues of the stomach and small intestine. From there it enters the bloodstream and is carried throughout the body. The majority of ingested radon is thought to be exhaled when the blood flow carries it to the lungs [4].Radon is considerably more soluble in water than the lighter noble gases, about 15 times as soluble as helium and neon. Radon is readily absorbed from the gastrointestinal tract and distributed among the tissues, in part because of its relative solubility in blood and in tissue [5].Temperature of drinking water is very important parameter because it affects the development dynamics of many physical, chemical and biochemical processes. In general, increasing the temperature of water also increases the speed of unfolding for some chemical and biochemical reactions, and it decreases the solubility of oxygen and some other gases [6]. In 1991, the United States Environmental Protection Agency (EPA) proposed a National Primary Drinking Water Regulation (NPDWR) for ^{222}Rn with a maximum contaminant level (MCL) of 11 BqL^{-1} .The National Academy of Sciences (NAS) revised the MLC and established an alternative maximum contamination level (AMCL). According to NAS (1999), the AMCL may be set higher than the MCL such that "the contribution of radon from drinking water to radon levels in indoor air is equivalent to the national average concentration in outdoor air". For the United States, this leads to an AMCL of 146 BqL^{-1} [7]. In 2013, the European Union adopted Directive EC2013/51/EURATOM laying down requirements for protecting the health of the general public with regard to radioactive substances in water intended for human consumption [8]. According to this Directive, Member States may set a level for radon that must not be exceeded, having in mind that the level set can exceed 100 Bq L^{-1} but not 1000 Bq L^{-1}.

2. MATERIALS AND METHODS

For the measurement of radon concentration are available many techniques and instruments that can be classified into two main groups: active and passive method. Here is presented one active measurement method based on the ionization chamber and widely used for measurement of radon concentration in air, water, soil gas and building materials. AquaKIT in combination with the AlphaGUARD PQ2000 PRO and AlphaPUMP can determine radon concentration in water samples directly and precisely (Figure 1). The main condition for correctness and exactness of the measurement is qualified sampling. The radon detector of the AlphaGUARD is based on an ionization chamber, designed for measuring radon in air, soil and water. Through optimal geometry of the chamber and intelligent signal evaluation this radon monitor is suitable for continuous monitoring of radon concentrations between $2\text{-}2\cdot10^6$ Bqm^{-3}. AlphaGUARD offers high detection efficiency, a wide measurement range, fast response and permanent, maintenance-free operation with long-term stable collaboration. In addition to the radon concentration in air AlphaGUARD also measures and records simultaneously ambient temperature, relative humidity and atmospheric pressure with integrated sensors. The measured radon concentration is shown in the upper line of display in Bqm^{-3}. The lower line shows the air temperature in ^{o}C, the relative humidity in %rH and the barometric pressure in hPa. Up to a maximum of 32 measuring series can be administrated in the memory of AlphaGUARD.

The PRO model of AlphaGUARD is capable for operating in two alternative modes: diffusion mode with a 10 or 60 minutes measuring cycle and flow mode in a 1 or 10 minutes measuring cycle. The PRO model's processor has twice the capacity of the standard processor providing extended functions, expanded memory and additional analogue and digital inputs [9,10].

AlphaPUMP is a battery operated electronically controlled handy pump for gases. For the tube connection between AlphaPUMP and an AlphaGUARD it has to taken into consideration that the stream of gas is always conducted from the outlet nozzle („OUT“) of the AlphaPUMP to the active adapter of the AlphaGUARD. This array grants that in the ionization chamber of the AlphaGUARD there is always a slight excess pressure [11].

AquaKIT is an optional accessory to the radon monitoring system AlphaGUARD PQ2000 PRO. AquaKIT is a set of accessories, mostly glass components, used for measuring directly radon gas of liquid samples. Radon measurements of water samples by using AquaKIT are precise and correct. In a closed gas cycle radon is expelled from water sample (placed in degassing vessel) by means of AlphaPUMP. The glass vessels of the AquaKIT measuring equipment grant a hermetically sealed enclosure of the radon expelled of the water samples as well as a fast change of samples which prevents incorrect measurements as a consequence of leakages. The security vessel is coupled at the outlet side of the degassing vessel. All water drops shall deposit in it if they have got into the gas cycle during the degassing process. By this the stress of the water vapor is minimized for the radon monitor. The background of empty set-up was measured for a few minutes before every water sample measurement. For measurement was used 100 ml of every water sample. When the sampling is placed the three-way taps of the degassing and security vessel shall be in 3 o'clock position, because of pressure balance. After that the plastic injection with water sample was injected slowly into the degassing vessel. The three-way taps of the degassing and security vessel must be set immediately into the 6 o'clock position for measuring mode. After that the plastic injection of the vertical connecting socket of the degassing vessel was removed and the AlphaPUMP and AlphaGUARD switched on. After ten minutes AlphaPUMP was switched off, but AlphaGUARD remained switched on for another twenty minutes where the radon measurement was continued. After that, the measured water sample was removed of the degassing vessel. The flow rate of the pump was 0.3 $Lmin^{-1}$. The determination of the radon concentration in the water samples is based on the radon concentration indicated on the radon monitor. This value is not the radon concentration in the measured sample because the radon drive out has been diluted by the air within the measurement set-up and a small part of the radon remains diluted in the watery phase.

For determination of radon concentration in water sample, the following equation was used:

$$C_{water} = \frac{C_{air} \cdot \left(\frac{V_{system} - V_{sample}}{V_{sample}} + k \right) - C_0}{1000} \quad ...(1)$$

where, C_{water} - radon concentration in water sample (BqL^{-1}); C_{air} - radon concentration in air after expelling of the radon, indicated by Alpha Guard (Bqm^{-3}); C_0-radon concentration in the measuring equipment before sampling (Bqm^{-3}); V_{system} - interior volume of the measurement equipment, (1122 mL); V_{sample} - volume of the water sample (mL); k-radon diffusion coefficient.

In most cases it is assumed that the radon concentration in the room air is below 100 Bqm^{-3}. For radon concentration in the measuring equipment before sampling ten minutes measuring time is sufficient for removal radon from equipment. The final result is calculated taking into consideration the value of diffusion coefficient, which depends on temperature:

$$k = 0.106 + 0.405\, e^{-0.052\, t} \quad ...(2)$$

where, k - diffusion coefficient of radon; t - temperature of the water (°C).

The diffusion coefficient is dependent on the temperature. With decreasing temperatures the quantity of radon soluble in water increases. Diffusion coefficient raises when the temperature drops. The influence of the radon diffusion coefficient is only low in the temperature range between 10 ºC and 30 ºC. When measurements are performed under normal conditions the value of k=0.26 can be used within the specified temperature range for a mean room temperature of 20 ºC [12]. Based on the results of the measurements radon activity concentration in the water sample the annual effective ingestion dose of radon was calculated, using the equation (3).

$$H_{ing} = C_{water} \cdot D \cdot G \quad ...(3)$$

where, H_{ing} - annual effective ingestion dose for radon, (μSvy^{-1}); C_{water} - radon activity concentration in the water sample, ($mBqL^{-1}$); D – conversion factor from the concentration of ^{222}Radon to the effective dose ($SvBq^{-1}$). The ingested dose conversion factors are 2.3×10^{-8}, 5.9×10^{-9} and 3.5×10^{-9} $SvBq^{-1}$ for infants (<1 y), children (2–7 y) and adults (≥17 y), respectively. G–annual water intakes by infants, children and adults of about 230, 330 and 730 L, respectively [13,14,15].

Figure 1. Measuring equipment for determination of radon activity concentration in water

3. RESULTS AND DISCUSSION

In Bosnia and Herzegovina many people receive their drinking water from private groundwater wells, drilled or dug type. Samples of water were collected from drilled and dug wells from Tuzla area. During the sampling processes it was determined that depth of drilled wells ranged between 6-9 meters in the investigated areas. Table 1 shows the mean values of radon activity concentration in water on the researched location.

Table 1. Mean values of radon activity concentration in investigated water samples

Type of source	C_{water}(m BqL^{-1}) Mean value
Drilled wells	887.2
Dug wells	1354.5

The radon activity concentration in investigated water samples varied from 887.2 $mBqL^{-1}$ to 1354.5 $mBqL^{-1}$ for drilled and dug wells, respectively. These values does not exceed the value of 11.1 BqL^{-1} recommended by Environmental Protection Agency for drinking water and European Commission reference level of 100 BqL^{-1} for radon in drinking water [16,8].

The values of annual effective ingestion radon dose for adults, children and infants, are given in Table 2, respectively. The annual effective dose was calculated on the basis of mean radon activity concentration in water samples for different type of wells. (Table 1).

The annual effective ingestion dose for adults was 2.26 μSvy^{-1} for water samples taken from drilled wells, and 3.5 μSvy^{-1} for dug wells, respectively. The estimated total annual committed effective dose received by the children as a result of ingestion of water was 1.73 μSvy^{-1} for drilled water and 2.6 μSvy^{-1} for dug wells, respectively. The annual effective ingestion dose of infants was 4.7 μSvy^{-1} for drilled wells, and 7.2 μSvy^{-1} for dug wells, respectively.

Table 2. Annual effective ingestion dose for all age groups humans

Type of source	H_{ing} (μSvy^{-1})		
	adults	*children*	*infants*
Drilled wells	2.26	1.73	4.7
Dug wells	3.5	2.6	7.2

The values of estimated annual effective doses are much lower than those obtained in Penang Malaysia, which were 3–48, 1-18 and 2–23 μSvy^{-1} for infants, children and adults, respectively, in the investigated drinking water samples [17]. In comparison with results from Croatia the estimated annual effective doses for adults are much lower than 24.2 μSvy^{-1} and 26.8 μSvy^{-1} that are values of the mean annual effective dose received by adult inhabitants of Osijek-Baranja County and Požega-Slavonia County, respectively [18, 19], in case of consummation of water from private wells. The values of annual effective here presented does not exceed the reference level of 0.1 $mSvy^{-1}$, recommended by the World Health Organization [20].

4. CONCLUSIONS

The values of radon activity concentration obtained in this research are much lower than the action levels of 11.1 BqL^{-1} and 100 BqL^{-1} proposed by United States Environmental Protection Agency and European Commission, respectively. The effective dose received by all age groups was less than standard limit recommended by the World Health Organization of 0.1 $mSvy^{-1}$.

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