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ACTIVITY OF ^{222}Rn IN TAP WATER IN KIELCE COUNTY

AKTYWNOŚĆ ^{222}Rn W WODACH WODOCIĄGOWYCH W POWIECIE KIELECKIM

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Abstract

Radon is known as a radioactive element that dissolves easily in water. It is worth mentioning that it is available in all possible reservoirs. Its concentration cannot be measured directly but only from the emitted radiation. Investigations of ^{222}Rn activity in water in the Kielce district were subjected to three selected water intakes: Bolechowice, Kołomań and Wola Kopcowa. This type of research was conducted for the first time in the discussed area. The results were analyzed in detail in terms of acceptable concentrations. Next, it was determined whether the geological location of the intakes in question may have an impact on the amount of radon present in water from the water supply network.

Keywords: radon concentration, water quality, geological structure

Streszczenie

Radon znany jest jako pierwiastek promieniotwórczy, który łatwo rozpuszcza się w wodzie. Warto zaznaczyć, że dostępny jest we wszystkich możliwych zbiornikach. Jego stężenia nie da się zmierzyć bezpośrednio, a jedynie na podstawie emitowanego promieniowania. Badaniom aktywności ^{222}Rn w wodzie w powiecie kieleckim zostały poddane trzy wybrane ujęcia wód wodociągowych: Bolechowice, Kołomań oraz Wola Kopcowa. Tego typu badania prowadzone są po raz pierwszy na omawianym terenie. Wyniki zostały poddane szczegółowej analizie, m.in. pod kątem dopuszczalnych stężeń. Następnie ustalono, czy położenie geologiczne omawianych ujęć może mieć wpływ na ilość pojawiającego się w wodach sieci wodociągowych radonu.

Słowa kluczowe: stężenie radonu, jakość wody, struktura geologiczna

1. INTRODUCTION

Uranium, thorium and radium are always present in rocks and soils in smaller or larger amounts. During the emanation process radon produced by the decay of radium can enter the pore spaces of rocks and soils. Radon is a radioactive gas that occurs naturally in nature. A characteristic feature of this element is its invisibility (it cannot be seen or smelled). It is found in the soil from where it constantly gets into the atmosphere. Studies prove that it is present in various amounts in households in the air and also in water [1]. Increased amounts of radon are observed mainly in areas where shale and granite rocks are found. As a noble gas, radon has a low binding capacity with solids, which is one of the reasons why it comes out of rocks. If we are dealing with places where rocks are predominant in the landscape, radon will get from them into the air and also into groundwater. So one should be aware of radon hazard in any place that is mountainous [2, 3].

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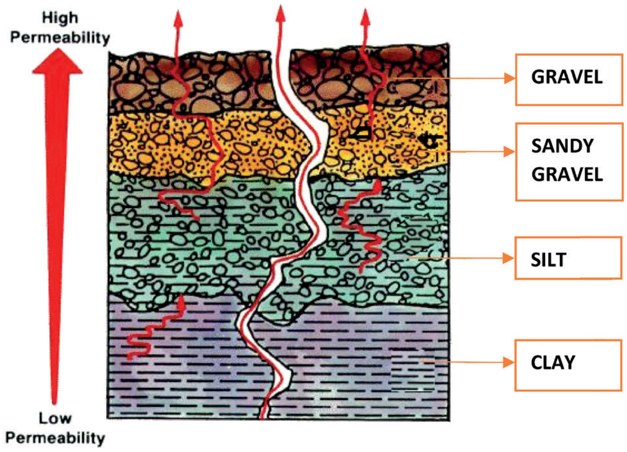

Fig. 1. Permeability of different geological formations [4]

Figure 1 shows the permeability of various geological formations. The ease and efficiency with which radon moves through pore space, including faults, exterminate how much radon will get into, for example, buildings, and tap water. The rate at which radon will penetrate soils depends on the soil's moisture content (how much water is contained in the pore space), porosity, and permeability (the soil's ability to transport water and air).

Radon is formed by the decay of uranium ^{238}U and thorium ^{232}Th. The geological structure and the concentration of uranium and thorium in the rock media are the main factors influencing radon emissions. The main carriers of uranium and thorium in magmatic rocks are accessory minerals such as monazite and xenotime. These minerals are resistant to weathering, hence they sometimes form quite rich accumulations in beach sandstones and scatters. The content of radioactive elements in rocks and the influence of tectonics on the possibility of their migration or penetration are the 2 main factors influencing the amount of radon emission.

Radon moves more slowly in water than in air. Up to the moment of decay, radon penetrates no more than about 2.5 cm in moist rocks or soils and up to about 180 cm in dry rocks. It is worth noting that water moves more slowly through pores in soil and faults in rocks, so radon travels shorter distances in moist geological formations before decaying [5].

The studies are aimed at determining whether water supplied to apartments in the in the Kielce district may be a source of radon hazard.

Additionally, the content of various substances indicating the quality of water was analysed, i.e. pH, conductivity, total hardness, chlorides, nitrites, nitrates, ammonium ions, phosphates, oxidisability and iron.

2. GEOLOGICAL STRUCTURE OF SELECTED SETTLEMENTS IN KIELCE COUNTY

Geologic structure is primarily the type of rocks, their age, and their mutual position in the earth's crust. Bolechowice, as shown in Figure 3, is mainly the remains of the Cretaceous (carbonate-siliceous rocks) and Jurassic, where the most calcareous rocks were formed. Kolomań is Wola Kopcowa is a mixture of Devonian (conglomerates and sandstones) and Silurian (limestone rocks, which today contain animal fossils) [6]. In the case of the Bolechowice and Wola Kopcowa intakes, the Middle Devonian is the geological layer from which water is drawn, whereas in the case of the Kołomań intake, it is the Triassic.

The villages of Bolechowice, Kołomań and Wola Kopcowa are above all the Świętokrzyskie Mountains, whose geological substrate is very rich. Their history covers 540 million years. It is worth noting that they have not always been mountains or land, and additionally it can be added that their location on the globe was not identical to the present one either. The oldest rocks found in the area of Świetokrzyskie Mountains are Cambrian rocks, available in many places, e.g. in Łysogóry. Ordovician and Silurian rocks are found mainly in Mycza, but also in various points of northern Kielce. Devonian for the Świętokrzyskie Mountains range was originally a terrestrial period. Silurian greywackes and the Lower Devonian conglomerates covering them are still visible, for example, in the Prągowiec Gorge. The upper part of the Devonian is again the sea (it reaches the maximum depth in the Carboniferous) where coral reefs developed. Rocks from this period include the Kadzielnia hills. Carboniferous movements are also called Hercynian or Variscan. The Świętokrzyskie region becomes land for millions of years.

It is worth noting that the current shape of the Świętokrzyskie Mountains is mainly the remains of movements during this period. The Permian, Triassic, Jurassic and Cretaceous rocks have unconformable positions on the folded rocks which can be seen in the quarry in Zagnańsk [7, 8]. Figure 2 shows angular and structural unconformities (Lower Triassic rocks lie on Middle Devonian dolomites).

Then we observe transgressions and regressions of the sea in the Upper Permian, Middle Triassic, Middle and Upper Jurassic, and the Upper Cretaceous leaving the Permian-Mesozoic margin of the Świętokrzyskie Mountains, complexes of weathering-resistant rocks, red sandstones of the Lower Triassic, and fossil dunes that can be seen in Tumlin.

Fig. 2. Quarry near Zagnańsk [9]

The sea in the area in question recedes with the end of the Jurassic, the Mesozoic era is mainly block movements. Paleogene and Neogene are the processes of weathering and denudation leading to the formation of the morphological equilibrium surface which becomes the starting point for the modern sculpture. The developing karst phenomena lead to the creation of caves with very phenomenal dripstone decoration. The best known is the Paradise Cave.

The Quaternary is the time of glaciations. On the ridges of hills there is strong mechanical weathering of exposed Paleozoic and Mesozoic rocks. Leading to the formation of the so-called gołoborza. A place very characteristic of the Świętokrzyskie Mountains [10, 11].

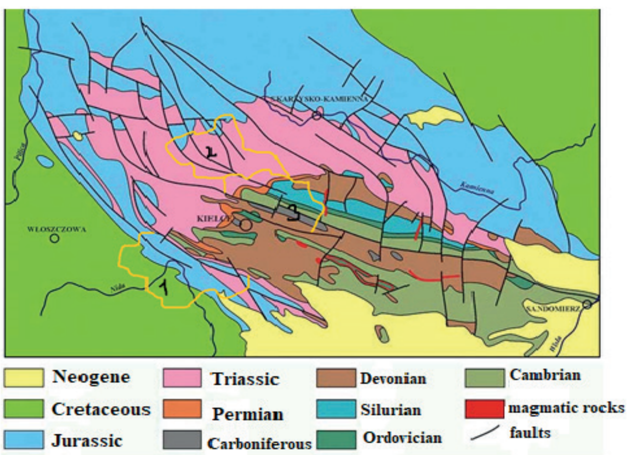

Fig. 3. Geological structure of the Świętokrzyskie Province with Bolechowice, Kołomań and Wola Kopcowa as particular locations [12]

The above figure shows the geological structure of the Świętokrzyskie province where Jurassic and Cretaceous remains are predominant. The surveyed intakes are presented numerically on the figure, where one is Bolechowice (dominance of Jurassic and Cretaceous); two is Kołomań (dominance of Triassic); three is Wola Kopcowa (mixture of Jurassic, Cambrian and Carboniferous, among others). Numerous faults appear in the studied areas, which translates into the movement of radon.

Table 1. Concentration of uranium and radium in rocks [13]

Rock type	Uranium [g/t]	^{226}Ra [Bq/kg]
Sands	0.45	1-27
Clay	1.8	77-124.1
Limestones	2.2	27.8
Granites	3	59.2
Bazslts	1	11.4
Phosphates	100-200 max 650	490

Table 2. Water quality parameters [14]

Water quality parameters	Permissible values
pH	6.5-9.5
Conductivity	2500 μS/cm
Total hardness	60-500 mg/dm^3
Chlorines	250 mg/dm^3
Phosphates	<5 mg/dm^3
Ammonium ion	0.50 mg/dm^3
Nitrates	50 mg/dm^3
Nitrites	0.5 mg/dm^3

2.1. Bolechowice

Bolechowice is located in the commune of Nowiny. This area is located within the south-western fragment of the Świętokrzyskie anticlinorium. Two main structural elements can be distinguished here: the Paleozoic core and its Permian-Mesozoic margin. As far as the Paleozoic structure is concerned, it has a folded structure and is strongly dislocated. The Branchzicko-Bolechowicko-Borkowska syncline consists of carbonate rocks of the Middle Devonian (limestone, dolomite). The core of the syncline is filled with rocks of the Upper Devonian (limestone, shale); Carboniferous rocks (claystones, shales, siltstones) and Permian (conglomerates, sandstones, siltstones, limestones). The syncline floor itself is mainly folded Lower Cambrian rocks (siltstones, sandstones, mudstones) [15].

2.2. Kołomań

Kołomań is located in the Zagnańsk municipality. This municipality includes its borders: the southwestern part of the Suchedniowski Plateau and the northern part of the Massif of the Holy Cross Mountains. As far as the structure is concerned, the Plateau is formed by the Lower Triassic sandstones, the so-called Pstry sandstones. There are also outcrops of limestone and marly dolomite, as well as small and sparse dunes. Referring to the geology of the substrate, the Zagnańsk commune is located within the Mesozoic shield of the Paleozoic core of the Świętokrzyskie Mountains, the structure of which consists of Permian and Triassic formations developed in the form of sandstones and limestones. The southern part of the community includes older formations of the Palaeozoic core of the Holy Cross Mountains, composed of Cambrian, Silurian and Devonian sediments. The Paleozoic is represented by Tsąpiascoides, Middle Cambrian and Upper Cambrian schists building the Krzemionki Mountains; quartzitic sandstones, siltstones and claystones (Barcza Mountain), as well as Middle Devonian dolomites (Góra Chełmowa). The Triassic formations (of the mottled sandstone) occupy the largest part of the municipality forming massifs of hills in the central, southern and western part of Zagnańsk. In these areas there are also thin and thick-bedded grey sandstones which may have a light grey shade; sandstones and siltstones, brittle, sometimes having a greyish-purple shade with mica, interlayered with slates known from the dolomite quarry. We are dealing here with Quaternary sediments such as: sands, clays, loess, silts, and also peats appearing mainly in the vicinity of the rivers, and also cover the whole commune in irregular patches. These are sediments of glacial, water-glacial, fluvial, aeolian, weathered (with fragments of bedrock) and deluvial origin [16, 17].

2.3. Wola Kopcowa

Wola Kopcowa is located in the Masłów Commune within the Paleozoic core of the Świętokrzyskie Mountains. Cambrian quartzite sandstones and quartzite and clay shales are the oldest formations. The Ordovician and Silurian represent sandstones, shales, and greywackes. Devonian is mainly carbonate sediments, dolomite, limestone and marl. Siliceous shales and claystones are the remains of the Carboniferous. The Permian is characterized by the remaining conglomerates and limestones. Quaternary sediments lie on top of the Palaeozoic basement and form a discontinuous cover of various shapes (clays, silts, gravels, sands, loess, peats and muds). As for the minerals occurring in the area, they are mainly sandstones, limestones, clay raw materials, sandstones and conglomerates. As far as economy is concerned, the greatest benefits are derived from Cambrian quartzitic sandstones (used as road aggregate and refractory material), which are mined from Wiśniówka deposit. In terms of obtaining other raw materials in the area of Masłów commune, it is visible that they are not fully exploited because of restrictions e.g. from the Świętokrzyski National Park. Due to the agricultural and tourist character of the commune, the development of the stone mining industry is not planned at the moment [18].

3. TEST METHODOLOGY

The first very important thing to study radon activity in water is proper sampling. Water was taken into 1.5l PET bottles after it had been slowly drained from the tap. For all studied intakes, both in January and February, water was taken after 4 min of slow flow from the tap. The water was poured full (under the stopper) to prevent the formation of air bubbles. Investigations of ^{222}Rn activity in water are performed using specialized equipment AquaKIT (manufactured by Genitron GmbH), which is connected to the AlphaGUARD PQ200PRO ionization chamber shown in Figure 4. To obtain correct results it must be remembered that half-life of radon – ^{222}Rn is only 3.825 days. Therefore, the collected water sample must be sent to the laboratory as soon as possible. Water in the water supply system was taken for measurement of radon activity after 4 min of free flowing. Three samples were taken from three different intakes in Kielce district (Bolechowice, Kołomań, Wola Kopcowa) in 1.5 l PET bottles. The samples were taken from 3 points on the intake of water supply network (initial middle and final).

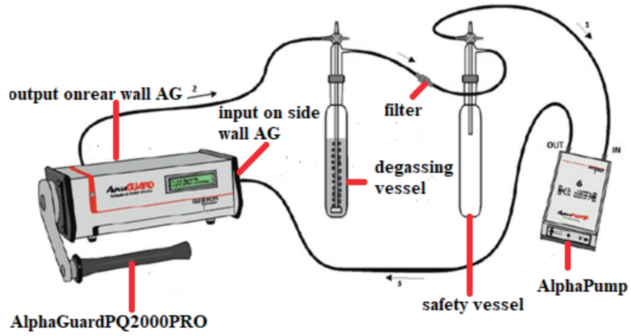

Fig. 4. Kit for measuring ^{222}Rn concentration in water samples [19]

The kit used to measure the concentration of ^{222}Rn in water samples, shown in Figure 1, consist of with a portable radon monitor (AlphaGUARDTM); a gas-tight pump with a step-regulated gas flow from 0.03 to 1.0 $dm^3 \cdot min^{-1}$ (AlphaPUMPTM); and a gas-tight kit for degassing the water sample and, most importantly, for releasing the radon dissolved in it. The above-mentioned components are connected in a hermetically sealed circuit. The measuring station also includes an immersion thermometer (measures the temperature of the water at the time of the measurement being performed); a stopwatch that measures the exact time of the measurement; a „docker" (thanks to it, it is possible to incorporate a syringe with the collected water sample into the system, as well as a syringe that is part of the AquaKITTM set with a volume of 100 cm^3. Thanks to the connection of the AlphaGUARDTM monitor with a computer, all the data from the measurement are sent and collected and then the necessary calculations are made. This monitor is the basic fundamental device found on the test bench, as it performs the actual measurement of ^{222}Rn concentration. The measurement takes place in an ionization chamber equipped with filters that retain radioactive radon decay products, as well as impurities. Radon enters the chamber along with atmospheric air thanks to the AlphaPUMPTM pump. Before entering the chamber, the air is pumped through a glass vessel designed to degas the water sample, followed by an assurance vessel, also called a desiccant, where excess moisture is retained there is a possibility of condensation (this protects the ionization chamber from damage). The water sample under test is placed directly into the degassing vessel by connecting the syringe outlet to the „docker" and gently injecting the water, in such a way as not to cause turbulent flow, which could cause partial escape of radon. Immediately after injecting a 100 cm^3 water sample into the degassing vessel, the valves in this vessel and in the assurance vessel are closed to hermetically seal the measurement system. After transferring the water sample with a volume of 100 cm^3 into the measuring system and its hermetic closure, and turning on the AlphaGUARD™ in a one-minute measurement cycle, in flow mode, the actual measurement of radon concentration in the water sample, actually in the air of the ionization chamber, into which ^{222}Rn has been extracted from the water, takes place. During the first 10 minutes of the measurement, the AlphaPUMP™ works by pumping 0.30 $dm^3 \cdot min^{-1}$ of air in a closed circuit: AlphaPUMP™-AlphaGUARD™-AquaKIT™. After 10 minutes of measurement, the pump is turned off and measurements continue, for another 20 minutes. After 30 minutes from the start of the measurement (turning on the pump), AlphaGUARD™ is turned off, ending the measurement. AlphaEXPERT™ software is used during processing of the obtained data. In this program, it is also possible to preprocess the data, including reading the average values of ^{222}Rn concentration and uncertainty of its determination for each 30-minute measurement. The average ^{222}Rn concentration along with the uncertainty of the determination is given in Bq/L. Conversion of this value to the concentration of ^{222}Rn in a water sample is possible using an equation proposed by the kit manufacturer [20, 21].

The water was additionally tested for pH, conductivity, total hardness, chloride, phosphate, ammonium ion, nitrate, and nitrite. These tests were carried out in the laboratory. The pH was measured with an Elmetron CP-551 pH meter. Conductivity was measured with a Hanna HI 8819 conductivity meter. Total hardness was measured by titration with disodium stannate. Chlorides were measured by the MOHRA method. Phosphate, ammonium ion and nitrite were measured by the calorimetric method. Nitrates were measured by the calorimetric method with phenolydisulfonic acid.

The results made it possible to determine the quality of water in a given locality.

4. RESULTS

Figures 5 and 6 summarize the results of ^{222}Rn activity in water for the Bolechowice, Kołomań, and Wola Kopcowa intakes.

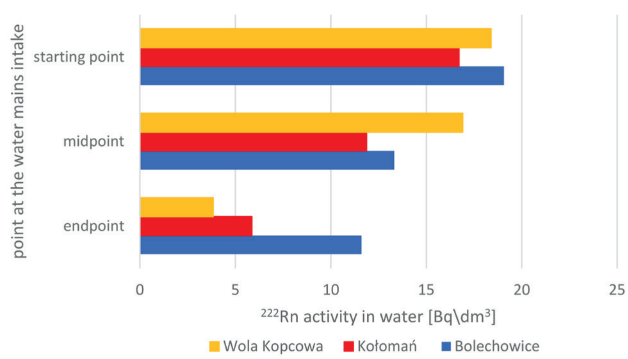

Fig. 5. ^{222}Rn activity in water for selected points at the intake during January

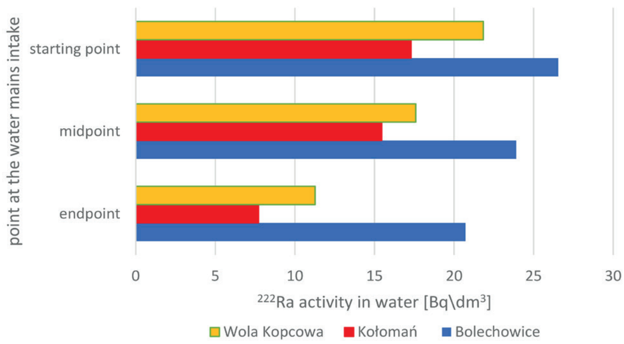

Fig. 6. ^{222}Rn activity in water for selected points at the intake during February

In January, average concentration of ^{222}Rn in water at the Bolechowice intake was 14.66 Bq/dm^3; for the Kołomań intake – 11.48 Bq/dm^3 and for the Wola Kopcowa intake – 13.04 Bq/dm^3. We observed that ^{222}Rn concentration in water is the highest at the beginning of the intake, with values decreasing in further sections. The highest concentration was recorded at Bolechowice intake – 19.06 Bq/dm^3 (beginning of intake). The lowest – 3.84 Bq/dm^3 was recorded in Wola Kopcowa (end of intake).

Table 3. Summary of water quality indicators for the Bolechowice intake in January and February

Index	Endpoint		Midpoint		Starting point	
	01.22.	02.22.	01.22.	02.22.	01.22.	02.22.
pH	6.40	6.41	6.80	6.37	6.40	6.47
total hardness [mg $CaCO_3$/L]	273.26	410.78	405.42	392.92	692.97	276.83
chlorides [mg/dm^3]	35.00	12.00	32.00	12.00	37.00	40.00
conductivity [mS]	0.62	0.34	0.90	0.64	0.64	0.63
phosporates [mg/dm^3]	0.15	0.28	0.11	0.03	0.14	0.05
ammonium ion [mg/dm^3]	0.03	0.30	0.20	0.30	0.00	0.30
nitrites [mg/dm^3]	0.010	0.010	0.010	0.001	0.004	0.010
nitrates [mg/dm^3]	15.23	17.02	4.43	4.13	7.61	6.21

Table 4. Summary of water quality indicators for the Kołomań intake in January and February

Index	Endpoint		Midpoint		Starting point	
	01.22.	02.22.	01.22.	02.22.	01.22.	02.22.
pH	6.40	6.72	6.40	6.60	6.30	6.52
total hardness [mg [$CaCO_3$/L]	367.92	357.20	453.64	285.76	128.59	142.88
chlorides [mg/dm^3]	15.00	12.00	13.00	13.00	16.00	13.00
conductivity [mS]	0.35	0.40	0.34	0.35	0.31	0.36
phosporates [mg/dm^3]	0.09	0.06	0.15	0.23	0.12	0.15
ammonium ion [mg/dm^3]	0.00	0,30	0.00	0,69	0,40	0.50
nitrites [mg/dm^3]	0.003	0.004	0.006	0.001	0.020	0.001
nitrates [mg/dm^3]	6.55	5.92	10.52	10.08	17.18	18.03

Table 5. Summary of water quality indicators for the Wola Kopcowa intake in January and February

Index	Endpoint		Midpoint		Starting point	
	01.22.	02.22.	01.22.	02.22.	01.22.	02.22.
pH	6.50	7.96	7.40	8.00	6.30	7.23
total hardness [mg $CaCO_3$/L]	292.90	267.90	23.22	375.06	528.66	500.08
chlorides [mg/dm^3]	12.00	15.00	12.00	15.00	39.00	41.00
conductivity [mS]	0.62	0.73	0.56	0.64	0.63	0.71
phosporates [mg/dm^3]	0.06	0.05	0.12	0.09	0.16	0.06
ammonium ion [mg/dm^3]	0.10	0.30	0.06	0.50	0.04	0.30
nitrites [mg/dm^3]	0.002	0.010	0.010	0.001	0.200	0.001
nitrates [mg/dm^3]	3.36	4.16	15.05	14.89	13.28	13.48

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In February, mean concentration of ^{222}Rn in water at the Bolechowice intake was 23.73 Bq/dm^3; for the Kołomań intake – 13.49 Bq/dm^3 and for the Wola Kopcowa intake – 16.9 Bq/dm^3. We observe that ^{222}Rn concentration in water is the highest at the beginning of the intake, with values decreasing in further sections. The highest concentration was again recorded at Bolechowice intake – 26.55 Bq/dm^3 (beginning of intake). The lowest concentration of 7.71 Bq/dm^3 was recorded in Kolomani (end of intake).

Tables 3-5 present results of selected quality indicators for water from Bolechowice, Kołomań and Wola Kopcowa intakes. Similarly to the studies on radon activity, water was taken from 3 points in the water supply network. For the Bolechowice intake, the pH value is usually slightly lower than recommended for human consumption, where the lower limit is 6.5. For the Kołomań intake, the range is 6.3 to 6.72. For the Wola Kopcowa intake, the range is 6.5 to 8.0 (the water has the best pH compared with the others). Total hardness was exceeded for the Bolechowice intake in January, at the beginning of the network, but in the next analyzed month a significant decrease can be observed, which is within the permissible ranges of values. The situation is similar for the Wola Kopcowa intake. The other analysed water quality indicators, i.e. chlorides, phosphates, ammonium ions, nitrites and nitrates, occur in small quantities which do not impair the water quality at all. The values of conductivity are within the range for drinking water.

5. CONCLUSIONS

The Bolechowice, Kołomań and Wola Kopcowa intakes under study are located in picturesque areas of the Świętokrzyskie Mountains. Hence, their geological structure is quite rich and the occurrence of various raw materials and minerals is connected with it. Bolechowice is mainly remains of Jurassic and Cretaceous (limestone rocks, carbonate-silicate rocks). Kołomań is Triassic (red sandstones – claystones). And Wola Kopcowa is Jurassic and Cambrian (limestone rocks, conglomerates and sandstones). As far as the geological layers the water is drawn from are concerned, for Bolechowice and Wola Kopcowa it is Middle Devonian, while for the Kołomań intake it is Triassic.

Radon is released from the ground mainly to the air by emanation i.e. release of radon from grains of rocks and minerals; transport i.e. migration of released radon in a given space and exhalation i.e. release of this element from soil and various minerals. The fact that it dissolves well in water may be a reason for radon transport to much further distances from its origin.

Water quality tests carried out for the intakes in January and February showed that the pH of the Bolechowice intake is quite low, but acceptable for drinking water. Total hardness, chlorides, conductivity, phosphates, ammonium ion, nitrites and nitrates are within permissible concentrations for water intended for human consumption. After analyzing all the results it can be concluded that the water is of very good quality.

Preliminary studies of radon activity in water show its presence. This indicates that the minerals present are conducive to the migration of this radioactive element into water.

According to WHO, the highest permissible concentration of ^{222}Rn in water intended for consumption is 100 Bq/dm^3, however in literature one can find more drastic restrictions which allow only 15 Bq/dm^3. It is worth noting that we rarely consume water directly from the tap. Filtered water loses many minerals and radon migrates from it to the air. Water after boiling will be free of radon. The amount of this element in water is trace so it should not have a negative impact on human health and life.

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