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ENVIRONMENTAL AND HUMAN HEALTH RISKS ASSOCIATED WITH THE PRESENCE OF RADIONUCLIDES IN THE GEOTHERMAL WATERS ORIGINATING FROM THE TRIASSIC AQUIFER OF ORADEA (BIHOR COUNTY)

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Abstract - Radium (^{226}Ra) and radon (^{222}Rn) are two of the most common naturally occurring radionuclides found in the groundwater. The analysis of the geothermal waters originating from Oradea Triassic aquifer, showed the presence of both radon and radium in these waters. Oradea geothermal area overlaps the hydrographic basin of Crişul Repede River. The only affluent with a permanent water course is Peța River, which collects the geothermal wastewaters exploited in the area and leads these waters towards Crişul Repede River. The geothermal wastewater discharged into the surface emissaries promotes the migration of pollutants through these rivers. In the present study, the possible impact on the environment and the human health, due to the geothermal exploitation of the thermal waters from Oradea perimeter, was evaluated.

Keywords: geothermal water withdraw, radionuclides contamination, effective dose.

1. INTRODUCTION

Currently, in the energy sector of most countries, there is a reconsideration of the priorities regarding the increasing of consumer safety and environmental protection. In this process, the renewable energy sources such as geothermal energy, offer an affordable solution for medium and long term guarantee. As such, the use of the alternative energy sources becomes relevant for today's world. Renewable energy technologies generate a small amount of pollutant emissions and waste, decreasing significantly the chemical and physical (thermal, radioactive) pollution.

However, the geothermal waters can contain different elements like heavy metals, petroleum hydrocarbons, or radionuclides and the release of these waters into the surrounding environment may raise concern about the aquatic ecology and the health risk for the local residents. Generally, in the geothermal waters exists high concentrations of radionuclides; as a consequence the continuous monitoring of the thermal waters radioactivity is necessary for the evaluation and the prevention of the possible negative health effect caused by the use and the disposal of these waters.

Radium (^{226}Ra) and radon (^{222}Rn) are two of the most common naturally occurring radionuclides found in the groundwater. ^{226}Ra ($T_{½}$ = 1620 y) is produced by the alpha decay of ^{230}Th ($T_{½}$ = 75200 y) in the ^{238}U decay series. Radium may enter the geothermal water by a number of processes including alpha recoil, desorption from aquifer surfaces, dissolution of aquifer solids [1], ion exchange, leaching from radiation-damaged crystals [2]; and the decay of dissolved parent isotopes, the last process is generally negligible because thorium isotopes are nearly insoluble in the ground water [3]. ^{222}Rn ($T_{½}$ = 3.8 days) is produced by the alpha decay of his parent ^{226}Ra. Because of its gaseous state and inert chemical properties, radon is very mobile and it can move either as a gas or be dissolved in geofluids.

Much of the previous research on the hydrogeochemistry and radioactivity of this thermal aquifer was conducted by Vasilescu et al. (1970) [4], Paal (1975) [5], Szabo (1978) [6], Țenu 1981 [7], and Gilău (1997) [8].

2. HYDROGEOCHEMICAL FEATURES OF THE TRIASSIC GEOTHERMAL AQUIFER FROM ORADEA

The geothermal deposit is located almost entirely in the basement of Oradea, covering an area of approximately 75 km^2 [5, 7].

The data from the drills located in the area, correlated with the results of the geological research carried out in the adjacent areas [9] showed that the geological structure in the area consists of formations belonging to the Quaternary, Neogene, Cretaceous, Jurassic and Triassic and the basement consists of metamorphic rocks.

The Triassic collector concerns limestone and dolomite at 2200-3400 m in depth. It belongs to the Inner Dacides, more precisely to Bihor Unit, as an underground extension of the rocks exposed in Pădurea Craiului Mountains, and it is covered by the Cenozoic formations of the Pannonian Basin. The isobaths at the top of Triassic are indicative of depths ranging between 1600 and 3000 m. Their thicknesses are about 500 to 1000 m, gradually thinning westward and northward to less than 200 m [7]. The lateral extension of these rocks is about 113 km^2.

In the area of Oradea, at 2400 m depth, the thermal gradient has an average value of 25-42.5°C/km (Fig. 1).

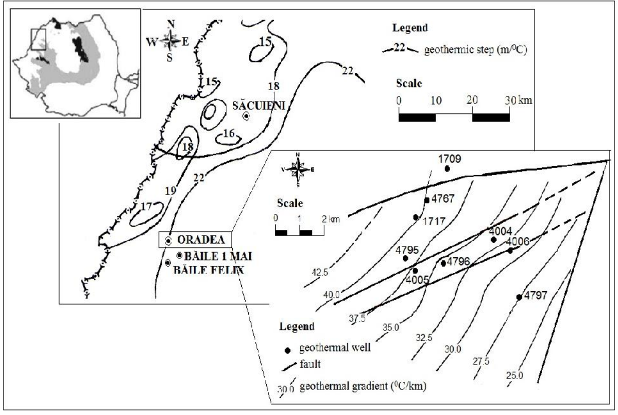


Fig.1. Geothermal features of the Triassic geothermal aquifer of Oradea, with changes after 1973 and Butac Paraschiv 1985.

The geothermal water is exploited through 12 wells: 11 production wells and a single one for reinjection. The wells debits range between 518.4-3456 m^3/day, in artesian regime. The wellhead temperatures are between 70 to 105°C, decreasing from northwest towards southeast [7]. The amount of the total dissolved solids (TDS) is relatively low, ranging from 1031 to 2308 mg/l. The waters originating from this aquifer are classified as SO_4-HCO_3-Ca-Mg type. These waters have low contents of dissolved gases, mainly CO_2 (8-42 mg/l) and H_2S (0.2 mg/l) [8].

This aquifer has a presumed natural recharge; the waters are probably originating from Apuseni Mountains, about 80 km East of Oradea. The main flow streams trend of these waters is from northeast towards west [7]. A secondary flow direction is trended north-northwest towards south-southeast; both these directions seem to connect [7]. The geothermal reservoir is bounded by faults. The tectonic pattern is mostly due to the Latest Cretaceous/Paleogene tectogenesis and the Neogene subsidence. Therefore, there are internal faults into the reservoir dividing it into several distinct blocks, causing discontinuities in the water circulation.

3. MATERIALS AND METHODS

3.1. The analysis of ^{222}Rn and ^{226}Ra from water

The geothermal waters were sampled from the wellhead, in 500 ml polyethylene bottles, which were previously washed three times with ultra pure water (Millipore Milli-Q). Before sampling, they were rinsed two times on the site, with geothermal water. They were fully filled up and they were screw up in order to prevent the possible radon losses.

Radon was analyzed within a maximum of 12 h from the sampling time. For radium measurement, after the arrival in the laboratory, the water samples were transferred into the glass bottles completely filled and hermetically sealed. Radium analysis was made by measuring the generated radon, after a period of about one month from the time of sampling, when radium-radon equilibrium was established at a rate of over 98% [10]. Radon loss by diffusion through the container walls was prevented by keeping the samples in glass bottles.

Radon and radium measurement in water samples was made by using a system equipped with Lucas cells and a glass container which was used for the extraction of the dissolved gas [11]. In the glass container (V=500 ml), a known volume (300 ml) of water sample was introduced. After one minute of strong shaking, the equilibrium between the activities of radon from the water (A_w) and radon from the air (A_a) above the liquid (V_a=air volume above the liquid) was established. The solubility constant was calculated as it follows [11]:

$$\alpha = \frac{A_W}{A_a} \cdot \frac{V_a}{V} \quad (1)$$

The glass container was coupled to a Lucas cell (previously held under vacuum) by a stopcock and to a Janet syringe containing 150 ml of distilled water. By opening the stopcock, the air containing radon was transferred from the scrubber into the Lucas cell (145 ml). In order to prevent radon progeny from entering the cell, a filter was inserted between the glass container and the Lucas cell. The measurement of the activity in the Lucas cell was made by using the photomultiplier inserted in the LUK-3A instrument. For the normal temperature of 20°C, the solubility coefficient (α) of radon in water is 0.254. If the working temperature is other than 20°C, the solubility coefficient (α) is calculated by using the following formula:

$$\alpha = 0.1057+0.405 \cdot \exp(-0.0502 \cdot t) \quad (2)$$

where "t" is the water temperature in the scrubber.

For a temperature of 20°C between the activity of radon in the water A [Bq/l] and the number of counts/second N [c/s] recorded by the device, the following relation is obtained [12]:

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

Considering the fact that radon was analyzed within a maximum of 12 h from the sampling time, the following correction had to be done:

$$A_{(0)} = A \cdot \exp(\lambda \cdot \Delta t) \quad (4)$$

where: "$A_{(0)}$" represents the initial activity (at the sampling time), "A" is the measured activity (within a maximum of 12 h), "Δt" is the time period between the sampling moment and the time of the measurement, and "λ" (λ = ln2/T) is the radioactive constant of radon (T = 3.82 days, the half-life for radon).

For radon analysis, the detection limit (LD) of the method was 0.2 Bq/l, for the measured time of 300 sec, while for radium, the detection limit was 0.05 Bq/l, for a measuring time of 3000 sec [12]. The statistical errors associated with radon analysis were between 5 and 6%, while for radium were in the range of 5 and 15% [12].

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

3.2. The analysis of ^{222}Rn and ^{226}Ra from air

The radon level from the indoor air was analyzed by using CR-39 track-etched detectors, according to the NRPB (National Radiological Protection Board) protocol and US-EPA recommendations. The used method is detailed in the study made by Dinu et al. 2009 [13]. The detectors were placed, in the inhabited areas, such as bedrooms and living-rooms, at a height of 1-1.5 m from the floor. The detectors were exposed since 22 of August 2009 until 20 of December 2009, for almost four months.

Alpha track detectors are inexpensive, reliable and easy to use. Every CR-39 detector is placed under the cap of a cylindrical polypropylene container 55 mm high and 35 mm diameter with a small gap in its upper part which prevents radon decay products and also 220Rn from entering. Then, only the alpha particles from radon that has diffused into the container and from the polonium produced inside can strike the detector. After the exposure time an etching process is done. Radon concentration was determined by counting the tracks in a given area. In order to calculate the radon activity, the following equation was used [13]:

$$C_{Rn} = \frac{\rho \cdot F_c}{t} \quad (5)$$

where: “C_{Rn}” is the radon concentration (Bq/m^3), “ρ” is the tracks density (traacks/mm^2), “F_c“ is the calibration factor (F_c= 42.696 (kBq hours/m^3)/(tracks/mm^2)), and “t” is the exposure time (hours).

The individual error of radon measurements was estimated at less than 10% [13].

4. RESULTS

The content of radon and radium in the sampled geothermal waters is shown in Fig.2 and Fig.3.

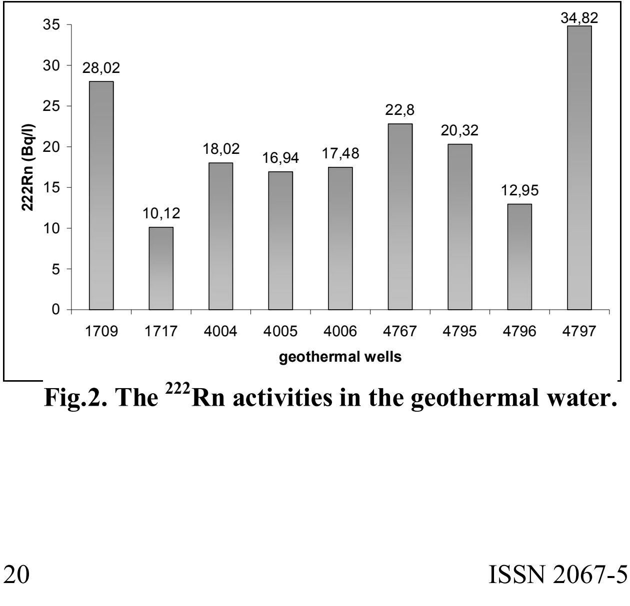


Fig.2. The ^{222}Rn activities in the geothermal water.

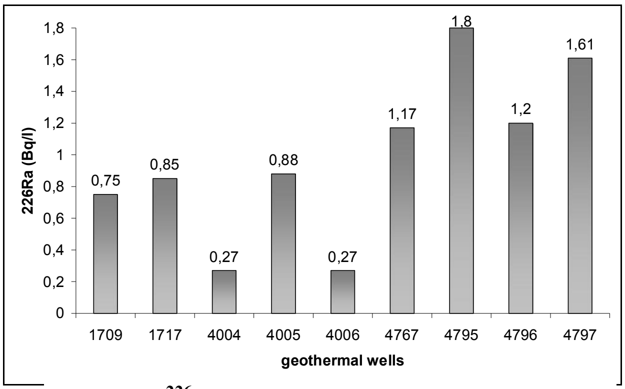


Fig.3. The ^{226}Ra activities in the geothermal water.

Radon had a wide range of activity, between 10.12 and 34.82 Bq/l, having the highest values in the wells 4797 (34.82 Bq/l) and 1709 (28.02 Bq/l). Radium activities ranged between 0.20 and 1.82 Bq/l. The highest radium levels were registered in the wells 4795 (1.8 Bq/l) and 4797 (1.61 Bq/l).

The ^{226}Ra and ^{222}Rn levels registered in the geothermal waters from Oradea are similar to those reported in the case of other geothermal aquifers like Wairakei-New Zealand [14], Bizovac Spa-Croatia [15], or some thermal aquifers from Hungary [16]. There are geothermal aquifers where ^{222}Rn activity can reach higher levels, up to 46.9 Bq/l (Bakreswar-India), 93.79 Bq/l (Stubica Spa-Croatia) [15], or even 1868 Bq/l (some thermal springs used as spas in Spain). Some thermal waters can have ^{226}Ra level up to 2 Bq/l (Budapest-Hungary), or 3.66 Bq/l (some thermal springs used as spas in Spain), or even 29.03 Bq/l (St.Mary-Louisiana from the Gulf Coast-USA).

The presence of radionuclides is a consequence of the contact between the waters and the rocks containing radioactive elements, the tectonic and geothermal features of the area, and hydrochemical and hydrodynamic features of aquifers.

5. ENVIRONMENTAL AND HUMAN HEALTH IMPACT ASSESSMENT

In Oradea, the geothermal waters are either used for the indoor heating or for the domestic hot water preparation. This was the reason for which we have analised the possible air and the domestic water contamination with radionulides.

The possible surface water contamination due to geothermal water withdraw was investigated, considering that the geothermal wastewaters are discharged in Peţa River.

5.1. Domestic hot water contamination

The geothermal water from the heat exchangers is used as a heating agent for the domestic hot water preparation. In order to evaluate the possible domestic water contamination in the heat exchangers, the domestic water samples were taken at the entrance and at the exit of heat exchangers. The results are shown in Table 1 and 2.

As shown in the up mentioned tables, the radionuclides content of the domestic waters did not suffered significant changes when passing through the heat exchangers.

In the geothermal waters, radon level ranged between 10.08 and 34.82 Bq/l, while in the domestic water the activity of radon was between 7.52 and 16.73 Bq/l.

Table 1. Domestic water contamination with radon in the heat exchangers.

Heat exchanger	^{222}Rn (Bq/l)			
	Geothermal water		Domestic water	
	heat exchanger entrance	heat exchanger exit	heat exchanger entrance	heat exchanger exit
4797	34.82	29.7	15.08	13.92
4767	22.8	20.62	14.84	16.73
4005	16.94	13.71	16.2	9.77
4796	12.95	10.08	12.48	7.52

Table 2. Domestic water contamination with radium in the heat exchangers.

Heat exchanger	^{226}Ra (Bq/l)			
	Geothermal water		Domestic water	
	heat exchanger entrance	heat exchanger exit	heat exchanger entrance	heat exchanger exit
4797	1.77	1.63	0.24	0.23
4767	1.73	1.5	0.17	0.15
4005	0.88	1.1	0.11	0.29
4796	1.22	0.82	0.2	0.12

Radon level in both geothermal and domestic heat water was considerably lower than 100 Bq/l, which is the reference level for drinking water mentioned by the UE-EPA legislation [17], respectively 1000 Bq/l, which is the action level for drinking water [17]. In all the analyzed water samples (geothermal and domestic water) the content of radon was below 74 Bq/l (2000 pCi/l); the waters which exceed this value are considered to be radioactive [18].

Radium level in the domestic water ranged between 0.11 and 0.29 Bq/l, having values under 0.5 Bq/l, which represents the maximum level recommended by the EU legislation [17].

5.2. Indoor air contamination

In Oradea there are several residential houses or public institutions heated with geothermal waters. Considering this, we have investigated the possible indoor air contamination with radionuclides in several (14) residential houses heated with the aid of the geothermal water. We have also analyzed radon level inside the doublet building Nufărul.

According to the data presented in Fig.4, the air from the houses heated with geothermal waters has low level of radon. In all the investigated houses the radon level was under the maximum limit recommended by WHO, which is 100 Bq/m^3, or 200 Bq/m^3 the level recommended by ICRP (International Commission on Radiological Protection). Inside the doublet building Nufărul, radon level was higher (111.95 Bq/m^3) than in the case of the residential houses. However, this value is below the maximum permissible limit for exposure to radon at work, which is 163 Bq/m^3 [19].

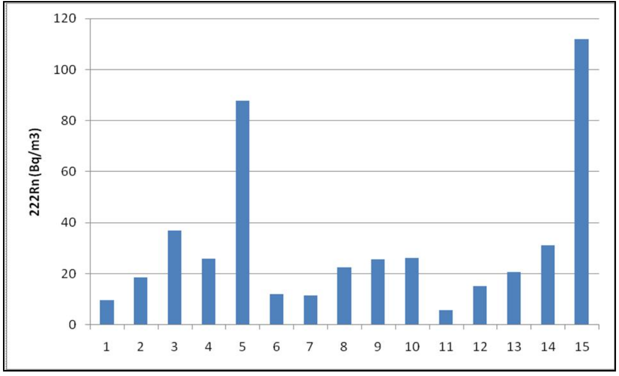


Fig.4. The radon level in indoor air.

These results show a low magnitude of radon exposure for the people living in these houses. The average values of radon activity recorded in the homes of Oradea (30.78 Bq/m^3) is considerably lower than the average level reported for Transylvania, which is 82.5 Bq/m^3 [15].

In the present study, we calculated the effective dose due to the exposure of the people to the indoor radon in the houses heated with the aid of geothermal water. The effective dose was estimated based on the relation proposed by UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation) [20]:

$$\text{Dose} = C_a\cdot(\varepsilon_r+\varepsilon_d F)\cdot O \quad (6)$$

where: "C_a" is annual averages of radon activity (Bq/m^3), "ε_r" and "ε_d" are conversion factors for the dose from radon and its decay products resulting short-lived ($\varepsilon_r = 0.17$ nSv·h^{-1}/Bq·m^{-3} and $\varepsilon_d = 9$ nSv·h^{-1}/Bq·m^{-3}), "F" is the equilibrium factor between radon and its short-lived progeny (F = 0.5), "O" is occupational factor for European countries (A = 0.7·8.76·103 h).

The results showed that the population of Oradea is exposed to a mean effective dose of 0.118 mSv/year. The doses are much lower than the value of 3 to 10 mSv/year, which is the action level recommended by the ICRP.

In some studies lower radon levels are reported inside the houses located in the geothermal perimeters, than those registered in Oradea. Thus, in Rotorua (New Zealand) there are reported activities between 10-80 Bq/m^3 radon, with an average of 18 Bq/m^3. There are also many cases, where radon from the indoor air is considerably higher than the one from Oradea. For example, in several cities near Rome (Italy) concentrations up to 282 Bq/m^3 were recorded. Similar results were obtained in Dikhili (Turkey) where radon level ranged between 31-280 Bq/m^3. In Finland radon from the indoor air ranged between 95 and 1200 Bq/m^3, with an average of 370 Bq/m^3.

5.3. Surface water contamination

In order to evaluate the surface water contamination due to the geothermal wastewaters withdraw, water samples from the discharging point, upstream (100 m) and downstream (100 m) from the discharging pace were analyzed. The sampling points are shown in Fig.5.

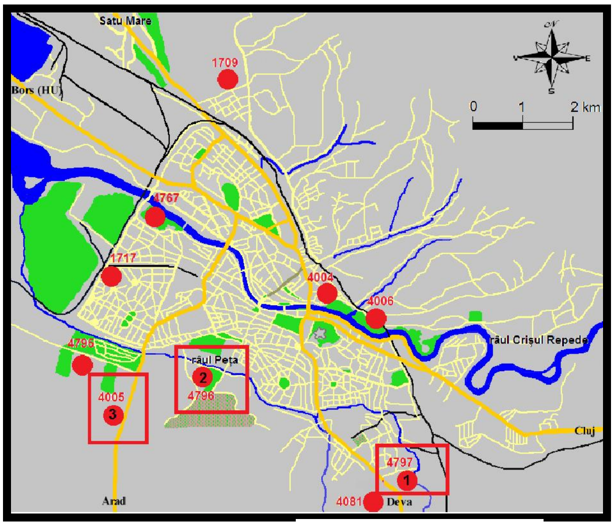
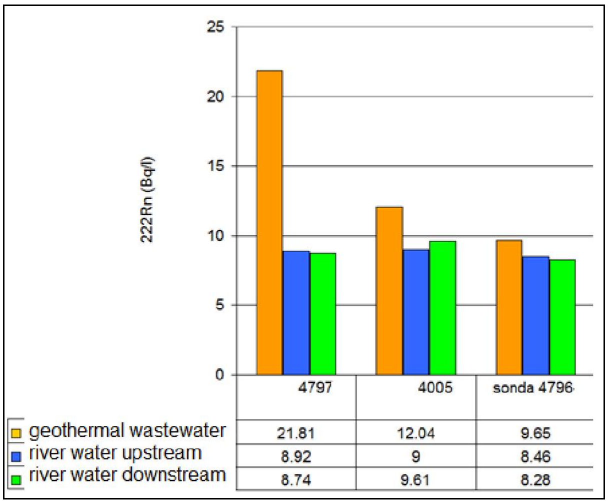


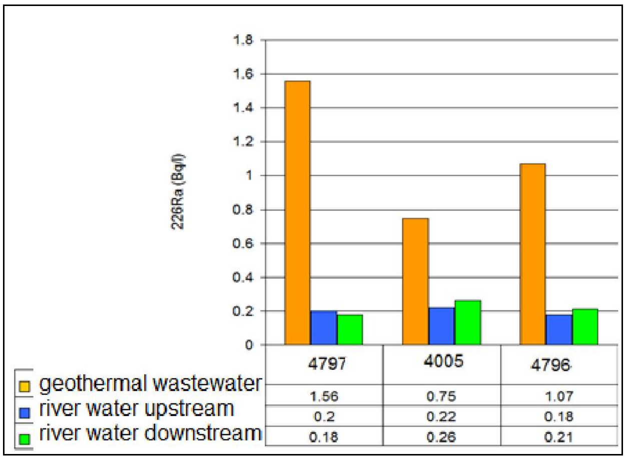
Fig.5. Locations where the geothermal wastewaters are discharged into the surface water (Peța River).

The results are shown in Fig. 6 and Fig.7. The analysis did not show significant changes in radionuclides levels, before and after the geothermal wastewaters discharge. Therefore, the presence of radionuclides in the geothermal wastewaters, which are discharged into the surface water (Peța River), do not present risk for the environment.



	4797	4005	sonda 4796
🟧 geothermal wastewater	21.81	12.04	9.65
🟦 river water upstream	8.92	9	8.46
🟩 river water downstream	8.74	9.61	8.28

Fig.6. The variation of radon level in Peța River, before and after the geothermal wastewater discharge.



	4797	4005	4796
🟧 geothermal wastewater	1.56	0.75	1.07
🟦 river water upstream	0.2	0.22	0.18
🟩 river water downstream	0.18	0.26	0.21

Fig.7. The variation of radium level in Peța River, before and after the geothermal wastewater discharge.

5. CONCLUSIONS

1. Radon had a wide range of activity, between 10.12 and 34.82 Bq/l, having the highest values in the wells 4797 (34.82 Bq/l) and 1709 (28.02 Bq/l). Radium activities ranged between 0.20 and 1.82 Bq/l. The highest radium levels were registered in the wells 4795 (1.8 Bq/l) and 4797 (1.61 Bq/l).

2. The results proved that there is no contamination of domestic hot water due to the presence of radionuclides in the geothermal waters.

3. According to the present study, the air from the houses heated with geothermal waters had low radon level. In all the investigated houses the radon level was under the maximum limit recommended by WHO or by ICRP.

4. The results proved that there is no contamination of the surface waters due to the geothermal wastewaters withdraw.

5. This study indicates the presence of some potential risk factors for the environment and human health due to the presence of radionuclides in the geothermal waters from Oradea Triassic aquifer.

6.Although the geothermal energy is classified as a clean energy, the management and exploitation of the geothermal resources should be done with most care to avoid negative effects on the environment and human health that could result from chemical or physical contamination.

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