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RADON AND RADIUM IN TAP DRINKING WATERS IN THE CITY OF KAVADARCI

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Abstract: Radon and radium could pose serious risk to human health and that is why our preliminary research was concentrated on their concentration in tap drinking waters from the Kavadarci municipality. We sampled three locations within the city limits and there have been determined radon and radium concentrations respectively. Radon concentrations of 0.53 Bq/l; 0.70 Bq/l and 0,87 Bq/l were determined for samples K1, K2 and K3, respectively, which are of 12 to 21 times less than the strictest reference values. Radium values were 0.21 Bq/l; 0.09 Bq/l and 0.20 Bq/l in samples K1, K2 and K3, respectively, which are 5 to 11 times less than usual reference values. Also, total exposure doses due to inhalation and ingestion of radon and ingestion of radium were calculated and their respective values were 4 to 8 times lower than actual allowed reference values.

Key words: radon measurements; water samples; Kavadarci; reference levels; exposure dose

INTRODUCTORY REMARKS

Radioactive elements in nature are present in a wide range of concentrations in all rocks, soils and waters. The presence and distribution of radionuclides in ground water is primarily a matter of local geology and the chemical composition of rocks and waters (Sloto, 2000). Among the heavy radioactive elements, the most common are ^{238}U and ^{232}Th, which produce other radioactive isotopes of radium and radon. Among the isotopes of radium, the most common in water is ^{226}Ra, which is a radioactive element of the ^{238}U decay series, with a period of radioactive half-decay of about 1622 years. Within alpha decay, ^{226}Ra produces radon (^{222}Rn). These radioactive isotopes (^{226}Ra and ^{222}Rn) have different chemical properties, but both can potentially cause health problems, such as cancer, after their intake with water consumption [WHO, 2011].

"Natural killer"

Radon in the true sense of the word is an invisible deadly weapon. It is a gas that is invisible and has no smell, but is radioactive. It is an "inert gas" that does not react with other elements. People can be exposed to radon primarily by breathing radon into the air that comes through cracks and voids in buildings and homes (but also from a variety of building materials built into them), as well as by consuming water with increased concentrations of radon. Because radon comes naturally from earth, humans are continuously exposed to it.

When we inhale radon together with other gases, it does not react but immediately dissolves in the blood and travels through it throughout the body. When it reaches equilibrium, that is, when its concentration is equal to that of the surrounding air, we release it out through the mouth and skin. This process seems completely harmless, unless we take into account that some atoms decompose during this journey through our body, while emitting radiation and creating particles of radioactive metals that accumulate in our bones, organs and adipose tissue.

During its decomposition in air, radon atoms are converted into radioactive metals that do not precipitate but constantly float. When we inhale them, some of them "stick" to the surface of our lungs, where their decay and radiation continue. At each step of the radioactive decay sequence, the decaying atoms emit ionizing radiation that kills or damages living cells – alpha particles (a helium nucleus with 2 protons and 2 neutrons), beta particles (high-velocity electrons), and gamma rays (X-rays with very high energy). Alpha particles are the largest and most deadly to living cells. From a medical point of view, the problem is not the dead cells but the surviving cells with altered DNA. They can multiply like cancer or mutate and damage future generations. Radon kills very slowly, but statistically speaking very confidently. In non-smokers, it is the leading cause of lung cancer, killer cancer no. 1. Radon is the second leading cause of lung cancer after cigarette smoking. The U.S. Environmental Protection Agency and the U.S. Office of Surgery have determined that radon is responsible for more than 21,000 lung cancer deaths each year in the United States (US-EPA, 1999a; 1999b; 2000). If you smoke and live in a home with high levels of radon or consume water with high concentrations of radon, you increase the risk of developing lung cancer, i.e. stomach cancer (IAEA, 2014). The only effective way to determine if you and your family are at risk for high radon exposure is to test your home for air and water.

Where is the radon?

Radon is all around us. This gas is created naturally within the long-term sequence of decay of radioactive heavy metals, uranium and thorium, which, as is commonly known, are scattered throughout the Earth's crust. Radon is also present in ground waters and surface waters that pass through various geological environments (rocks, sediments, soils). Uranium and its decay products, radium and radon, can be found in almost all rocks and soils. Most of them contain only 1–3 ppm uranium, but some such as granites, dark shales, light-colored volcanic rocks, and phosphate sedimentary rocks can contain up to 100 ppm uranium (WHO, 2009; 2018). Thorium, which is even more common in nature, also produces radium and radon. The sequence of decay of uranium and thorium constantly produces radium which then "magically" decomposes into radon gas, i.e. its isotopic varieties ^{222}Rn (radon) and ^{220}Rn (thoron). On average, about two atoms of radon are emitted every second per square centimeter of soil anywhere on Earth, every day. Radon has been around since the creation of planet Earth. Outdoors, everywhere, and indoors in many areas, radon levels are low and the health risk is low. However, in some areas radon can reach very high concentrations outdoors and indoors (Bartram, 2015.). As radioactive heavy metal atoms decompose, they change into lighter and lighter radioactive heavy metals until they end up as non-radioactive lead. Radon is the only gas in the long range of radioactive decay of uranium. Its "parent" (predecessor) is radium while its successor is polonium. The half-life of radium (^{226}Ra) is about 1600 years. The half-life of radon (^{226}Rn) is only 3.82 days, but then the sequence of radioactive decay continues with polonium, bismuth and lead. After 22 years, half of the radon atoms end up as lead (^{226}Pb), a stable non-radioactive element. Of the approximately 20 known isotopes of radon, only two more occur naturally in nature (EURATOM, 2010; 2013). These are thoron (^{220}Rn) and actinon (^{219}Rn). Thoron has a half-life of 55 seconds and is the product of a series of radioactive decays of thorium (^{232}Th). It emits from building materials, such as cement, and can contribute 5% to 20% of the total radon level in homes. Actinon has a half-life of 4 seconds. It is the product of a series of ^{235}U (actinouranium) radioactive decay. The Earth will never run out of radon gas. You will wonder why? Because the radioactive decay of uranium is very slow. The half-life of uranium (^{238}U), when half of its atoms decayed into another element, is 4.5 billion years. As we have said, thorium (^{232}Th) is as common as uranium, and its period of radioactive half-life is even longer 14.1 billion years.

OBJECTIVES FOR MAKING THE MEASUREMENTS AND THE PAPER

We want to emphasize that more than 50% of the effective annual dose of radiation received by a human being is related to radon and its "ancestors", i.e. progeny. Among the main mechanisms that bring radon into habitats is soil evaporation as well as evaporation and release from water [ICRP 60; ICRP, 65; UNSCEAR 1993]. Given the mechanism of evacuation, it was estimated that this mechanism of radon release from water represents about 89% of the risk of cancer, while drinking water with high concentrations of radon is associated with about 11% of the risk of cancer [Oner et al., 2009]. The concentration of radon in water can be subject to various factors such as the geology of the area, the bottom sediments, the influx of streams, the temperature, the atmospheric pressure, etc. In this regard, it is important to emphasize that the solubility of radon in water is about 510 cm^3 kg^{-1} at 0ºC and decreases at higher temperatures [Corrêa, 2006]. Surface and ground waters contain radionuclides as natural components in different concentrations depending on their origin. Radon is released into water as a result of natural processes, such as the decay of its nuclide "parent" ^{226}Ra, while dissolution from the surrounding geological environment (rocks, soils) is predominant, as highlighted by Moreno et al. (2014); Fonollosa et al. (2016). Radon in water can also originate from the dissolution of radon from air into water, as well as mixing with other waters from the catchment area, with increased concentrations of radon.

As for radium (^{226}Ra), its concentration in water depends on the content of ^{226}Ra in rocks and soil, the permeability of the soil and the solubility of uranium and radium compounds in water. Typically, surface waters have lower concentrations of radium (^{226}Ra) than ground waters [Somalai et al, 2002]. Radium (^{226}Ra) is considered to be the second largest source of radiation in water [WHO, 2005, 2008, 2009; US-EPA, 2000]. Several agencies set concentration limits for total water activity to control radium exposure levels. The World Health Organization (WHO) in 2011 sets a limit of 0.5 Bq/l for gross alpha radiation for safe water consumption when no decision should be made to reduce or reduce the dose [WHO, 2011]. The US Environmental Protection Agency (US-EPA) considers bone cancer and various types of soft tissue cancer to be the main health effects associated with radium intake (^{226}Ra). It is chemically similar to calcium and is absorbed by plant from the soil, which in turn transfers it to the human food chain. Due to its calcium-like chemical behavior, it is mainly distributed in the bones of the human skeleton and its amount is greater when the individual is in the process of growth. Therefore, it significantly contributes to the dose absorbed into children's tissue [US-EPA, 2000].

Pursuant to Article 35 and Article 36 of the EURATOM Agreement (EURATOM, 2010), monitoring and reporting on environmental radioactivity is an obligation for EU Member States. Drinking water should be analyzed for radon content in accordance with the new EURATOM directive for drinking water called E-DWD (EURATOM, 2013). To protect the health of citizens from radon in drinking water, different levels of radon concentration are introduced. For water intended for human consumption, E-DWD establishes parametric values, WHO (WHO, 2008) uses level guidelines, while maximum concentration levels are introduced in the United States. As explained in the publications of the E-DWD and the World Health Organization (WHO), guidance levels and parameter values should not be considered as limit or reference values. The country concerned determines guidance levels and parametric values based on whether that value poses a risk to human health in terms of radiation protection or not (i.e. if additional remedial action is required).

The guidelines and parametric levels have been reviewed for all Member States and are summarized in Table 1. For water intended for human consumption in European Union countries, the guidelines and parametric levels are in accordance with the E-DWD, i.e. between 100 and 1000 Bq/l.

In the Republic of Northern Macedonia, according to Article 20 and Article 22 of the Rulebook on limits for exposure to ionizing radiation and the conditions of exposure in special cases and extraordinary events (Official Gazette of RM, no. 29/2010), the action level of the radon per unit volume of drinking water from the public water supply system should be 1000 Bq/l.

Table 1

International guidelines and parametric values of radon in drinking water (Bq/l)

Directive / Recommendation	Activity concentration	Reference
EURATOM DWD (E-DWD)	100-1000	EURATOM, 2013
24 EU member states*	100	National law of the member state
Ireland, Portugal, Spain	500	National law of the member state
Finland	1000	National law of the member state
Guidance level according to WHO	100	WHO, 2008
Maximum level of contamination according to US-EPA	~11.1	US-EPA, 1999
Alternative maximum level of contamination according to US-EPA	148	US-EPA, 1999

Note: * Austria, Belgium, Bulgaria, Croatia, Cyprus, Czech Republic, Denmark, Estonia, France, Germany, Greece, Hungary, Italy, Latvia, Lithuania, Netherlands, Poland, Romania, Slovakia, Slovenia, Sweden, United Kingdom (until recently – before the Brexit)

In the United States, two different levels are given for the maximum level of concentration (US-EPA, 1999a). Exceeding the highest alternative concentration level may result in increased health risks of indoor radon (i.e. radon disappearing from water and indoor air). Higher concentration levels can contribute approximately one tenth (14.8 Bq m^{-3}) of total indoor radon, which is equivalent to the average U.S. radon concentration outdoors (Bartram, 2015). The lower, stricter, concentration level defined by the US-EPA is approximately 11.1 Bq/l (Table 1). The World Health Organization has set water levels up to 100 Bq/l in the third edition of the WHO Guidelines for Drinking Water (WHO, 2008).

INSTRUMENTS AND METHODS USED IN MEASUREMENTS OF THE ^{222}RN AND ^{226}RA IN WATER

The measurement of the presence of radon (Rn^{222}) and thoron (Rn^{220}) in the air at positions of interest was performed using the AlphaGUARD DF2000 professional radon monitor for multi-parametric analysis with gas impermeable chamber for pulsating ionization (0.6 l). The radon measurement range is from min 2 to max 2 000 000 Bq/m^3 Rn^{222}. Radon sensitivity is: 1 cpm at 20 Bq/m^3 (0.5 pCi/l). Sensitivity for radon determination in relation to toron: radon minimum 1 cpm at 60 Bq/m^3 (1.6 pCi//l); thoron (1 l·min^{-1}) minimum 1 cpm at 200 Bq·m^{-3} (5.5 pCi·l^{-1}) and thoron (2 l·min^{-1}) minimum 1 cpm at 140 Bq/m^3 (3.8 pC/l).

The measurement of the presence of radon (Rn^{222}) and radium (Ra^{226}) in water samples from localities of interest to the client was performed using the AlphaGUARD DF2000 professional radon monitor for multi-parametric analysis with gas impermeable chamber for pulsed ionization (0.6 l) which for these specific measurements worked in conjunction with portable equipment, for direct determination of radon concentrations and indirect determination of radium in water samples, AquaKIT (2 vessels for rinsing / releasing radon gas, respectively, 2·100 ml). The radon measurement range is from min 2 to max 2 000 000 Bq/m^3 Rn^{222}. Radon sensitivity is: 1 cpm at 20 Bq/m^3 (0.5 pCi//l).

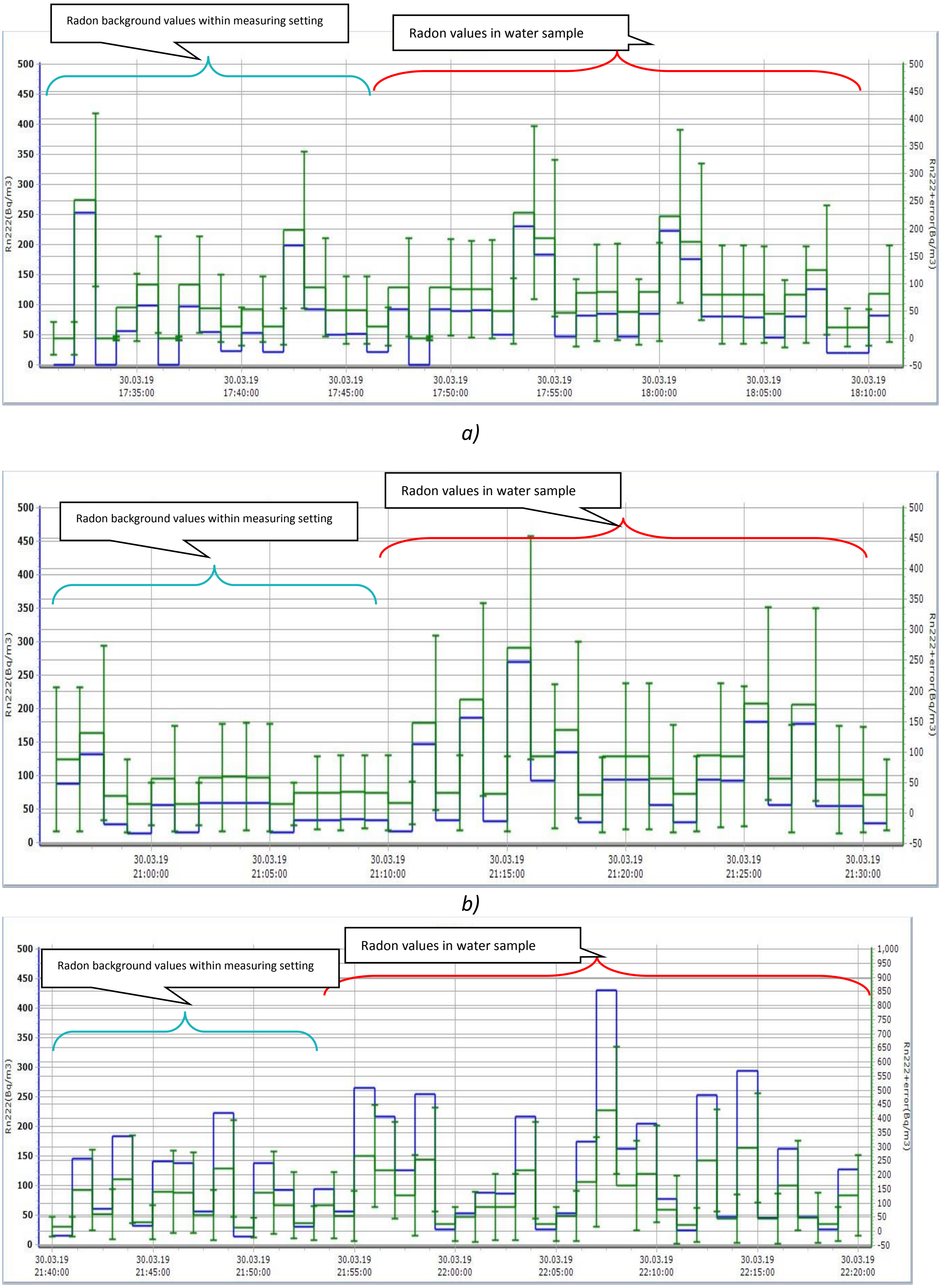
Emanometry is based on aeration (degassing) of the sample followed by detection of alpha particles by the ionization chamber. When the water sample is degassed, ^{222}Rn is transferred to the measuring cell (ionization chamber) by air flow / circulation in the system. The technique is sensitive to water temperature, as it can affect the level of degassing during transport (ISO 13164-3, 2013). Detector contamination is checked to see if radon analysis at low concentrations is possible. One approach to degassing is to place a sample of water in a degassing cell and to introduce radon-free (or very small concentrations) air into a closed system. In this way, the radon is released from the sample and with the help of a pump is transferred to the ionization chamber, i.e. the detector. In the ionization chamber the concentration of radon activity in the air is analyzed, but the initial concentration of radon in water can be calculated based on this measurement.

$$C_{water} = \frac{C_{Air}\ x\ \left[\frac{V_{system} - V_{sample}}{V_{sample}} + k\right] - C_o\ x\ \left[\frac{V_{system} - V_{sample}}{V_{sample}}\right]}{1000}$$

C_{water} – radon concentration in the water sample (Bq/l),
C_{air}- – concentration of radon (Bq·m^{-3}) in the measuring system after aeration of radon from water,
C_0 – concentration of radon (Bq·m^{-3}) in the measurement system before the start of measurement ("zero level"),
V_{system} – internal volume of the measuring system (ml)
V_{sample} – water sample volume (ml)
k – water / air distribution coefficient

RESULTS OF RADON AND RADIUM MEASUREMENTS IN WATER AND DISCUSSION

Emanometric measurements of water samples were followed by detection of alpha particles by the ionization chamber. When the water sample is degassed, ^{222}Rn is transferred to the measuring cell (ionization chamber) by air flow / circulation in the system. As pointed out, the technique is sensitive to water temperature, as it may affect the level of degassing during transport (ISO 13164-3, 2013).



a)

b)

c)

Fig. 1. Diagrams for the course of radon measurements in tap water supply network in the city of Kavadarci; a) sample K1; b) sample K2 and c) sample K3

The results of the measurements of the concentration of radon (in Bq/m^3) in the air during the aeration / degassing of drinking water from the taps in the water supply network of the city of Kavadarci is given in Table 2.

Table 2

Measurements of the radon concentration in drinking water from the taps in the water supply network of the city of Kavadarci

Sample	N (number of measurements)	Min$_{Air}$ (Bq/m^3)	Max$_{Air}$ (Bq/m^3)	Average$_{Air}$ (Bq/m^3)	Median$_{Air}$ (Bq/m^3)	Water ^{222}Rn (Bq/l)
K1	22	19.37	229.62	94.91	81.91	0.53
K2	22	15.96	270.36	90.42	74.38	0.70
K3	28	24.18	429.94	130.92	90.78	0.87
				average	K1-K3	0.70

The diagram given in Figure 1 graphically shows the course of radon measurements, both in the initial phase when the background values of radon within the measuring system setting are determined, and in the second phase of determining the radon concentrations in the degassed (aerated) sample (samples K1, K2 and K3) of water.

Based on the measurements and calculations (Table 1 and Figure 1), we can conclude that the concentration of radon in the measured samples of drinking water from the taps in the water supply network of the city of Kavadarci have values like those given in Table 3.

For a more illustrative view, the concentration of radon in the drinking water from the taps in the water supply network of the city of Kavadarci is given with a diagram, where the position of the radon concentrations in the measured samples is given very vividly compared to the national and world action/reference levels. (Figure 2).

Table 3

Concentration of radon in drinking water from taps in the water supply network of the city of Kavadarci (Bq l^{-1})

Sample	Measured value	MKD action level	WHO	US-EPA
K1	0.53	1000	100	11.1
K2	0.70	1000	100	11.1
K3	0.87	1000	100	11.1

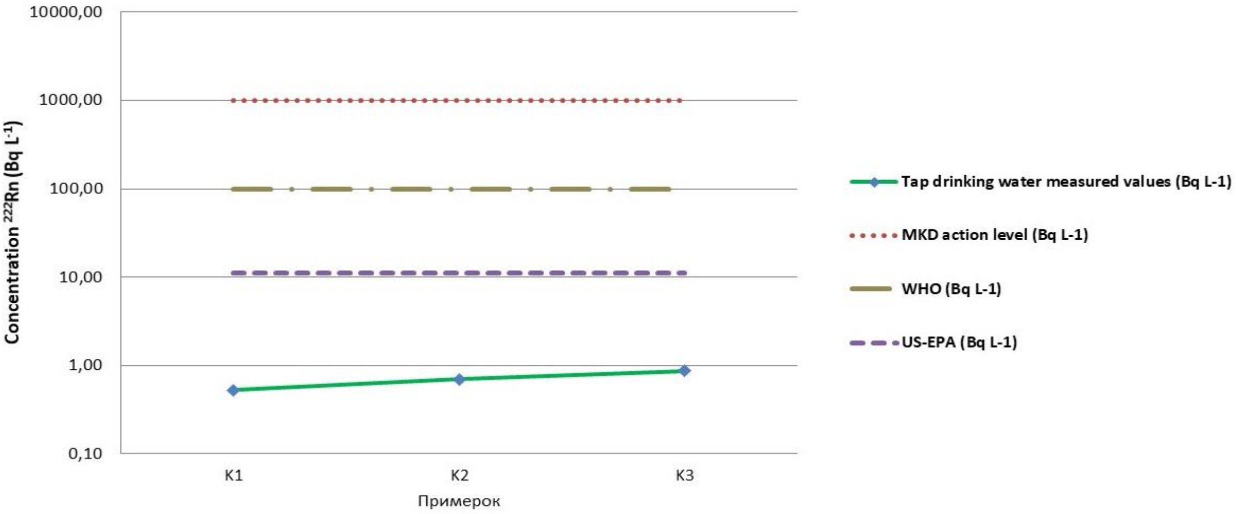


Fig. 2. Diagram of radon concentration in drinking water from the taps in the water supply network of the city of Kavadarci in comparison with the reference values, Macedonian action level (MKD), the World Health Organization (WHO-World Health Organization) and the United States Environmental Protection Agency (US-EPA)

As can be seen from Table 2 and Figure 2, the concentrations of radon in all waters (K1; K2 and K3) in the water supply network of the city of Kavadarci are below the maximum allowed values according to the MKD action level and the values recommended by the World Health Organization (WHO, 2009). However, even more important is the fact that the concentrations of radon in all these samples (K1, K2 and K3), which go directly to consumers and are consumed, are below the strictest, maximum allowed values, by the United States Environmental Protection Agency (US-EPA, 2000).

In terms of radium (^{226}Ra) and its measurements, we first want to highlight a few important facts. Radium 226 (^{226}Ra) and its products of radioactive half-life / decay products are responsible for much of the internal dose that humans receive from natural radionuclides. It has long been known in the scientific community that many mineral springs contain relatively high concentrations of radium and radon. Radium (^{226}Ra) is generally a direct precursor to radon (^{222}Rn), and is in secular equilibrium with it. It has been found that λ_{Ra} and λ_{Rn} (radioactive decay constants of radium and radon) correspond to the number of radium and radon atoms N_{Ra} and N_{Rn} (Shivakumara et al., 2014). In the case of secular equilibrium, during t << T1/2 (Ra), where T1/2 (Ra) = 1620 years, the rate of radium disintegration is actually constant, so it can be roughly said that $e^{-\lambda_{Rn}t} \approx 1$, which means $N_{Ra} = N_{Ra}$ (0) and the number of radon atoms is given by the equation:

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

In addition, even if the condition $t \geq$ T1/2 (Rn) is met, where T1/2 (Rn) = 3.82 days, then $e^{-\lambda_{Rn}t} \approx 0$, leading to equation:

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

Or that $\lambda_{Rn} \cdot N_{Rn} = \lambda_{Ra} \cdot N_{Ra}$, which actually means that the activities of the parent (^{226}Ra) and the "daughter/product" (^{222}Rn) become equal. This in practice means that the concentration of radon is equal to the concentration of radium, this occurs after a period of 30 days when radium can be considered in secular equilibrium with radon. Duplicates of the original samples, in which the concentration of radon in the drinking water from the taps in the water supply network of the city of Kavadarci was measured, were taken and stored in hermetically sealed bottles for 30 days in order to eliminate the radon diluted in the sample. At the same time, after 30 days, the radon that is still present in the water sample (and is the subject of analysis) is directly related to the dissolved radium in it.

After 30 days when the water sample is stored in an airtight container, the radon reaches the secular equilibrium and its measured activity can be assessed as the activity of the dissolved radium concentration. In all the used calculations we will refer to the measurement of the radon concentration, which is actually the concentration of the radium activity, in this case, i.e. the measurements after 30 days from taking the initial water samples from the subject locality. The results of the radium measurements in the water samples gave the values as shown in Table 4.

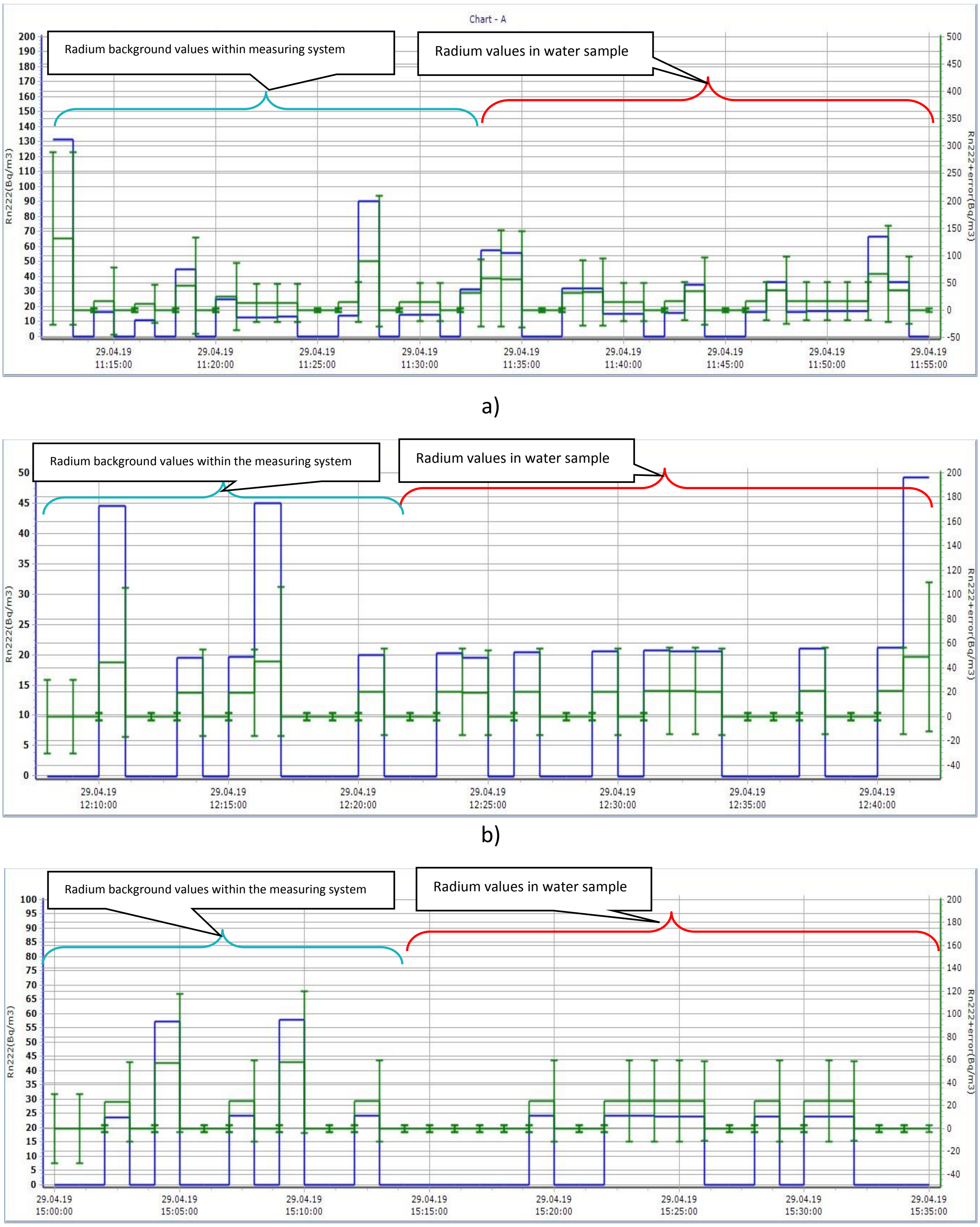
Table 4

Concentration of radium in drinking water from taps in the water supply network of the city of Kavadarci

Sample	N (number of measurements	Min_{Air} (Bq/m^3)	Max_{Air} (Bq/m^3)	$\text{Average}_{\text{Air}}$ (Bq/m^3)	$\text{Median}_{\text{Air}}$ (Bq/m^3)	Water ^{226}Ra (Bq/l)
K1	15	14.96	66.58	28.19	16.81	0.21
K2	10	19.55	49.29	23.41	20.61	0.09
K3	7	23.79	24.26	23.98	23.92	0.20
				average	K1–K3	0.17

The diagram given in Figure 3 graphically shows the course of radium measurements, both in the initial phase when the background values of radium in the system are determined, and in the second phase of determining the radium concentrations in the degassed (aerated) sample (samples K1, K2 and K3) of water.

Based on the measurements and calculations (Table 4 and Figure 3), we can conclude that the concentration of radium in the analyzed samples of drinking water from the taps in the water supply network of the city of Kavadarci have values such as those given in Table 5:



a)

b)

c)

Fig. 3. Diagrams for the course of radium measurements in tap water from the water supply network in the city of Kavadarci:; a) sample K1; b) sample K2 and c) sample K3

Table 5

Concentration of radium in drinking water from taps in the water supply network of the city of Kavadarci in comparison with the reference values of the World Health Organization existing and newly proposed

Sample	Measured value ^{226}Ra (Bq/l)	WHO$_{\text{existing}}$ ^{226}Ra (Bq/l)	WHO$_{\text{suggested}}$ ^{226}Ra (Bq/l)
K1	0.21	1	0.5
K2	0.03	1	0.5
K3	0.20	1	0.5

For a more illustrative view, the concentration of radium in the analyzed samples of drinking water from the taps in the water supply network of the city of Kavadarci is given with a diagram, where the position of the concentrations of radium in the measured samples is given in comparison with the world referent levels (Figure 4).

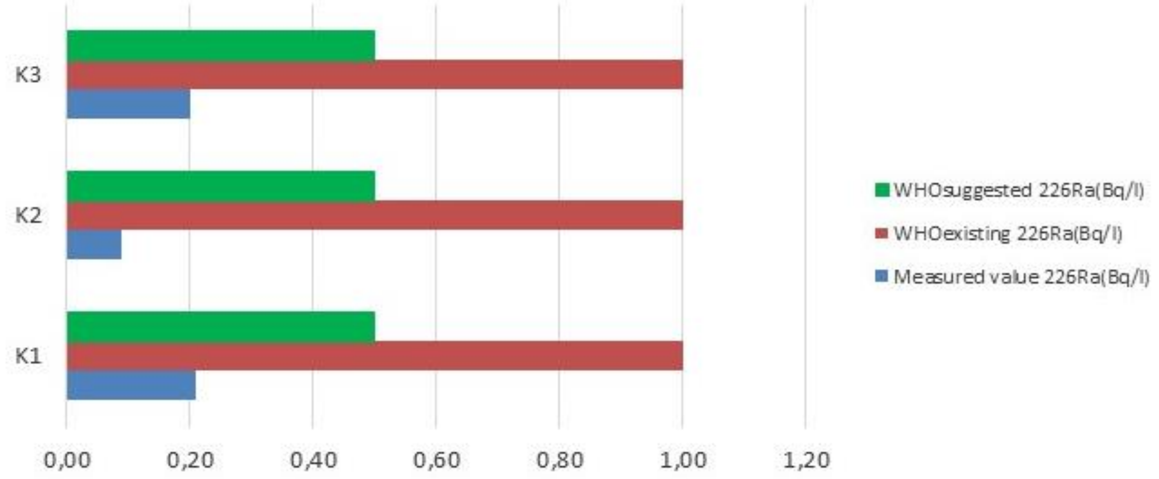


Fig. 4. The measured values of ^{226}Ra in the drinking water from the taps in the water supply network of the city of Kavadarci, compared with the reference values given by the World Health Organization (WHO)

None of the analyzed samples of drinking water from taps in the city of Kavadarci, the reference value for radium given by the World Health Organization (WHO) in the amount of 1 Bq/l that is, the newly proposed value of 0.5 Bq/l was not exceeded.

Going one step further, we calculated the exposure to radon inhalation and ingestion of radon and radium (Table 6).

In this regard, we want to emphasize that the parameters for calculating the exposure during inhalation were the concentration of radon (^{222}Rn) in water, the ratio of air to water concentration of 10^{-4}, indoor residence time (7000 h per year, equilibrium factor 0.4, as well as the exposure dose conversion ratio for inhalation of 9 μSv (Bq h/m^3)$^{-1}$.The effective ingestion dose mainly depends on the amount of water consumed by the consumer in 1 (one) day ionization doses (exposure dose) on inhalation and ingestion are calculated according to the formulas given below (UNSCEAR, 2000; 2006).

$$\text{Inhalation dose } (\mu\text{Sv}) = {}^{222}\text{Rn}_{\text{conc}}\ (\text{Bq l}^{-1})\cdot 10^{-4}\cdot 7000\text{ h}\cdot 0.4\cdot 9\ \mu\text{Sv (Bq h m}^{-3})^{-1}$$

$$\text{Ingestion dose } (\mu\text{Sv}) = {}^{222}\text{Rn}_{\text{conc}}\ (\text{Bq l}^{-1})\cdot 365\text{ l y}^{-1}\cdot 3.5\ \mu\text{Sv Bq}^{-1}\cdot 10^{-3}$$

$$\text{Ingestion dose } (\mu\text{Sv}) = {}^{226}\text{Rn}_{\text{conc}}\ (\text{Bq l}^{-1})\cdot 365\text{ l y}^{-1}\cdot 0.28\ \mu\text{Sv Bq}^{-1}\cdot 10^{-3}$$

Table 6

Calculated values for exposure to radon inhalation and ingestion of radon and ingestion of radium in drinking water from taps in the water supply network of the city of Kavadarci

Sampling location	K1	K2	K3
Number of measurements	22+15	22+10	28+7
pH	7	6.9	6.1
EC (μSv cm^{-1})	120	100	100
^{222}Rn (Bg l^{-1})	0.53	0.7	0.87
^{226}Ra (mBg l^{-1})	210.00	90.00	200.00
Inhalation dose ^{222}Rn (μSv y^{-1})	1.34	1.76	2.19
Ingestion dose ^{222}Rn (μSv y^{-1})	0.68	0.89	1.11
Ingestion dose ^{226}Ra (μSv y^{-1})	21.46	9.20	20.44
TOTAL dose (μSv y^{-1})	23.47	11.86	23.74
TOTAL dose (mSv y^{-1})	0.023474675	0.01185625	0.023743825
TOTAL ingestion dose (μSv y^{-1})	0.022139075	0.01009225	0.021551425
Inhalation and ingestion dose ^{222}Rn (μSv y^{-1})	2.01	2.66	3.30

The radon dose (^{222}Rn) is divided into two parts, namely the ingestion dose and the inhalation dose. For ingestion and inhalation, ^{222}Rn and its progenitors in water give a dose of radiation to the stomach and lungs, respectively. Based on the obtained radon concentrations in the waters subject to this monitoring, it was concluded that the inhalation dose in sample K1 was 0.53 μSv y^{-1}, sample K2 0.70 μSv y^{-1} and sample K3 0.87 μSv y^{-1}. Swallowing dose calculations, depending on the radon concentration, showed that in sample K1 it was 0.68 μSv y^{-1}, sample K2 0.89 μSv y^{-1} and sample K3 that value was 1.11 μSv y^{-1}. The calculated combined inhalation and ingestion doses based on the measured radon concentrations in the water in question are: for sample K1 2.01 μSv y^{-1}, sample K2 2.66 μSv y^{-1} and sample K3 3.30 μSv y^{-1}. Regarding the obtained values for the radium during the analysis of the subject waters and based on them the calculated doses during swallowing, we emphasize that in the sample K1 it is 21.46 μSv y^{-1}, in the sample K2 is 9.20 μSv y^{-1} and the sample is K3 31.68 .Sv y $^{-1}$.

Of course, the absolute and perhaps most relevant values in the calculations of this type were the absolute values of the total exposure when inhaling radon and swallowing radon and radium (Table 6). These absolute values for the specific water samples were in sample K1 23.47 μSv y^{-1}, sample K2 11.86 vSv y^{-1} and sample K3 23.74 μSv y^{-1}. The absolute values of the total exposure to radon inhalation and ingestion of radon and radium are given in the diagram below for greater illustration (Figure 5), where we compare them with the most relevant reference values (100 μSv y^{-1}) given by the World Health Organization (WHO).

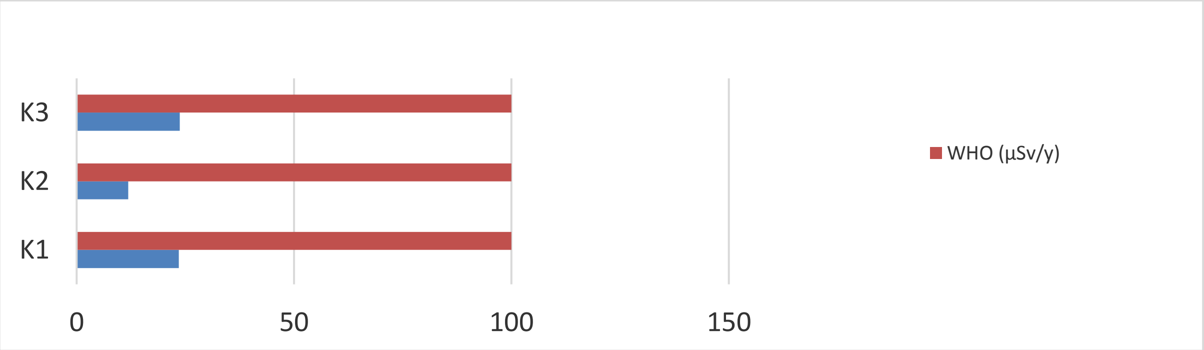


Fig. 5. Calculated values of the total dose of exposure during inhalation and drinking/consumption (^{222}Rn + ^{226}Ra) in the samples of drinking water from the taps in the water supply network of the city of Kavadarci, compared with the reference values given by the World Health Organization

As can be vividly seen from the diagram above (Figure 5), all samples of analyzed drinking water from the water supply system in the city of Kavadarci, which go to the final consumers, have a total dose of exposure during inhalation and drinking/consumption lower for slightly more than four times the value of the maximum recommended reference values given by the World Health Organization (WHO).

CONCLUSION

Our radiological (radon and radium) study of tap drinking water network in the city of Kavadarci showed excellent results in comparison the recommended reference values. Namely, ^{222}Rn values ranged from 0.53 to 0.87 Bq/l averaging 0.70 Bq/l, which is of several magnitudes bellow the strictest reference values of 11.1 Bq/l given by the US-EPA. Also, similar were findings in regards to ^{226}Ra which values ranged from 0.09 to 0.21 Bq/l averaging 0.17 Bq/l. Those values were at least five times lower than the actual reference values of 1 Bq/l given by the WHO, as well as lower than newly suggested values of 0.5 Bq/l by WHO. Calculations of exposure to radon and radium due to radon inhalation (1.34 – 2.19 μSv y^{-1}) and ingestion (0.68–1.11 μSv y^{-1}) and radium ingestion (9.20–21.46 μSv y^{-1}) totaled values in range of 11.86–23.74 μSv y^{-1}, which is just a fraction compared to the reference values of 100 μSv y^{-1} recommended by the WHO. These results proved that the tap drinking water in the city of Kavadarci is safe for consumption for the city habitants in regards to radon and radium. However, we would like to suggest that regular monitoring in suitable time frame should be established.

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Резиме

РАДОН И РАДИУМ ВО ВОДИТЕ ЗА ПИЕЊЕ ВО ГРАДОТ КАВАДАРЦИ

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Клучни зборови: мерење на радон; примероци на вода; Кавадарци; референтни нивоа; доза на изложеност

Радонот и радиумот можат да претставуваат сериозен ризик за човековото здравје и затоа нашето прелиминарно истражување е насочено кон нивните концентрации во водите за пиење во општина Кавадарци. Испитавме три локации од потесното градско подрачје и определивме соодветни концентрации на радон и радиум во вода. Во примероците во вода. K1, K2 и K3 концентрациите на радон изнесуваа 0,53 Bq/l; 0,70 Bq/l и 0,87 Bq/l, соодветно, што е 12 до 21 пат помалку и од најстрогите референтни вредности. Вредностите на радиумот изнесуваа 0,21 Bq/l; 0,09 Bq/l и 0,20 Bq/l во примероците K1, K2 и K3, соодветно, што е 5 до 11 пати пониско од вообичаените референтни вредности. Исто така беа одредени вкупни дози на изложеност поради вдишување и голтање на радонот и голтање на радиумот, а нивните соодветни вредности беа 4 до 8 пати пониски од актуелните дозволени референтни вредности.